Instabilities of drops detaching from a circular capillary tip

[1,2]Kairaliyeva T.*, [1]Karbaschi M.,
[1,3]Taeibi-Rahni M., [1]Faraji S., [4]Schano K.H.,
[1]Born M., [2]Aidarova S.B., [1]Miller R.

[1]Max Planck Institute of Colloids and Interfaces, 14424 Potsdam/Golm, Germany
[2]K.I. Satpayev Kazakh National Technical Research University, Almaty, Kazakhstan
[3]Sharif University of Technology, Teheran, Iran
[4]Gamma-Service, Berlin, Germany

*E-mail: *talmira.kairaliyeva@mpikg.mpg.de*

When drops are formed at circular capillaries at high liquid flow rates, the relation between surface tension and drop volume is often not linear but shows irregularities. These volume bifurcations and other more complex pattern of detaching drops for pure liquids depend on the bulk viscosity and the surface tension. In this study, it has been shown that liquids with high surface tensions and low viscosities, such as pure water, show complex pattern of drop times. When the surface tensions are much lower, such as for pure ethanol, and the bulk viscosity is high enough, such as for water:glycerol mixtures with a glycerol content of 75% and more, the bifurcations decrease or disappear.

Keywords: Single drop dynamics; drop volume tensiometry; hydrodynamic instabilities; drop size bifurcations; effects of viscosity; effect of surface tension.

Дөңгелек тектес капиллярлардан шығатын тамшылардың тұрақсыздығы

[1,2]Кайралиева Т.*, [1]Карабаши М.,
[1,3] Таейби-Рахни М., [1]Фараджи С.,
[4]Шано К.К., [1]Борн М.,
[2]Айдарова С.Б., [1]Миллер Р.

[1]Коллоид және фазааралық беттер Макс Планк Институті, Потсдам/Гольм, Германия
[2]Қ.И. Сатпаев атындағы Қазақ ұлттық техникалық зерттеу университеті, Алматы, Қазақстан
[3]Шариф Технология университеті, Тегеран, Иран
[4]Gamma-Service, Берлин, Германия

*E-mail: *talmira.kairaliyeva@mpikg.mpg.de*

Дөңгелек тектес капиллярларда және сұйықтықтың ағу жылдамдығы жоғары болған жағдайда пайда болған тамшының беттік керілуі мен тамшы көлемінің арақатынасы көп жағдайда сызықты бола бермейді. Бұл үдеріс тамшы бөлінген кезіндегі сұйықтықтың көлемдік тұтқырлығы мен беттік керілуіне байланысты. Беттік керілуі жоғары сұйықтықтарда, мысалы суда, тамшының өмір сүру уақыты күрделі таралады. Ал беттік керілуі төмен сұйықтықтарда мысалы этанолда, көлемдік тұтқырлығы жоғары су-глицерин қоспасында (75% глицерин) бұндай ауытқулар мүлдем байқалмауы мүмкін.

Түйін сөздер: жеке тамшы динамикасы; тамшының көлемі бойынша тензиометрия; гидродинамикалық тұрақсыздық; тамшы көлемінің бифуркациясы; тұтқырлық әсері; беттік керілудің әсері.

Неустойчивость капель, вытекающих из капилляров круглого сечения

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[4]Шано К.К., [1]Борн М.,
[2]Айдарова С.Б., [1] Миллер Р.

[1]Институт коллоидов и поверхностей Макс Планка, Потсдам/Гольм, Германия
[2]Казахский национальный исследовательский технический университет им. К.И. Сатпаева, Алматы, Казахстан
[3]Технологический университет им. Шарифа, Тегеран, Иран
[4]Gamma-Service, Берлин, Германия

*E-mail: *talmira.kairaliyeva@mpikg.mpg.de*

При образовании капель на капиллярах круглого сечения и высоких скоростях потока жидкости соотношение между поверхностным натяжением и объемом капли часто не является линейным, а проявляет особенности. Такие бифуркации объема или иная, более сложная форма отделяющихся капель, в случае чистых жидкостей зависят от объемной вязкости и поверхностного натяжения. В данной работе показано, что для жидкостей с высоким поверхностным натяжением, например, для чистой воды, характерно сложное распределение времени жизни капли. В случаях, когда поверхностное натяжение намного меньше, чем у воды, например, для этанола, и объемная вязкость достаточно высока, как, например, в смесях вода:глицерин с содержанием глицерина не менее 75%, такие бифуркации менее существенны или вообще отсутствуют.

