Dynamic contact angle and three-phase contact line of water drop on copper surface

E G Orlova, D V Feoktistov and K A Batishcheva
Thermal Theory and Engineering Department, Power Engineering Institute, National Research Tomsk Polytechnic University, 30, Lenin Avenue, 634050, Tomsk, Russia

E-mail: enja1991@rambler.ru

Abstract. Nowadays there is a lack of experimental data describing the physical process of drop spreading on a solid metal surface for developing wetting and spreading theory. The experimental data obtained by using the high speed video-recording will allow to identify unknown previously spreading modes as well as the change of the dynamic contact angle and the three-phase contact line. The purpose of the work is to determine the effect of the drop growth rate and the copper substrate surface roughness on the dynamic contact angle and the three-phase contact line speed at distilled water drop spreading. Shadow and Schlieren methods are used to obtain experimental data. Three drop spreading modes on the rough surfaces were identified. Time dependences of the dynamic contact angle and contact line speed were obtained. Experimental results can be used for assessing the validity of the developed mathematical models of wetting and spreading processes in the field of micro- and nano-electronics, ink jet printing, thin-film coatings, spray cooling, and optoelectronics.

1. Introduction
The surface microstructure and conditions of drop formation (growth rate) are well known [1-6] to influence the hydrophobic and hydrophilic properties of materials. The theory of wetting and spreading processes is not developed at a level providing an opportunity of heat exchangers’ construction with a patterned surface that intensifies heat transfer. One of the factors suppressing the development of scientific achievements in this sphere is an insufficient number of experimental results describing the spreading process taking into account the droplet formation conditions. Experimental data of the dynamic contact angle (DCA) and the three-phase contact line speed can be used to assess the validity of the developed mathematical models of wetting and spreading processes.

The purpose of the presented work is to obtain experimentally dependences on DCA and the three-phase contact line speed from the drop growth rate and the surface roughness of copper substrates.

2. Research technique
Research was conducted by using the experimental setup (figure 1) consisting of the equipment for shadow and Schlieren methods implementation [7, 8].

In the shadow optical method light source 3 with lens (placed in the widest part of the light source case) was used to produce a beam of plane-parallel light illuminating the drop on the substrate. The distance between the lens and the object under investigation must be greater or equal to the focal length (h ≥ F).
In Schlieren method incoherent light source 4, ground glass 10 and coding filter 11 were used for a light flux with a stepped decrease of intensity on the space. A light beam from source 4 passed through collimating lens 5, which transformed it into a plane-parallel one. Then it was reflected from beam splitter 9, fell on to the substrate, passed to lens 8 and projected on the sensor of the high-speed video camera 7. The transparent shield with an opening 6 is set for reducing the influence of external light sources.

![Figure 1](image)

**Figure 1.** A schematic view of the experimental setup: 1 – photographic camera; 2 – macro lens; 3 – light source of Shadow method; 4 – light source of Schlieren method; 5 – collimating lens; 6 – transparent shield with an opening; 7 – high-speed video camera; 8 – lens; 9 – beam splitter; 10 – ground glass; 11 – coding filter.

Video recording of spreading drops on the surface was carried out simultaneously in two coordinate directions. The equipment for Schlieren method implementation was used to control the drop symmetry. If the drop lost symmetry, the experiment was repeated.

Three copper substrates (54 mm in diameter and 4 mm thick) with a through opening (1 mm in diameter) located on the center were used. The surface microstructure of two substrates was formed by bombardment with Al₂O₃ 10 and 100 µm sized particles. The third substrate was made of the flexible copper.

According to results of the preliminary experiment, the values of influencing factors were defined (table 1).

| Parameter                  | Value                                      |
|----------------------------|--------------------------------------------|
| Drop volume (ml)           | 0.3                                        |
| Drop growth rate (is controlled by syringe pump, Figure 5) (ml/s) | 0.005; 0.02; 0.04; 0.08                   |
| Substrate material         | Copper                                     |
| Roughness parameter of surface – Ra (µm) | Sample № 1 – flexible copper with Ra 0.591; Sample № 2 – copper with Ra 5.190; Sample № 3 – copper with Ra 6.210; |
| Wetting liquid             | nondeaerated distilled water                |
Surfaces of substrates were investigated on the profilometer "Micro Measure 3D station" and microscope TM-3000. The parameter of roughness (arithmetic average roughness Ra) and microstructure were defined. Surfaces microstructure and substrates view are presented in figures 2-4.

**Figure 2.** Sample No 1 – flexible copper Ra 0.591 [µm]: (a) - surface microstructure (Magnification: x3000); (b) - substrate view.

**Figure 3.** Sample No 2 – copper Ra 5.190 [µm]: (a) - surface microstructure (Magnification: x2000); (b) - substrate view.

