

Lateral wetting angle of falling film in dense fluid

Y. Sutjiadi-Sia, R. Eggers*

Hamburg University of Technology, Eissendorferstrasse 38, 21073 Hamburg, Germany

Received 7 June 2007

Available online 20 December 2007

Abstract

An investigation of the wetting ability of a liquid-falling film on vertical steel and glass surfaces is performed by measuring the thickness and the width of the water falling film at 313 K up to 27 MPa. An attempt to reconstruct the two-dimensional cross-section of the falling film was made. The cross-section of the falling film is assumed to have the shape of a circular segment.

The falling film wetting angle is compared with the sessile drop contact angle. The sessile drop contact angle represents the upper limit of the film wetting angle. A continuous increase in the mass flow at a constant pressure causes the spreading of the film. This happens when the force balance between the interfacial tensions and the dynamic forces, which deform the film geometry, is exceeded. However, if the pressure increases, the wettability goes down. This is partly due to the accumulation of liquid mass, which is caused by a larger buoyancy.

The critical mass flow, that is, the minimum mass flow needed to guarantee a wide covering film is reported. The disintegration point of a liquid film is directly affected by its wettability.

© 2007 Published by Elsevier Ltd.

Keywords: Falling film; Wettability; Contact angle; Wetting angle; Film instability; Film disintegration

1. Introduction

In process engineering, numerous processes make use of a wide falling film. This is a liquid (e.g. water) which flows under the influence of gravity down a flat vertical support material such as glass or stainless steel. Regarding the heat and mass transfer, it is important to have adequate knowledge about the exchange geometry in the process. In the past, it has been assumed that the whole area of the support material provided can be entirely covered by the liquid film, and thus, the exchange area is equal to the whole available area. In the literature [1–3], the film thickness is often estimated by means of the Nusselt film condensation theory [4] which assumes a shear-stress-free-flow. Such a simplification does not take the wettability of the system and the surface shear stress into account.

Speaking about the wettability of a liquid on a solid material, it nearly always concerns only the static sessile drop [5,6]. But in fact, in most industrial processes, liquid – driven by the gravitational force – flows along a solid material. It is therefore important to know the wetting ability of a flowing liquid on a vertical surface in a dense fluid environment, and how that compares a static contact angle. Kern [7] and Towell [8] reported on the optical, direct method of measuring the falling film wetting angle, but Kern's main interest was the hydrodynamics. There was no report on the magnitude of the wettability and of a universal parameter which can be used as a measure for the wetting. Furthermore, the measurement method is best applied at atmospheric pressure, not at high-pressure conditions.

The focus of the present paper is the study and the characterization of the wettability of falling liquid films on a vertical wall under the presence of dense carbon dioxide by means of a universal measure, i.e. the wetting angle that serves as the wetting parameter. A stability study of the

* Corresponding author. Tel.: +49 40 42878 3191; fax: +49 40 42878 2859.

E-mail address: r.eggerts@tu-harburg.de (R. Eggers).

Nomenclature

b film width
 R circle equivalent radius of film cross-section

Greek symbols

α_0 angle
 δ thickness
 θ_0 film wetting angle/contact angle
 σ interfacial tension

Subscripts

F film
 lv liquid–vapour
 sl solid–liquid

falling film and its dependence on operating conditions is included at the end of this report.

2. Experimental

Falling film experiments are performed in an approximately 350 ml pressure-resistant-view-cell made of stainless steel. The view-cell is designed for a maximum temperature of 393 K and a maximum pressure of 50 MPa (see Fig. 1).

Liquid can be pumped directly from the high-pressure vessel onto the wall in the view-cell. The wall has dimensions of approximately 115 mm × 15 mm or an area of 1725 mm². Through the windows, the formed falling film can be recorded by means of a camera. By taking pictures from two sides of the view-cell, the film thickness and the wetting area can be recorded simultaneously and the figures can be analysed by means of image processing software.

In this work pure water is used as the film phase and pressurized carbon dioxide as the continuous phase. Solid materials used in the experiments are glass and stainless steel. The experiments are performed at 313 K and up to 27 MPa at a water mass flow between 1 g/min and 9 g/min.