Ключевые слова: динамика индивидуальной капли; тензиометрия по методу объема капли; гидродинамические неустойчивости; бифуркации объема капли; влияние вязкости; влияние поверхностного натяжения.

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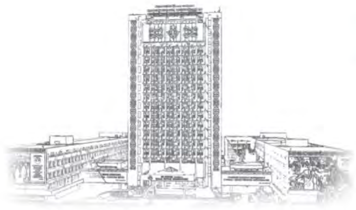

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[1]Max Planck Institute of Colloids and Interfaces, 14424 Potsdam/Golm, Germany
[2]K.I. Satpayev Kazakh National Technical Research University, Almaty, Kazakhstan
[3]Sharif University of Technology, Teheran, Iran
[4]Gamma-Service, Berlin, Germany

*E-mail: *talmira.kairaliyeva@mpikg.mpg.de*

1. Introduction

Dynamics of drops and bubbles is an old and at the same time modern topic in fluid dynamics. Recently, the review article summarising many aspects of the coupling of dynamic interfacial aspects and fluid dynamics phenomena was published [1]. The combination of experiments based on single bubbles and drops combined with simulations of various types turns out to be an efficient approach to proceed from more empirical to quantitatively understood phenomena [2].

The formation of tailored single drops and bubbles is required in various technical applications, such as in the ink jet printing [3] where drops of a definite size have to be produced in a short time and the deposited on a certain material. Also in 3D printing, drops of a molten material are deposited on a definite place in the 3D space having a well-defined volume [4]. When microfluidics is used for producing multiple emulsions [5] and drop targeting in medical diagnostics [6], drops of an exact volume have to be produced.

For more than 150 years, the drop volume tensiometry is known as a method to measure the surface tension of liquids. It was possibly first used in 1864 by the pharmacist Tate [12], who took an exact number of drops to produce a definite volume of a liquid medicine. Tate postulated that the weight W or volume V of a drop detaching from a capillary with radius r_{cap} is given by the liquid's surface tension γ

$$W = 2\pi r_{cap}\gamma \quad \text{or} \quad V\Delta\rho g = 2\pi r_{cap}\gamma \tag{1}$$

where $\Delta\rho$ and g are the density difference and gravitational constant. Later, Lohnstein [7,8] showed that Eq. (1) is only a rough estimation, and a correction factor has to be introduced to apply it for an accurate measurement of surface tension. More problems arise from a discussion of the dynamics of drop formation on the basis of a fluid dynamics point of view [1]. It becomes clear that the volume of a detaching drop of a pure liquid is different for different growth rates of the drop [9], in the way as the profile of a quickly growing drop deviates from that of a drop at rest [10]. It was even shown that the volume of drops formed under dynamic conditions do not obey a simple linear relation with the surface tension, as given by Eq. (1) or better by the corrected form

$$V = \frac{2\pi r_{cap} f}{\Delta\rho g}\gamma \tag{2}$$

with the correction factor (f) introduced by Lohnstein [7] and modified in [9] or later for viscous liquid in [11]. It was experimentally observed that the volume of drops formed at rather high liquid flow rates show bifurcations [12], i.e. there is no exact volume for a given liquid at certain drop formation times.

The goal of this work was to extend the experimental basis towards the instability of drops detaching from circular capillaries. As it was earlier shown, at certain liquid dosing rates, the volumes of detaching drops show bifurcations with pattern of various complexity. The presented data shows the drop detachment stability for liquids with different viscosities (water/glycerol mixtures) and different surface tension values (water/ethanol mixtures). Increasing bulk viscosity and decreasing surface tension improves the stability of drop detachments, i.e. lead to a damping of bifurcations in the detaching drop volume. Our experimental results are in match with the theoretical description of this phenomena considering Rayleigh-Plateau instability as the cause of droplet detachment from the capillary tip. Lowering the surface force or increase of the viscosity results in a decrease in Rayleigh-Plateau instability.

The impact of surfactants providing additional dynamic surface effects was not studied in this work.

