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Influence of Surfactant Concentration on the Critical Current Density

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1. Introduction

Boiling heat transfer allows larger heat transfer coefficient than single phase heat transfer. When the heat flux goes over a critical limit, the heat transfer is disturbed by the vapor film and the surface temperature abruptly increases, which is called Critical Heat Flux(CHF). Particularly in the nuclear power plants, the measurement of CHF is important as it denotes the upper heat transfer limit [1]. However, it is difficult to perform the CHF experiments due to the extreme test conditions, which results in failure of test specimen and measurement devices. To avoid this experimental difficulty, an alternative experimental method is explored.

In an electrochemical hydrogen evolving system such as water electrolysis, the cell potential increases as the current density increases due to the increased hydrogen generation rate. However, when the current density arrives a certain critical limit, hydrogen film is formed and the cell potential increases abruptly. This phenomenon is known as Critical Current Density(CCD), which is quite similar to the CHF [2]. The gas film formation phenomenon from gas bubbles seems to be governed hydrodynamically. Thus, we postulated that there is a certain analogous relationship between the CHF and CCD [3].

The CHF depends strongly on the surface tension [4]. There have been many studies to manipulate the surface tension. Typical methods of manipulating the surface tension are the modification of heat transfer surface through nanoparticle coating and use of surfactants. In the existing researches, it has been demonstrated that the CHF is decreased as the surfactant concentration is increased [5,6].

Raza et al. [6] used various surfactants and observed change of the CHF. In the existing CHF model, the CHF is proportional to the surface tension to power 0.25 [5]. Raza et al. [6] identified that experimental measurements of CHF did not follow this relationship and proposed that CHF can not be explained in surface tension by one. Raza et al. [6] suggested that the bubble behavior according to bubble size is another factor, which affects the CHF. In this paper, the CCD values were measured according to the surfactant concentration. The analytic model from Raza et al. [6] was imitated to explore similarities between the CHF and CCD.

2. Experimental setup

2.1 Test matrix

The range of present study is specified in Table 1. The surfactant used in this experiment is cetrimonium bromide (CTAB). We carried out experiments near the critical micelle concentration (CMC; 298 ppm at CTAB [7]). The surface tensions of working fluid were measured using surface tensiometer.

Table 1. Range of present study

Concentration of CTAB (ppm)	4	11	66	74	146	219	291
Surface tension (mN/m)	73.22	66.50	60.81	35.17	36.80	35.58	35.11
Contact angle (degree)	73.49	73.09	68.82	64.59	64.59	54.17	53.62

2.2 Test Apparatus

Figure 1 shows the experimental apparatus and electric circuit. We used 1.5 M aqueous solution of sulfuric acid (H_2SO_4) and added CTAB. 10 mm diameter copper disk cathode is used. The cathode and anode are located in the H_2SO_4 solution at atmospheric condition and room temperature (294 K). The high speed camera is used to record the bubble behavior at the cathode. Power supply is used to control the cell potential and current density. The measured data were recorded by the data acquisition (DAQ) system.

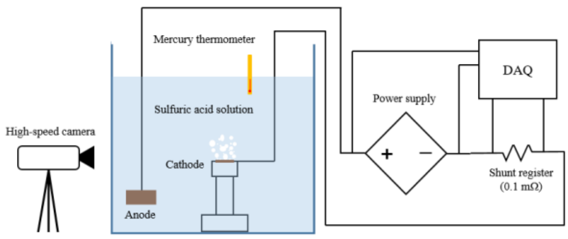

Fig. 1. Experimental apparatus and test section.

3. Result and Discussion

3.1 Bubble behaviors according to the surfactant concentration

Figure 2 shows the influence of surfactant concentration on the bubble behavior in both boiling system and hydrogen evolving system near the CHF and CCD respectively. In the boiling system, the size of bubble is decreased as the surfactant concentration increased [6]. In the hydrogen evolving system, similar tendency was observed. Increased surfactant concentration allows increased surfactant diffusion in liquid–vapor interface, which results in the decreased bubble size [8,9].

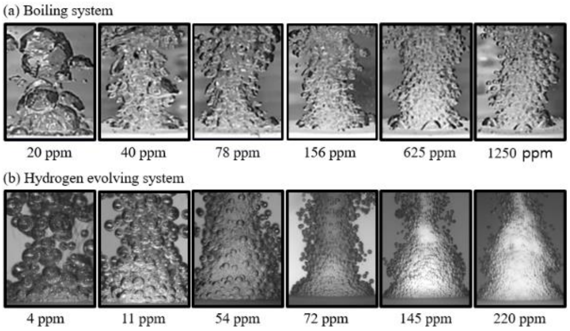

Fig. 2. Variation of bubble size according to the surfactant concentration; (a) Near the CHF (SDS) [6]. (b) Near the CCD (CTAB).