3. Results and discussion

3.1. Qualitative

Some authors [8–12] distinguish between a rivulet and a falling film. A falling film is a continuous, wide-covering liquid flowing along a vertical wall. In contrast, a rivulet is a streamlet whose width and thickness are of the same order of magnitude [12]. Such a distinction is not made in this work. The whole experiment is performed in a high-pressure chamber having a viewable width of only 18 mm. The ratio of the liquid film’s width to its thickness

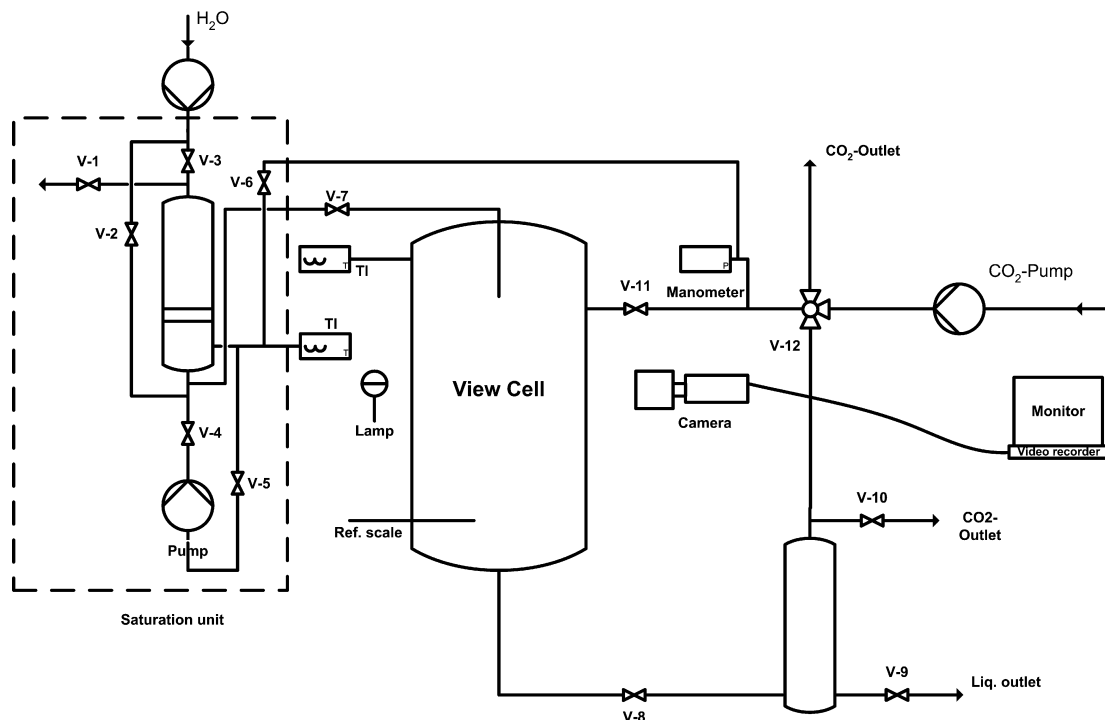


Fig. 1. Experimental setup.

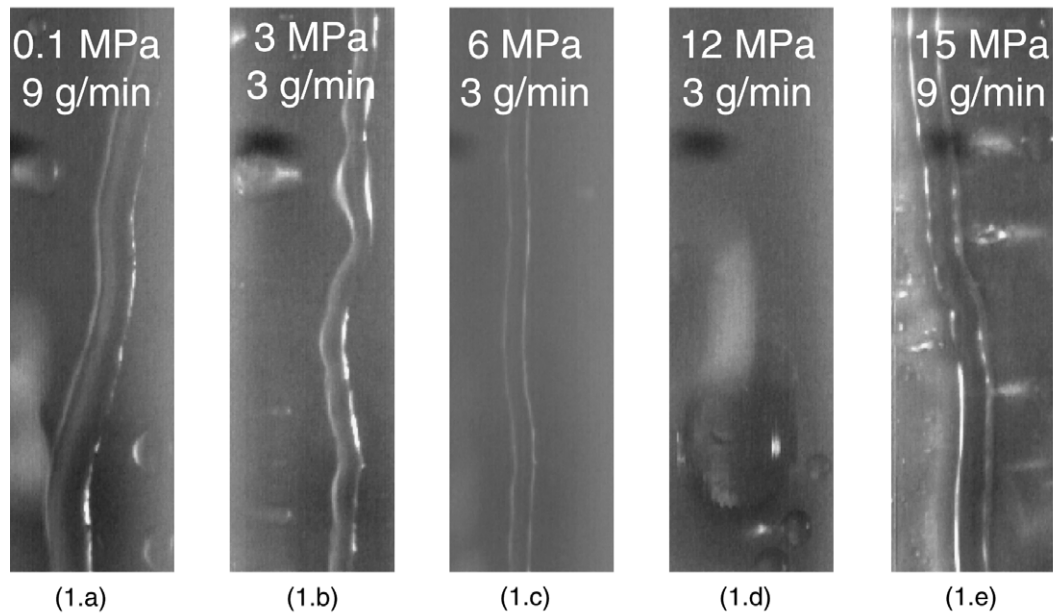
is quite large. It varies from 5 to 25 depending on the solid material studied and the operating conditions. It is justifiable to speak of a falling film in this case. Furthermore, it is believed that the wetting of a wider film can only be better than that of a rivulet. The knowledge obtained in this study increases the understanding of the wettability of a wide falling film.