2. Experiment

The used glycerol and ethanol have been purchased from Sigma-Aldrich. The viscosities of the studied water/glycerol mixtures were 3.7 and 36 mPa·s, for the 40% and 75% glycerol content, respectively. The surface tensions of water, ethanol and glycerol at room temperature were 72.3, 22.1 and 62.9 mN/m, respectively. All mixtures were prepared with MilliQ water.

The details of the setup used for the determination of drop times were described recently in [1]. In brief, liquid from a reservoir flows smoothly (driven by the hydrostatic pressure) through a valve and forms drops at the end of a tube with a cylindrical capillary. A detector is provided at the down side of the capillary to detect the detachment of drops, as shown in Figure 1. The detector linked via an interface to a PC accurately registers the time intervals between subsequent detachments. With each detached drop, the liquid height in the glass column (reservoir) decreases slightly, and by this the drop formation rate decreases, i.e. continuously increasing the drop formation time. Depending on the column diameter and the capillary tip size, we can arrange different changes of the liquid flow rate while the starting flow rate can be set by the valve. In the experiments presented here, we used a reservoir with a cross section diameter of 94 mm and a cylindrical steel capillary with an outer tip diameter of 2.98 mm.

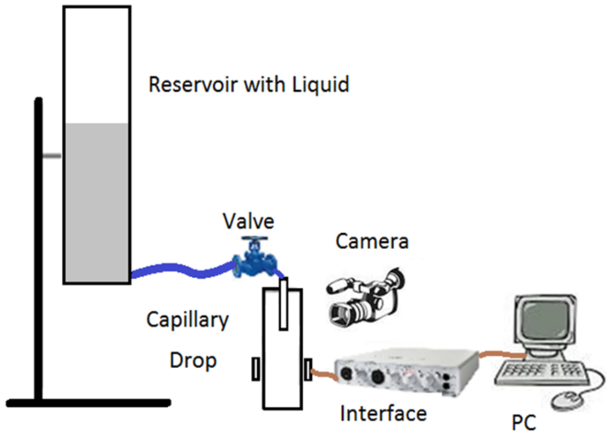

Figure 1 – Scheme of the single drop analyzer based on the hydrostatic pressure of a liquid in a circular column; as proposed in [12]

The accuracy of drop times measurement, i.e. time intervals between two subsequent detachment measured by an opto-coupler and a special electronic interface, is on the order of ±0.2 ms, with the drop formation times in the order of 0.2 to 0.3 s for the given experimental conditions.

3. Results and Discussion

The results of the present investigations have been obtained in terms of drop time as a function of drop number. Due to the dosing principle via the hydrostatic pressure of the liquid in a circular container, the changes in the dosing rate are given by the changes in the level of liquid in the reservoir. Using a valve, it is possible to set the liquid flow such that the initial drop time is of the order of about 0.2 s. The drop formation time slowly increases over time due to a continuous decrease in hydrostatic pressure assuming no change in position of the valve or connections (constant flow resistance). In Figure 2, the change in drop time over a large number of drops (about 28,000 drops) is shown.

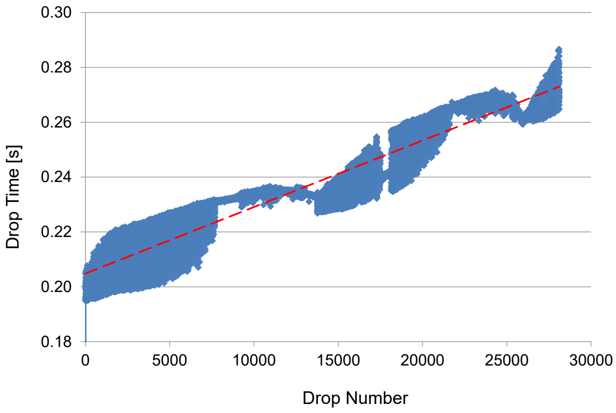

Figure 2 – Dependence of drop formation time on drop number for pure water

What we would expect from the known relation between drop volume and surface tension given by Eq. (2) is shown by the dashed line – a simple linear correlation between drop time and drop number. However, the data show irregular pattern, i.e. a range of drop times probable for subsequent drops. The drop time values, however, do not scatter in a certain time range but show clear pattern, as it was already discussed in [12]. This is clearly seen in Figure 3, where we zoom in into a particular range of data points, where drop number varies between 510 and 525. In this range of data points, we can see a repeated pattern consists of three subsequent drops. From drop number 525 on, another pattern starts to establish. Note, the jumps in drop time are of the order of about 10%, i.e. 100 times larger than our measurement accuracy for the determination of drop time. Note, to characterize drop formation time in classical studies such as drop volume/weight tensiometry, the drop formation times are typically larger than 5 s. Under these conditions, only a clear straightforward dependence of drop formation time is obtained [9].