Figure 3 shows the bubble behavior due to the heat flux and the current density. In the boiling system, as the heat flux increased, the size of bubble is increased [6]. In the hydrogen evolving system, the similar tendency is appeared. When the heat flux and current density are increased, the generation rate of bubble is increased then the coalescence is forced due to the crowding effect.

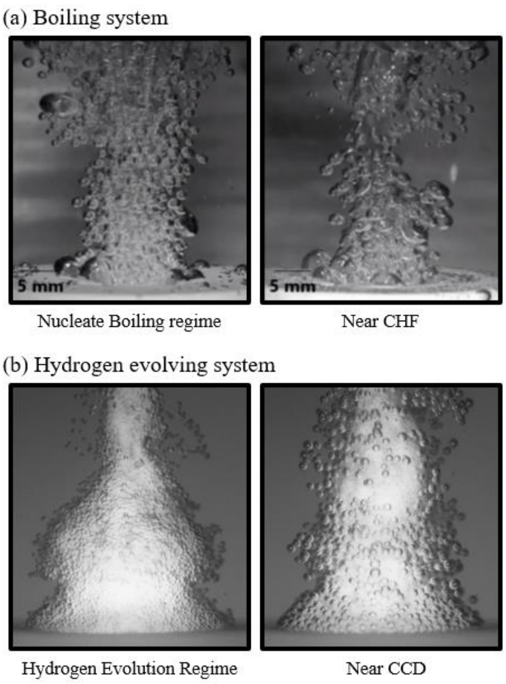

Fig. 3. Comparison of bubble behavior according to the bubble generation rate (a) SDS solution in boiling system [6]. (b) CTAB solution in hydrogen evolving system.

3.2 Influence of bubble behavior on CCD

To analyze the influence of surfactant on the CCD, we borrowed Raza et al.'s method [6]. The CHF can be calculated by the heat balance equation as expressed in the Eq. (1) [10].

$$q''_{CHF} = h_{lv}\rho_v U A_R. \tag{1}$$

Where A_R is the area ratio and U is superficial velocity. By the force balance between the buoyancy and drag force, the bubbles must be reached terminal velocity (U_T)

$$(\rho_l - \rho_v) g \frac{1}{6}\pi D^3 = \frac{1}{2} C_d \rho_l \frac{\pi}{4} D^2 U_T^2. \tag{2}$$

Solving for the U_T, we get

$$U_T = \sqrt{4/(3C_d)}\sqrt{(\rho_l - \rho_v) g D/\rho_l}. \tag{3}$$

Then the U in the Eq. (1) can be substituted by U_T. Hence, combining the Eq. (1) and Eq. (3) we get

$$q''_{CHF} = h_{lv}\rho_v A_R \sqrt{4/(3C_d)}\sqrt{(\rho_l - \rho_v) g D/\rho_l}. \tag{4}$$

By applying analogous relationship, the CCD can be calculated as follow:

$$I''_{CCD} = \frac{nF}{m}\rho_v U A_R. \tag{5}$$

Where n is valence number of copper ion, F is Faraday constant and m is molar mass. Combining the Eq. (5) and Eq. (3), we get

$$I''_{CCD} = \frac{nF}{m}\rho_v A_R \sqrt{4/(3C_d)}\sqrt{(\rho_l - \rho_v) g D/\rho_l}. \tag{6}$$

The diameters of hydrogen bubbles, D were estimated by measuring bubble diameter just before the CCD. We obtained the area ratio, A_R and drag coefficient, C_d as follow. We assumed that the bubbles are packed in hexagonal close packing and bubble based area (A_{bubble}) was estimated by using the contact angle.

$$A_R = \frac{A_{bubble}}{A_{hexagonal}} = \frac{\pi \sin^2\theta}{2\sqrt{3}} \tag{7}$$

The relationship between the measured CCD and bubble diameter was obtained as shown in the Fig. 4(a). The measured CCD was proportional to the $\sqrt{D}$, which implies the C_d can be regarded as the constant value based on the Eq. (6). The C_d was estimated as 4.87 by performing regression analysis. The similar results were reported in the CHF as shown in the Figure 4(b).