The recording of pure water film on steel (1.a–1.e) and on glass (2.a–2.e) surfaces at different operating conditions can be seen in Fig. 2. Comparing the images with each other, obviously the film width depends on the system pressure and the film flow rate. The liquid does not always take

the shortest path between two points (as in Fig. 2(1.c)), but meanders instead [6] (Fig. 2).

The side projection, which shows the film thickness, can be observed in Fig. 3. An even film surface such as shown in Fig. 3a is rather an exception. Normally, the film surface is wavy and the waviness itself is influenced by the operating conditions, such as the system pressure and the film rate of flow. Increasing the pressure at a constant flow rate causes a higher buoyancy experienced by the film due to the smaller difference in density between the phases. The liquid is retained and accumulated. As a result, a thicker, uneven film such as shown in Fig. 3c and d is obtained.

Water rivulet on steel surfaces



Water rivulet on glass surfaces

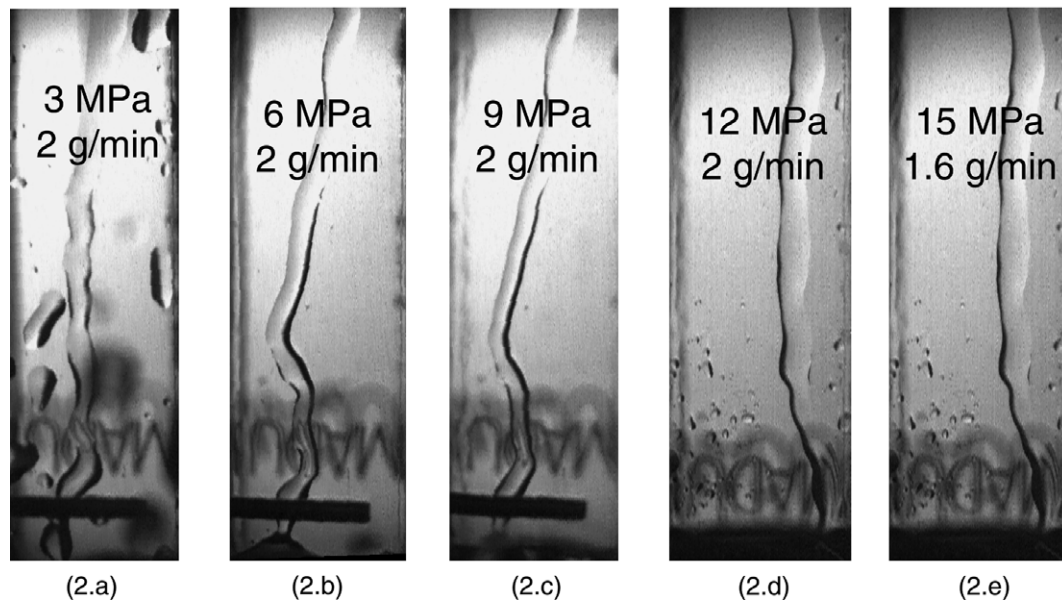


Fig. 2. Film wetting area on steel (1) and on glass (2) surfaces. The magnification used to record films on steel are different from that applied for films on glass surface.