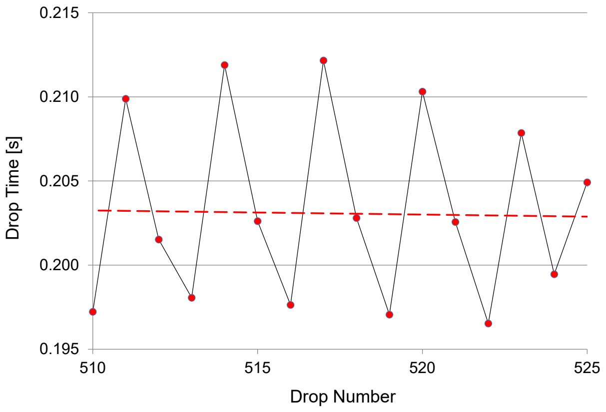

Figure 3 – Dependence of drop formation time on the drop number, a zoomed-in part of data points taken from Figure 2

In Figure 4 we show the behaviour of the system for a mixture of water (85% v/v) and ethanol (15% v/v). The viscosity of this mixture is similar to that of water (1 cP), while the surface tension is remarkably reduced to 42.2 mN/m. The measured drop formation times again show individual specific pattern changing over time. The shift between subsequent drop formation patterns are around 15% of the total time of measurement. The results show that a reduction in surface tension does not remove the effect of data scattering.

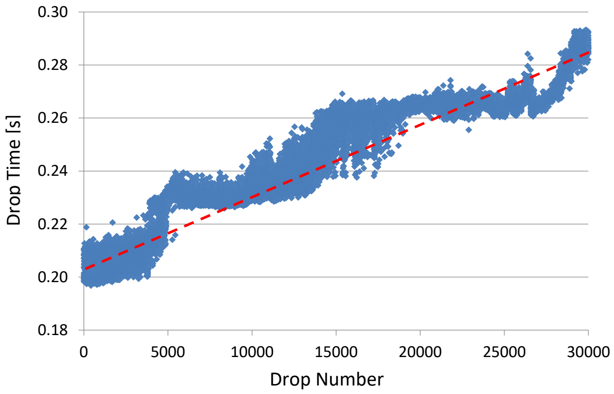

Figure 4 – Dependence of drop formation time on the number of drops for a water/ethanol (85/15) mixture

Pure ethanol has approximately the same viscosity as water, but its surface tension is only 22.1 mN/m (Figure 5). What we see are three branches, each with a certain slope. A detailed analysis shows that the drop formation time recording system is failed in accurately measuring the formation time of each drop, and it measures only the time for a drop randomly chosen out of each two or three drops formed in series. This is mainly due to a decrease in drop detachment volume to a range not detectable by our recorder. As surface tension was reduced by a factor of about 3.2, using the same capillary, the cross-section area of the falling drop is significantly reduced and, therefore, an accurate adjustment of the sensor is required to register each falling drop. Considering the data points of the lower branch of data, the most likely accurately measured ones, the measurement represents a rather linear dependence of drop formation time vs drop number. This linear trend for pure ethanol system is in match with what can be analytically described by Eq. (2).

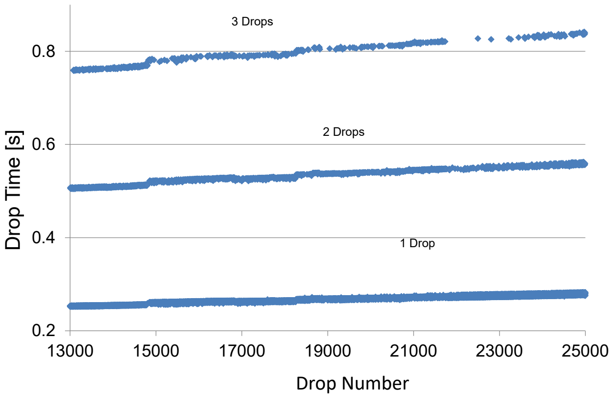

Figure 5 – Dependence of drop formation time on the number of drops for pure ethanol