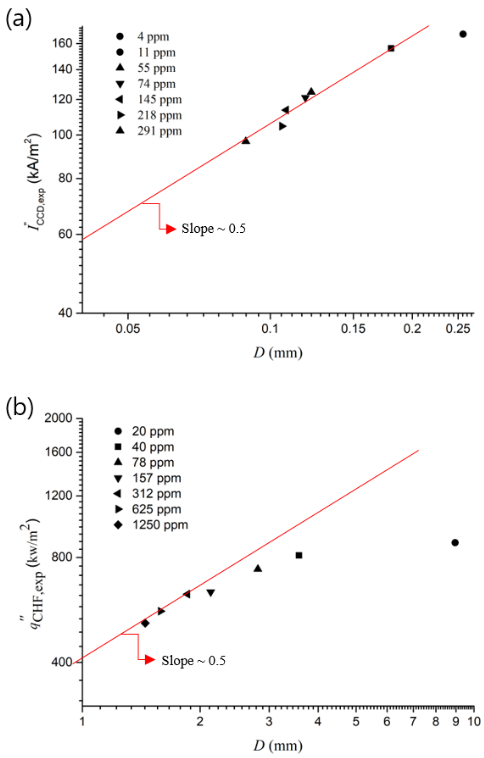

Fig. 4. Relationship between CCD/CHF and bubble diameter. (a) Present result. (b) Reproduced from Raza et al.'s result [6].

Based on the previous analysis, the prediction of CCD using Eq. (6) was represented in Figure 5. The Eq. (6) predicts well for the high surfactant concentration. Because in real situation the C_d would be increased in the large D cases, the discrepancies (within 20%) were appeared.

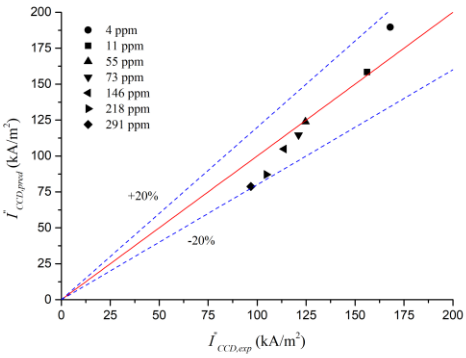

Fig. 5. Prediction of CCD using D, C_d and A_R.

Figure 6 shows the complex influence of the surfactant concentration on the CCD/CHF and bubble diameters. It is concluded that the influence of surfactant concentration on the CCD showed similar influence to the CHF.

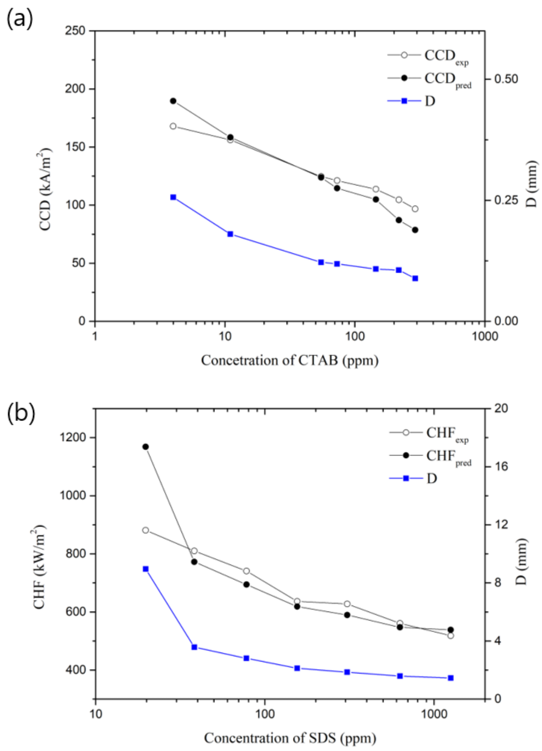

Fig. 6. Surfactant concentration influence on bubble diameter and CCD/CHF. (a) Present result. (b) Reproduced from Raza et al.'s result [6].

4. Conclusion

Influence of the surfactant concentration on the bubble behavior and CHF were simulated by the hydrogen evolving system. The CCD and bubble diameter were measured according to the concentration of surfactant. The similarities of the surfactant concentration influence between hydrogen evolving system and boiling system are discussed.

The hydrogen bubble diameter was decreased as the surfactant concentration was increased. And the hydrogen bubble diameter was increased as current density increased due to the vigorous bubble coalescence. These phenomena were identically observed at the boiling system.

The CCD is closely related to the bubble diameter. When the bubble diameter was decreased, the CCD was decreased due to the reduced U_T. The CCD is predicted well using the bubble parameters D, C_d and A_R, which was similar to the CHF.

It is concluded that the CCD phenomenon is closely related to the bubble behavior, which is based on the hydrodynamic parameters.

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