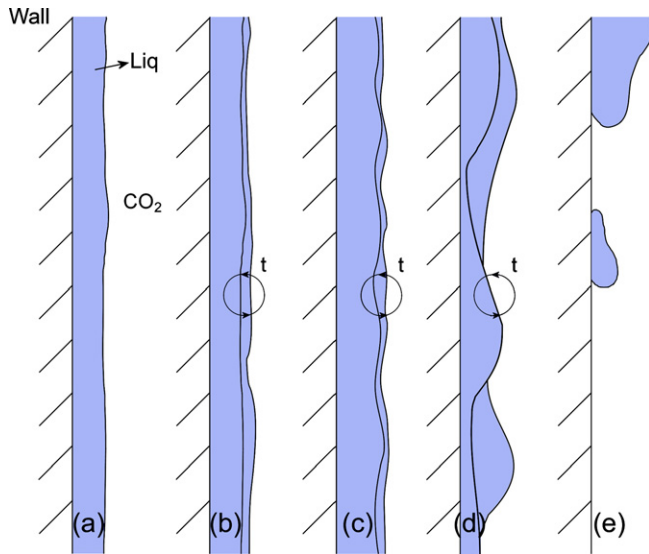


Fig. 3. Film thickness seen from the side view.

A further increase in pressure causes film disintegration (Fig. 3e).

If the flow rate is increased and the pressure kept constant, the waviness can be reduced. A large increase in the flow rate causes not only an even film surface but also a wider and thicker film, so that the wettability cannot be judged solely by means of the film width or its thickness.

3.2. Measurement results

The film measurement results on steel and glass surfaces at 313 K can be seen in Figs. 4 and 5. On the steel surface, in a pressure range of 0.1–27 MPa, the film thickness varies from 0.4 mm to 0.9 mm. An increase in the flow rate from 6 g/min to 9 g/min does not result in any appreciable effect on the film thickness. Due to the irregular film thickness

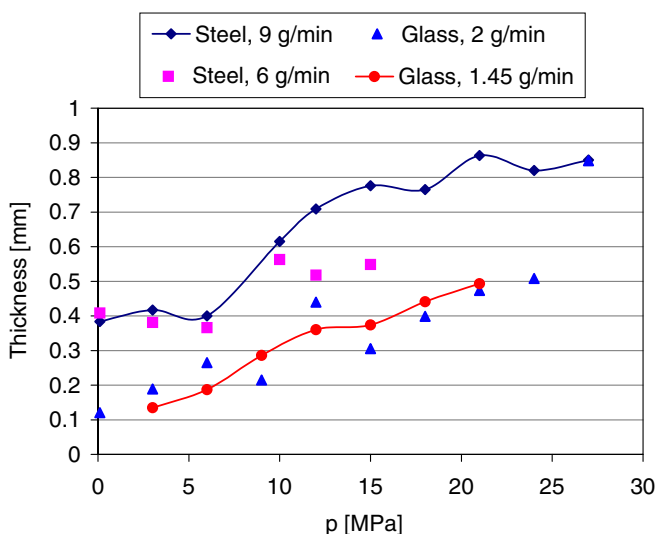


Fig. 4. Film thickness on steel and glass surfaces as a function of pressure.

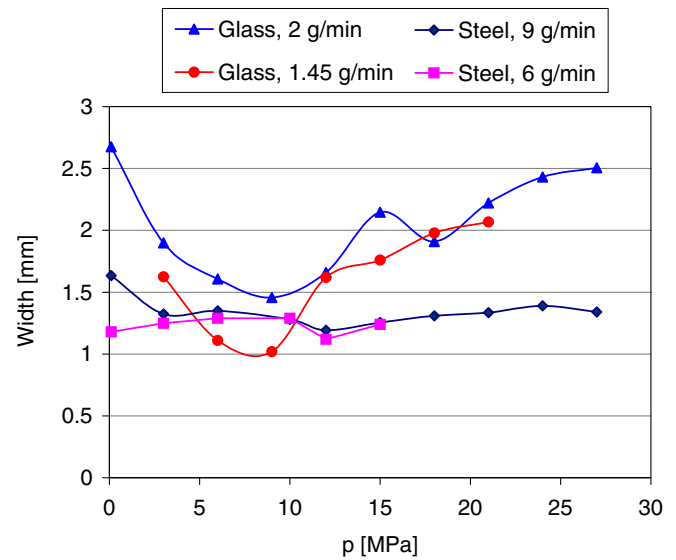


Fig. 5. Water film width on glass and steel surfaces as a function of pressure.

and the film width along the distance, the reported results are the mean values.

