Heat Transfer in Nucleate Boiling of Binary Mixtures*

(Development of a Heat Transfer Correlation)

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Heat transfer coefficients in nucleate pool boiling of five binary mixtures were measured under atmospheric pressure on an upward-facing heated surface for a wide range of heat flux from about 15 percent of the critical heat flux to close to it. Mixtures were: methanol/water, ethanol/water, methanol/ethanol, ethanol/*n*-butanol, and methanol/benzene. As has been observed in many previous experiments, heat transfer coefficients of mixtures were reduced in comparison with interpolated values between their constituent components. This reduction was a function of the mixture composition and became more pronounced with an increase in heat flux. To provide a simpler and more reliable prediction of heat transfer coefficients of mixtures, Thome correlation was modified so as to include the effect of heat flux in a dimensionless form. A newly developed correlation in this way was verified to predict heat transfer coefficients for the present five mixtures well within ± 20 percent accuracy lines.

Key Words: Heat Transfer, Nucleate Boiling, Binary Mixture, Correlation

1. Introduction

Heat transfer coefficient in nucleate boiling of mixtures is reduced below the coefficient for an imaginary pure substance of the same physical properties as the mixture, or the linearly interpolated coefficient between two pure components comprising the mixture. This reduction was observed to greater or lesser degrees in every boiling experiment of mixture irrespective of the type of mixtures, the size, posture and roughness of the heated surface, system pressure, and other experimental conditions. Thus heat transfer reduction is now an overwhelming knowledge.

Some explanations⁽¹⁾⁻⁽⁷⁾ for the reduction of heat transfer coefficient were given hitherto. As a physically convincing basis, they refer more or less to a growth theory^{(8),(9)} of a spherical bubble in uniformly superheated mixture. According to the theory vapor

bubble grows at slower rate in mixture than in pure substance because of mass transfer resistance in the liquid phase and a resulting temperature rise at the bubble surface. In the case of boiling, however, many bubbles grows at the same time on the heated surface, thus heterogeneously, and their form is non-spherical. Furthermore bubbles interact with each other during their growth period, resulting in deformation, coalescence and breaking up. Finally they depart from the heated surface. Such complex bubble behavior in boiling is very far or rather different from the ideal case of homogeneous spherical growth of a single isolated bubble. Hence the heat transfer reduction in boiling is not directly attributable to the growth rate reduction of a single bubble in mixture. To verify any proposed explanation for heat transfer reduction and resulting correlations, measured data for various mixtures are needed for a wide range of conditions such as composition, heat flux, system pressure and so on.

From the above mentioned background the authors measured boiling heat transfer coefficient for five binary mixtures of different type. The data⁽¹⁰⁾ were used to assess five typical correlations^{(3),(7),(11)-(13)}

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developed for mixtures in the past. It was found that some correlations are fairly good while their accuracy varies considerably with mixture and heat flux. Thus it is difficult to recommend any single correlation that is applicable irrespectively to any mixture. The present study is to develop a new empirical correlation that will enable to correlate the same data measured in the preceeding report⁽¹⁰⁾ well within $\pm 20\%$ accuracy.

2. Experimental Apparatus and Test Mixtures

2.1 Experimental apparatus

The experimental apparatus consists of a boiling vessel and a temperature-controlled air bath. The vessel is a hollow tube of stainless steel with an inner diameter of 104 mm and a height of 395 mm. Glass windows are open for visual observation. At lower position in the vessel is installed a cylindrical heating block of copper, an upper end of which, 40 mm in diameter, serves as the heated surface and is heated by nichrome heaters in slots at its bottom. A thin circular fin, 0.2 mm thick, outside of the heated surface is in a one unit construction with the cylindrical portion. This is to minimize the conduction heat losses and to secure uniform distribution of bubbles on the heated surface. Along the vertical axis of the heated cylinder, three thermocouples were embedded at depths of 1.1, 7.1 and 13.0 mm to determine the heated surface temperature and heat flux assuming one dimensional heat conduction. Refer to Ref.(10) for details of the experimental apparatus and method. To maintain the same surface conditions, the heated surface is carefully polished by No. 0/4 emery paper in advance of each series of measurement, resulting in a surface roughness of $0.125\ \mu\text{m}$ on average.