By using mixtures of water and glycerol, it is possible to check the impact of the liquid's viscosity on the drop formation time under fast drop formation regimes. The results of similar measurements for water/glycerol mixtures are shown in Figures 6 and 7 considering 3:2 and 3:1 water/glycerol mixing ratio respectively. For 40% glycerol solution, the viscosity increases from 1.0 cP for pure water to 3.7 cP while the surface tension is close to the surface tension of pure water (69 mN/m for glycerol solution vs. 71 mN/m for pure water). For the mixture shown in Figure 7, the viscosity is 36 cP and the surface tension is 65.5 mN/m. In both cases, we do not observe patters in terms of bifurcations as it was the case for water and water/ethanol mixtures. However, for lower glycerol contents, not a clear straight dependence between drop formation time and drop number is obtained, as it would have been expected (red dashed line). For the mixture with higher glycerol contents (75% glycerol) the measured data are much closer to the expected

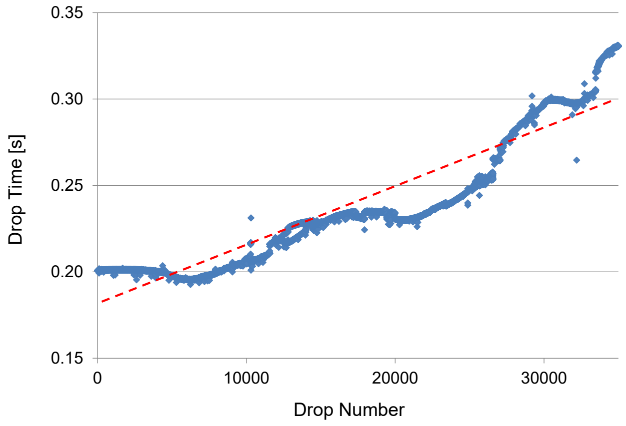

Figure 6 – Dependence of drop formation time on the number of drops for a 3:2 water/glycerol mixture (40 % glycerol with a viscosity of 3.7 cP)

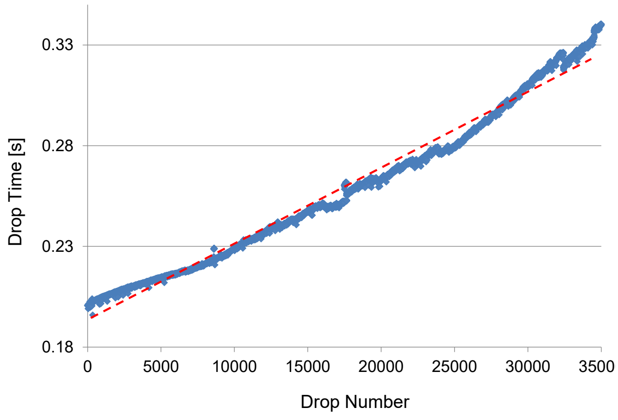

Figure 7 – Dependence of drop formation time on the number of drops for 3:1 water/glycerol mixture (75% glycerol with a viscosity of 36 cP)

linear dependence given by the red dashed line. The reason for this behaviour is yet unclear and would require a detailed visual inspection of the drop formation and detachment process.

4. Conclusion

The investigations on the stability of drop formation of liquids with different surface tension and viscosity show that under certain conditions, bifurcations in the drop formation time take place. These bifurcations originate from drop instabilities happening during the detachment process. Thus, subsequent larger and smaller drops are formed over a certain narrow interval of dosing rate with differences of 10% and even more. For higher surface tensions and low bulk viscosities, as it is the case for pure water, such bifurcations are magnificent over a broad range of dosing rates. The drop formation time oscillates between two values with a peak-to-peak "amplitude" of about 10%. At lower viscosities, and for lower surface tension values, at least for drop formation times of 200 to 300 ms, these amplitudes are even increased. For pure ethanol, however, no bifurcations are observed, and a simple linear dependence of drop time on drop number (smooth change in the liquid dosing rate) is obtained. For more viscous liquids, the Rayleigh-Plateau instability responsible for the observed drop formation time (or drop volume) bifurcations gets damped, and the effect of that on making specific patterns of data points disappears.

For a quantitative analysis of the observed bifurcations and other instabilities, a fast video technique would be required to instantly record the onsets of drops detachment from the capillary tip.

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