Looking at Figs. 4 and 5 it can qualitatively be concluded that water wets steel better in gaseous states than it does in supercritical carbon dioxide. Under increasing pressure the film width remains nearly constant while film thickness rises.

As on the steel surface, the film thickness on the glass depends nearly linearly on the pressure and varies from 0.1 mm to 0.5 mm. The film thickness on the glass is smaller than the film thickness on steel. This thin film is probably not only due to the smaller water flow rate but also controlled by its own wetting characteristics. The film width has a kind of parabolic dependence on the system pressure. It varies from 1 mm to 2.5 mm.

The film geometrical data does not allow a direct interpretation of the wetting ability of the water film on a glass surface. One parameter which contains and therefore combines two sets of information – the thickness and the width – will be needed in order to characterize the wettability of a system clearly and universally.

4. Falling film wetting angle

4.1. Theory

The width and the thickness of the falling film are used to reconstruct the film geometry. For this purpose, theoretically, the film is cut perpendicularly to its flow direction as shown in Fig. 6a. It is assumed that the cross-sectional area has the shape of a circular segment [8,12,13] as shown in Fig. 6b and c. The film's lateral wetting angle θ_0 is defined as the angle between the tangent at the three-phase-contact-point (see Fig. 6b) and the solid surface, or, according to the contact angle definition, the angle between the vec-

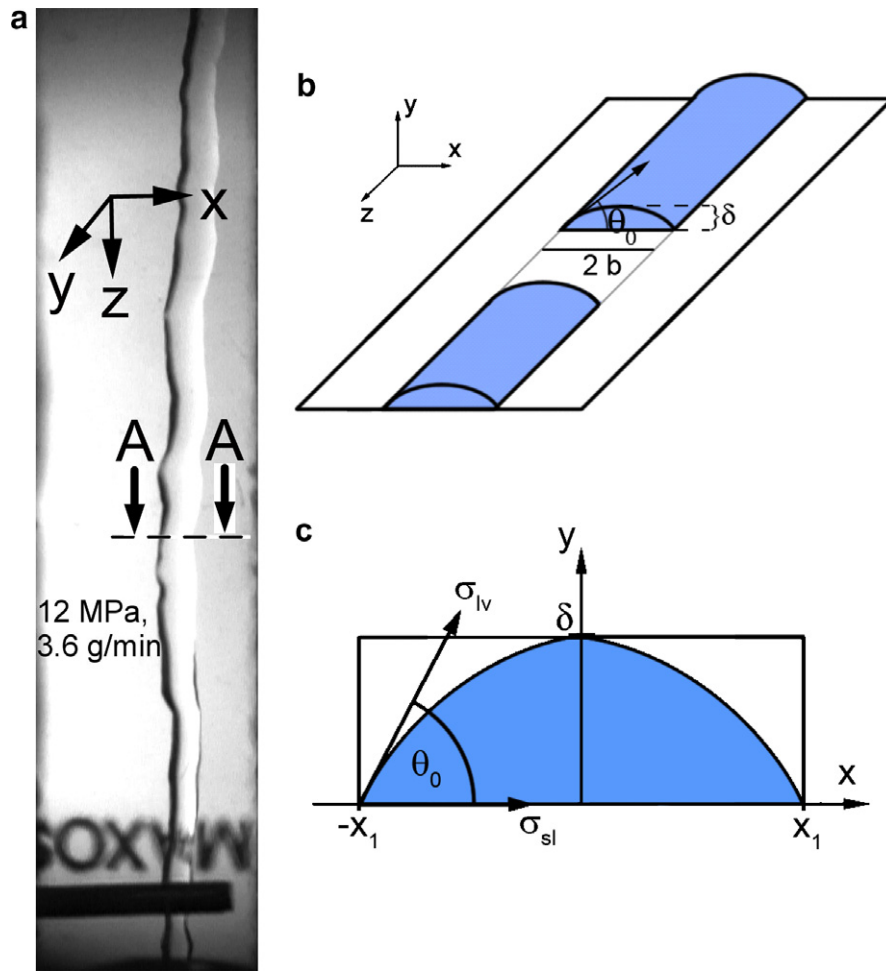


Fig. 6. Theoretical approach in understanding the wetting phenomena by means of the wetting angle. Circular film cross-section is assumed.