Experiment begins with degassing the vessel, pouring the test mixture to a 100 mm level above the heated surface, and heating the mixture to the saturation temperature under a pressure of 0.101 MPa by auxiliary heaters. Thereafter, the auxiliary heaters are kept at constant output, the heated surface is powered on and the boiling experiment starts. The vapor formed on the heated surface is condensed in a water-cooled condenser outside of the vessel and returned to the vessel, thus maintaining a constant liquid level and a constant mixture composition during the experiment.

The data were taken for decreasing heat fluxes stepwise from the highest value to avoid the boiling hysteresis and no less than 20 minutes were allowed at each step to stabilize the boiling state. The reproducibility of data was confirmed by repeating the same experiments on different days.

Table 1 Features of five test mixtures

M/W: methanol/water E/W: ethanol/water
E/Bu: ethanol/n-butanol M/E: methanol/ethanol
M/Bz: methanol/benzene

	M/W	E/W	M/E	E/Bu	M/Bz
aqueous	○	○			
non-aqueous			○	○	○
non-azeotropic	○		○	○	
azeotrope -forming		○			○
larger $ Y_1 - X_1 $ and ΔT_{bp}	○	○		○	○
smaller $ Y_1 - X_1 $ and ΔT_{bp}			○		
linear property variation			○	○	
non-linear property variation	○	○			○

2.2 Selection of mixtures

In the present measurement, methanol/water, ethanol/water, methanol/ethanol, ethanol/*n*-butanol, and methanol/benzene were selected as the test mixtures in consideration of availability of phase equilibrium data, reliability of estimated mixture physical properties, and moderate level of temperature for experimental convenience. Other key intention is, as shown in Table 1, to include a variety of mixtures in the measurements: aqueous, organic, non-azeotropic, and azeotrope-forming mixtures, mixtures with a different range of the flash temperature difference, ΔT_{bp} , as well as the vapor-liquid concentration difference, $|Y_1 - X_1|$, and mixtures with their physical properties varying either non-linearly or rather linearly against composition.

3. Presentation of Experimental Results

A total of 765 heat flux vs. wall superheat data were obtained for 39 concentrations of the five mixtures including their pure components. Mole fraction of the more volatile component, X_1 , is used here to express the mixture concentration. Thus $X_1=0$ is for the less volatile component and $X_1=1$ is for the more volatile component. Heat flux was ranged from a lowest about 15% of the critical heat flux (CHF) to a highest close to CHF. To distribute data points evenly in this range, heat flux was changed in almost the same increment on the logarithmic scale.

Most data are found in the fully developed boiling regime. Heat transfer coefficient, α in $\text{W}/(\text{m}^2\text{K})$, defined using the temperature difference between the heated surface and the bubble point of mixture specified at a bulk concentration can be expressed by a power function of heat flux, q in W/m^2 , as

$$\alpha = Cq^n \quad (1)$$

Table 2 Data presentation by $\alpha = Cq^n$, where α is in unit of $\text{W}/\text{m}^2\text{K}$ and q is in units of W/m^2