**Figure 4.** Sample No 3 – copper Ra 6.210 [µm]: (a) - surface microstructure (Magnification: x2000); (b) - substrate view.
The roughness of the sample No 1 (flexible copper) is formed by longitudinally arranged grooves. The roughness of the samples No 2 and No 3 (copper substrates) is formed by chaotically arranged asperities and cavities.

The drop was formed on the surface by the syringe pump (Cole-Parmer Touch Screen) (Figure 5). The nondeaerated distilled water was squeezed on the surface through the channel placed in the substrate. This bottom-up methodology of droplet formation in comparison with the known by syringe dispenser [9, 10] facilitates a precise control of droplet formation and size, as well as allows to reduce the error at maintaining the initial volume.

![Figure 5. The bottom-up methodology of droplet formation on the surface.](image)

The drop profile was obtained by the Shadow method. The equipment for implementation of this method includes the high-speed video camera (FastVideo-500M) and the light source (MI-150 Edmund). Geometrical parameters (contact diameter, height and contact angle of the drop) were defined at processing images in Drop Shape Analysis (DSA) software (KRUSS Company).

According to the obtained experimental data, the contact line speed was calculated. This parameter describes the rate of spreading drops on a surface and is determined from the following equation:

\[
V_k = \frac{C \cdot (d_3 - d_1)}{2 \cdot (n_3 - n_1)}
\]

where C is a frame capture speed, frame/s; \(d_1\), \(d_3\) are droplet diameters in the previous and the next moment, mm; \(n_1\), \(n_3\) are numbers of the frame at previous and the next moment in time, frame.

3. Results and discussion

Time dependences on DCA and the three-phase contact line speed at the drop growth rate from 0.005 to 0.08 ml/s are shown in figure 6.

Three drop spreading modes on the rough copper surfaces were conditionally subdivided. The first mode is characterized by an abrupt increase in the three-phase contact line speed. The advancing DCA and speed decrease monotonously in the second mode. The equilibrium contact angle at the constant wetted area is formed in the third mode.

Duration of identified modes in experiments from all time of spreading equaled: the first – 1-2%, the second – 39-50%, the third – 48-60%.

According to the data obtained by the authors [11, 12], the spreading process after collision of a drop with a solid occurs simultaneously in the two modes. The first mode is the movement of liquid within the grooves of a roughened surface. The second is spreading liquid over the grooves of a surface. At high velocities of collision the first mode dominates. At low velocities the movement over the grooves prevails. It is obvious, that these modes are implemented at drop spreading under the gravity force without collision with a substrate. However, when applied the syringe pump at drop placing (bottom-up method), at high drop growth rate (above 0.010 ml/s) the movement of liquid over the grooves is dominant. In this case cavities of microstructure are filled with gas, and the contact of liquid
with a solid substrate represents a heterogeneous interphase "liquid-solid-gas". So that, the liquid drop moves over "air cushion" [13]. At low drop growth rate (0.005ml/s) the dominant mode is the motion of liquid within the grooves, and the interphase is "liquid-solid".

Figure 6. Time dependences on DCA and the three-phase contact line speed. The drop growth rate, ml/s: a – 0.005; b – 0.02; c – 0.04; d – 0.08. The advancing DCA: ○ – sample No 1; Δ – sample No 2; ■ – sample No 3. The three-phase contact line speed: ☉ – sample No 1; ▲ – sample No 2; □ – sample No 3. Spreading modes are shown by Roman numbers: I, II, III.

During the second mode slight increase in DCA was observed at drop spreading on the surface of sample No1 (flexible copper) with the drop growth rate 0.005 ml/s. According to the "model of parallel grooves" proposed in [14-16], at a parallel arrangement of contact line relative to the grooves on the surface, a great number of pinning is possible. The increase in DCA on the surface with parallel grooves can be explained as follows: by increasing roughness, firstly, natural decrease of $\theta_d$ is found, but when cavities becomes deep enough, there is a complex structure with lower energy barriers, the value of $\theta_d$ increases.

It was found that the increase in the drop growth rate at spreading on the surface with $Ra=5.190 \mu m$ (Sample No 2, Figure 3) led to increasing the maximum value of DCA of 30%; on the surface with $Ra=6.210 \mu m$ (Sample No 3, Figure 4) of 37%. DCA increasing on the surface with $Ra=0.591 \mu m$ (Sample No 1, Figure 2) equaled no more than 8%. 
According to the analysis of the experimental results, it was established that non-wetting is replaced by wetting at drop spreading on the copper substrates. The advancing DCA during the contact line movement decreases and after a while (depending on the drop growth rate and the surface microstructure) the contact angle becomes smaller than 90°.

At high drop growth rate (0.8ml/s) change of the contact line speed occurs non-monotonically. Since at the movement on surface the drop loses an equilibrium state and oscillates.

Random error is calculated based on the experimental results and equals to 3.9% for contact angle and to 1.87% for the contact line speed. Three experiments at fixed factors were performed. Systematic error was measured by syringe pump error Cole Parmer ± 0.355%.

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