tors of the solid–liquid σ_{sl} and the liquid–vapour interfacial tension σ_{lv} in the x – y plane (see Fig. 6). The film’s lateral wetting angle is calculated according to Eq. (1) when the wetting angle is less than 90°

$$\theta_0 = \arcsin \left(\frac{2b\delta_F}{b^2 + \delta_F^2} \right). \tag{1}$$

In unwet cases, that is, when the wetting angle is equal to or larger than 90° , it is calculated according to Eq. (2)

$$\theta_0 = \alpha_0 + \frac{\pi}{2}, \tag{2}$$

with the definition of α_0 as it is shown in Eq. (3)

$$\alpha_0 = \arcsin \left(\frac{\delta_F - b}{b} \right), \tag{3}$$

b is half of the film width, δ_F is the film thickness, θ_0 is the film wetting angle and α_0 is the angle gained when the right angle is subtracted from θ_0 (see Eq. (2)). The circle equivalent radius of the cross-section is calculated by applying Eq. (4)

$$R = \frac{b^2 + \delta_F^2}{2\delta_F}. \tag{4}$$

When the wetting angle is larger than 90° , that is, the film thickness δ_F is greater than half of the film width b , the circle equivalent radius is equal to b .

4.2. Water falling film wetting angle

The calculation results of the water film wetting angle on steel and glass surfaces at 313 K can be seen in Fig. 7. A comparison is shown between the sessile drop contact angle [14] and the falling film wetting angle at different mass flow. It is obvious that in the whole pressure range, the sessile drop contact angle represents the upper limit of the falling film wetting angle. If there is no respective data available in the literature, the sessile drop contact angle can be used by rule of thumb as a prediction of the falling film wetting angle. The difference between the sessile water drop contact angle and the water film wetting angle on both steel and glass surfaces, ranges between 10° and 20° . In general water film wettability decreases when the pressure is increased.

The increase of the wetting angle due to the rise in the mass flow (Fig. 7) leads to the conclusion that the extra

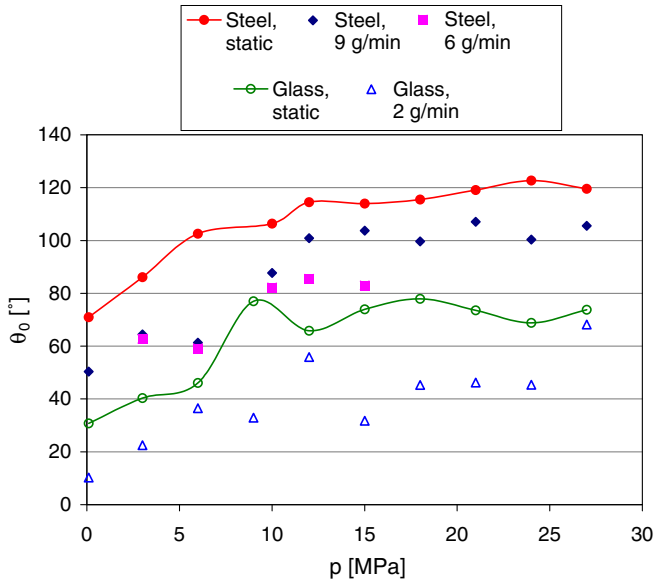


Fig. 7. A comparison of sessile water drop contact angle and film wetting angle on steel and glass surfaces over a pressure range of 27 MPa.

amount of liquid added onto the vertical surface accumulates in the *y*-direction (Fig. 6), and so causes a thicker film. But this only occurs when the available force which results from the interfacial tension σ_{lv} is able to hold the whole geometry stable. Once the force equilibrium point is reached, that is, the supplied force is just as large as the one needed to maintain the film geometry, no more liquid can be added in the *y*-axis. A further increase in mass flow will cause a spreading of the film. This spreading can be explained two ways. The first is the force inserted by the pump which acts in a disruptive way. Higher mass flow means simply an increase in this disruptive force which promotes the spreading.