X_1 : mole fraction of the more volatile component NOD: number of data points AVD: average deviation in % ROH: range of heat flux in kW/m^2					
(a) methanol/water					
X_1	C	n	NOD	AVD %	ROH kW/m^2
0	0.540	0.82	13	3.13	33–610
0.1	1.151	0.73	8	2.68	37–510
0.2	0.504	0.79	13	4.58	41–580
0.3	1.341	0.71	8	1.57	37–520
0.4	0.724	0.76	14	2.48	36–510
0.6	0.736	0.75	9	2.10	28–500
0.8	0.530	0.80	23	4.29	21–580
0.9	0.162	0.90	9	1.73	26–500
1.0	0.0876	0.96	21	4.69	25–520
(b) ethanol/water					
X_1	C	n	NOD	AVD %	ROH kW/m^2
0	0.540	0.82	13	3.13	33–610
0.07	2.030	0.70	13	4.68	17–590
0.17	0.947	0.74	9	2.34	40–570
0.20	0.543	0.78	14	3.50	40–550
0.31	0.360	0.81	23	3.26	36–550
0.40	0.460	0.79	12	3.68	30–450
0.55	0.196	0.88	18	3.30	22–600
0.60	0.203	0.87	15	3.39	23–500
0.80	0.350	0.86	15	6.08	24–460
0.88	0.138	0.93	18	1.56	25–460
1.0	0.189	0.90	23	2.89	19–400
(c) methanol/ethanol					
X_1	C	n	NOD	AVD %	ROH kW/m^2
0	0.189	0.90	23	2.89	19–400
0.2	0.112	0.94	25	2.55	23–430
0.4	0.0933	0.95	18	2.55	24–420
0.6	0.0813	0.96	17	2.75	21–400
0.8	0.122	0.93	24	4.01	20–500
1.0	0.0876	0.96	21	4.69	25–520
(d) ethanol/ <i>n</i> -butanol					
X_1	C	n	NOD	AVD %	ROH kW/m^2
0	0.332	0.86	17	2.58	22–330
0.2	0.212	0.87	16	1.55	19–370
0.4	0.151	0.89	15	2.39	25–380
0.6	0.166	0.88	16	2.78	28–390
0.8	0.121	0.91	26	3.45	23–500
0.9	0.199	0.89	7	2.79	15–390
1.0	0.189	0.90	23	2.89	19–400
(e) methanol/benzene					
X_1	C	n	NOD	AVD %	ROH kW/m^2
0	0.398	0.84	31	4.83	18–280
0.1	0.265	0.83	14	3.41	21–350
0.2	0.283	0.83	12	4.54	23–280
0.3	0.262	0.85	13	5.43	20–270
0.4	0.314	0.84	22	5.39	16–280
0.5	0.200	0.88	14	3.89	19–240
0.6	0.0948	0.95	19	1.96	21–280
0.7	0.0924	0.95	15	1.40	23–320
0.8	0.122	0.93	15	2.71	20–280
0.9	0.133	0.93	14	3.39	24–350
1.0	0.0876	0.96	21	4.69	25–520

where the constant, C , and exponent, n , were determined for each concentration by fitting Eq. (1) to the data points mostly lying in the fully developed boiling region. Table 2 compiles the values of C and n together with number of data (NOD), average deviation (AVD) and range of heat flux (ROH). Equation (1) is verified to reproduce well the data within an average deviation of 5% at most. A few data points at lower heat fluxes deviate on the upper side of Eq. (1) since they belong to the partial nucleate boiling region where natural convection remains influential.

4. Heat Transfer Results

4.1 Effect of mixture concentration

Figure 1 shows variations of heat transfer coefficient with mole fraction of the more volatile component for the five mixtures with taking heat flux as a parameter. Data points in the plots are not the measured ones but the calculated from Eq. (1) to make heat flux at the same value. On the top of each figure are shown for reference variations of the boil-

ing range, ΔT_{bp} , defined as the temperature difference between the dew and bubble points and the vapor-liquid concentration difference, $|Y_1 - X_1|$, defined in a mole fraction of the more volatile component.

As having been observed in many previous measurements, heat transfer coefficient is reduced below an interpolated value between their pure components for non-azeotropic mixtures or between one of pure component and an azeotrope for azeotropic mixtures. A distinct minimum in heat transfer coefficient occurs at a certain intermediate concentration of each mixture. Detailed examination of the variations in α , ΔT_{bp} and $|Y_1 - X_1|$ against X_1 indicates that the largest reduction of heat transfer coefficient does not necessarily correspond to the concentration at which either ΔT_{bp} or $|Y_1 - X_1|$ becomes maximal.