The second explanation is a geometrical one: having the same area, a cross-section with a smaller wetting angle but a larger circle equivalent radius delivers a greater length in circumference (see Fig. 8). This in turn, provides larger interfacial force. Interfacial force is the force gained by multiplying the interfacial tension by the circumference around which the tension acts. The attempt to enlarge the circumference does not take place as long as the supplied force is enough to hold the whole film construction in the *x*-*y* plane stable. As soon as the force is exceeded, for instance by an increase in the flow rate, liquid needs

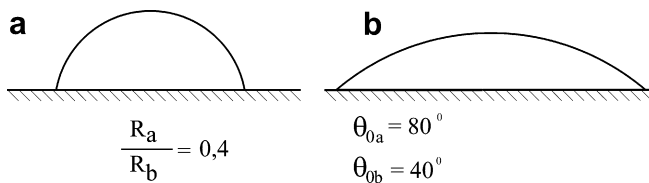


Fig. 8. The lengthening of circumference due to spreading of drop. A wetting angle decrease from 80° (a) to 40° (b) causes a 150% rise in the circle equivalent radius and a 22% increase in the circumference whereas the drop area remains unchanged.

to move sideways and thus spreads in order to provide the required additional force. Thereby the whole construction is held stable by changing its own geometry to a more advantageous one. For example: decrease in the wetting angle from 80° to just half, that is 40°, delivers – at a same cross-sectional area – a circle equivalent radius which is 2.5 times larger than the former one and gives a 22% increase in the circumference (see Fig. 8).

A direct comparison between the wettability of the water film on a steel and a glass surface shows us that glass – like the cases of sessile drops [10,11] – is better covered by the film. The wetting angle on glass varies from 10° to 60°. On steel, the angle ranges between 50° and 110°. The poor wettability of the water film on steel surfaces can also be seen in the circle equivalent radius of the cross-section calculated according to Eq. (4). On steel the radius ranges between 0.5 mm and 0.8 mm whereas on glass it varies from 0.6 mm to 2.5 mm.

5. Film instability

The water film wetting angle becomes smaller with decreasing flow rate as can be seen in Fig. 7. But decreasing the flow rate consistently causes an instability in the film flow. Instead of flowing continuously down the vertical plate, liquid stagnates and accumulates at certain points on the wall. At the beginning there is a mixture of liquid film and liquid drops, which are created by the narrowing down of the film (see Fig. 2, image (1.d) and Fig. 3e). This mixture flows unsteadily down the wall. The critical mass flow at which falling film becomes unstable is called break-up- or film disintegration-points. In Fig. 9 the break-up-points of water film on glass and steel surfaces up to 27 MPa can be seen. For both liquids the break-up-points increase as the pressure goes up. This agrees very well with the wetting angle data reported before (Fig. 7), where the wettability decreases as the pressure increases.

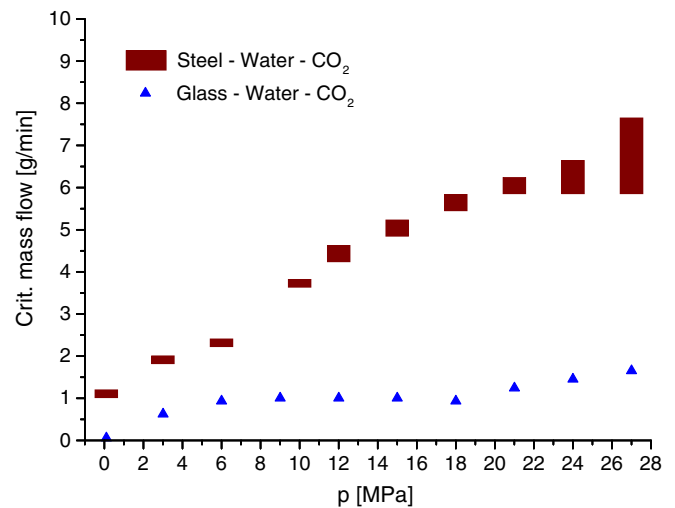


Fig. 9. Critical mass flow of water falling film on steel and glass surfaces at 313 K.

For the system of water film on a glass surface, only the beginning of the film disintegration is shown in Fig. 9. At and above this rate of flow a pure film regime can be expected. Below the given rate of flow, there will be either a mixture of film and drop or only water drop.

The top of the bars in Fig. 9 shows the start of film disintegration on steel, that is to say, the end of the pure film regime. The lower end of the bar shows the end of the film-drop regime. This also indicates the start of the pure drop regime from which only drops flow unsteadily down the wall. The bar itself represents a transition zone, from the film regime down to the drop regime. The same pressure effect, as on glass, can also be observed here.