4.2 Effect of heat flux

When compared at a same concentration in Fig. 1, heat transfer coefficient is more significantly reduced as heat flux is increased. To make clear this heat flux dependence we normalize heat transfer coefficient as a

Fig. 1 Variation of heat transfer coefficient with mixture composition, along with variations of flash temperature difference, ΔT_{bp} , and composition difference, $|Y_1 - X_1|$

ratio to an ideal coefficient interpolated between pure components. Let ΔT_1 and ΔT_2 the wall superheats for pure components at the same heat flux as the mixture, and α_1 and α_2 the corresponding heat transfer coefficients, then an ideal wall superheat and an ideal heat transfer coefficient are defined respectively as follows.

$$\Delta T_{id} = X_1 \Delta T_1 + (1 - X_1) \Delta T_2 \quad (2)$$

$$\alpha_{id} = \frac{q}{\Delta T_{id}} = \frac{1}{X_1/\alpha_1 + (1 - X_1)/\alpha_2} \quad (3)$$

Here subscripts "1" and "2" refer to the more and the less volatile components respectively. For azeotrope-forming mixtures the above interpolation is done between one of pure component and an azeotrope. Wall superheat in mixture boiling is exceeding the ideal superheat, ΔT_{id} , calculated from Eq. (2), resulting in a reduction of heat transfer coefficient of mixture below the ideal coefficient, α_{id} .

Figure 2 shows the normalized heat transfer coefficients for methanol/water and ethanol/water

Fig. 2 Normalized heat transfer coefficient showing larger reduction at higher heat flux

mixtures. A reduction of α/α_{id} is more significant as heat flux is increased.

5. Development of a New Correlation

5.1 Background of existing correlations

For the reduction of heat transfer coefficient, α , below an ideal coefficient, α_{id} , four mixture effects are proposed as attributable. The first is mass transfer resistance^{(2),(4)} for the more volatile component to be replenished from bulk liquid to the bubble surface. The second is a local temperature rise in the vicinity of the heated surface caused by depletion of the volatile component and a resulting decrease^{(1),(3),(7)} in the effective wall superheat. The third is an increase of energy needed for forming a bubble nucleus of mixture and a resulting decrease^{(3),(6)} in nucleation sites. The fourth is nonlinear variation⁽⁵⁾ of mixture properties with composition. Various correlating parameters are deduced from the above mentioned controlling mechanisms and they are taken into consideration in developing empirical correlations to predict heat transfer reduction.

In a case that mass transfer is a controlling resistance, the concentration difference of the volatile component in vapor and liquid, $|Y_1 - X_1|$, is taken up as a correlating parameter. This parameter was introduced in such correlations as developed by Stephan and Körner⁽³⁾, Calus and Leonidopoulos⁽¹¹⁾, Schlünder⁽¹²⁾, and Jungnickel et al.⁽¹³⁾. Although Stephan and Körner used only this parameter, another parameters were additionally included in other correlations. Calus and Leonidopoulos added two parameters, i.e. Lewis number and the sensible to latent heat ratio, while Schlünder employed mass transfer coefficient and the conversion ratio of the total heat to latent heat of bubbles. Schlünder and Jungnickel et al. included heat flux in their correlations to account for an increased heat transfer reduction with heat flux.

Considering a decrease in effective wall superheat as a main cause of reduced heat transfer, Thome⁽⁷⁾

introduced the flash temperature difference, ΔT_{bp} , defined as the temperature difference between the dew and bubble points, thus determined solely from the phase diagram. His correlation runs as,

$$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + (\Delta T_{bp}/\Delta T_{id})} \quad (4)$$

According to the assessment⁽¹⁰⁾ of the above mentioned five correlations in comparison with the present data, Stephan and Körner correlation predicts rather well the experimental data as naturally expected because it includes an empirical constant accommodated to each mixture. The second-best is Schlünder correlation. However, it includes mass transfer coefficient, information of which is very deficient for most mixtures and its estimation is tedious even when possible. Hence the mass transfer coefficient in Schlünder correlation is regarded as an empirical constant adjustable to each mixture. Thome correlation is in a simplest form while fails to correlate the data. As shown in the following and hinted by Thome, his correlation is of limited use in higher heat flux region close to the critical heat flux.