6. Conclusions

The geometry of a falling film of water flowing down a stainless steel or glass surface by gravitational force has been measured. The flow may be decelerated by the wall, the buoyancy and the shear stress on the film surface. For easier handling of measurement data, and thus gaining clearer characterization of the wetting ability, the lateral film wetting angle θ_0 is introduced.

Comparing the results of the wetting angles of water films with the sessile drop contact angles on both, steel and glass surfaces, it can be stated that the tendency of the wettability of water on these media remains the same. The wettability on a vertical wall also decreases when pressure rises. The sessile drop contact angle represents the upper limit of the film wetting angle on both solid surfaces. Depending on the operating conditions, the difference between the two varies from 10° to 20° . Water film can, under all investigated operating condition, wet a glass surface better than a steel surface.

An increase in mass flow up to a certain value leads to a decrease of the wettability of water. However, a further increase in the film rate of flow beyond this limit causes film spreading.

In contrast, a continuous decrease in the water mass flow will destabilize the film. The film disintegrates into droplets. At this point, the available amount of flowing liquid on the surface is not enough to guarantee a wide covering film. The mass flow at which a film breaks into drops

is referred to as the critical mass flow. On both surfaces, the critical mass flow increases if the pressure is raised.

Acknowledgement

The authors gratefully acknowledge the financial support provided by the German Research Community (DFG), project DFG Eg 72/12-2.

References

- [1] W. Brötz, Über die Vorausberechnung der Absorptionsgeschwindigkeit von Gasen in strömenden Flüssigkeitsschichten, *Chem. Ing. Tech.* 26 (8/9) (1954) 470–478.
- [2] W.L. McCabe, J.C. Smith, P. Harriot, *Unit Operations of Chemical Engineering*, fifth ed., McGraw-Hill, New York, 1994, pp. 325–346.
- [3] A.B. de Haan, J. de Graauw, Mass transfer in supercritical extraction columns with structured packings for hydrocarbon processing, *Ind. Eng. Chem. Res.* 30 (1991) 2463–2470.
- [4] W. Nusselt, Die Oberflächenkondensation des Wasserdampfes, *Z. Vereines Deutscher Ingenieure* 60 (27) (1916) 541–546, 569–575.
- [5] T. Young, An essay on the cohesion of fluids, *Philos. Trans. Roy. Soc. London* 95 (1805) 65–87.
- [6] J.B. Culkun, S.H. Davis, Meandering of water rivulets, *AIChE J.* 30 (2) (1984) 263–267.
- [7] J. Kern, Untersuchungen über die Hydrodynamik der Rinnsal, Ph.D. Thesis, Technische Universität Berlin, 1970.
- [8] G.D. Towell, L.B. Rothfeld, Hydrodynamics of rivulet flow, *AIChE J.* 12 (5) (1966) 972–980.
- [9] B. Janocha, Veränderung von Benetzung und Adsorption an Kunststoff-Wasser-Grenzflächen unter dem Einfluß externer elektrischer Felder und der Kunststoffoberflächenpolarität, Ph.D. Thesis, Eberhard-Karls-Universität Tübingen, 1998.
- [10] P. Jaeger, Grenzflächen und Stofftransport in verfahrenstechnischen Prozessen am Beispiel der Hochdruck-Gegenstromfraktionierung mit überkritischem Kohlendioxid, Ph.D. Thesis, Technische Universität Hamburg Harburg, 1998.
- [11] Y. Sutjiadi-Sia, H. Marckmann, R. Eggers, C. Holzknecht, S. Kabelac, Zum Einfluss von in Flüssigkeiten unter Druck gelösten Gasen auf Grenzflächenspannungen und Benetzungseigenschaften, *Forsch. Ingenieurwes.* 71 (2007) 29–45.
- [12] J. Kern, Zur Hydrodynamik der Rinnsale, *Verfahrenstechnik* 3 (1969) 425–430.
- [13] S.G. Bankoff, Minimum thickness of a draining liquid film, *Int. J. Heat Mass Transfer* 14 (1971) 2143–2146.
- [14] T.S. Martin, Wetting Characteristics and Surface Tension of Several Solids under Elevated Pressure of Carbon Dioxide, Diploma thesis, Technische Universität Hamburg-Harburg, 2005.