5.2 Effective temperature difference

When binary mixture is in boiling, the more volatile component is preferentially evaporated into bubbles. Thus its concentration, $X_{1,p}$, at the vapor-liquid interface becomes lower than the bulk liquid concentration, X_1 , as shown in Fig. 3. This results in a rise of bubble point temperature, T_h , above the bulk temperature, T_s . As a consequence, the available temperature difference for evaporation is reduced from the apparent wall superheat of ΔT by an amount of ΔT_h . This local temperature rise ΔT_h , should be dependent on boiling intensity or vapor generation rate, thus be a function of heat flux. It will remain nearly zero just after the boiling inception because the rate of evaporation is very low and will increase to its highest value at the critical heat flux point. Thus the maximum local temperature rise is limited at most by the flash temperature difference, ΔT_{bp} , shown in

Fig. 3 Temperature rise at the vapor-liquid interface of bubbles on a phase diagram

Fig. 3. Consequently,

$$\Delta T_h = F(\text{heat flux}) \times \Delta T_{bp} \quad (5)$$

where F is a function of heat flux and should asymptotically satisfy the following conditions at the both ends of nucleate boiling region.

$$F \rightarrow 0 \text{ at } q \rightarrow q_{INB} \text{ (the boiling inception)}$$

$$F \rightarrow 1 \text{ at } q \rightarrow q_{CHF} \text{ (the critical heat flux)}$$

The bubble surface temperature is high above the saturation temperature of bulk liquid, T_s , by an amount of ΔT_h . Hence the effective superheat is reduced by ΔT_h from the apparent wall superheat, ΔT , defined as the heated wall temperature minus T_s . It is assumed here that the ideal wall superheat given by Eq.(2) is identical to the effective temperature difference. Then

$$\Delta T_{id} = \Delta T - \Delta T_h \quad (6)$$

$$\therefore \frac{\alpha}{\alpha_{id}} = \frac{q/\Delta T}{q/\Delta T_{id}} = \frac{1}{1 + \Delta T_h/\Delta T_{id}}$$

Equation (5) is substituted into the above equation to give

$$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + F \times (\Delta T_{bp}/\Delta T_{id})} \quad (7)$$

Thome correlation (4) is a special case that $F=1$ in Eq.(7). As noted by Thome⁽⁷⁾, an assumption of $F=1$ holds true only at higher heat fluxes close to the critical flux.

5.3 Derivation of a new correlation

According to Thome assumption that heat transfer reduction of mixtures is attributable to a local temperature rise around growing bubbles or in liquid close to the heated surface, it is very important to determine F in Eqs.(5) and (7) as a function of heat flux. Inoue and Monde⁽¹⁵⁾ proposed $F=0.25+0.45q \times 10^{-5}$, while Fujita and Tsutsui⁽¹⁴⁾ offered $F=1-0.8 \exp(-q \times 10^{-5})$, respectively with use of the unit W/m^2 for heat flux. These formulas include dimensional constants and fail to satisfy the limiting condition that $F \rightarrow 1$ as $q \rightarrow q_{CHF}$. Hence their validity to a wide range of heat flux remains uncertain.

Nucleate boiling is prevailing in a wide range of

heat flux from the inception value, q_{INC} , to the critical heat flux, q_{CHF} . The inception heat flux is usually less than 1% of the critical heat flux. Evaporation rate at such low heat fluxes is not so intensive that the reduction of heat transfer coefficient is not so significant as compared with that in the fully developed boiling at higher heat fluxes. Hence it is reasonable to pursue a functional relationship of F in Eq.(5) in terms of heat flux ratio, q/q_{CHF} , normalized by the critical heat flux. Then a more general formula is expected to be found.

As for the critical heat flux of single components the relation, $q_{CHF} = K \rho_v h_{fg} [\sigma g (\rho_L - \rho_v) / \rho_v^2]^{1/4}$, is well established. Assuming the similar relation holds true for mixtures, a functional relationship of F against heat flux is to seek in terms of the following dimensionless heat flux normalized in reference to the critical heat flux.

$$F = F \left\{ \frac{q}{\rho_v h_{fg} [\sigma g (\rho_L - \rho_v) / \rho_v^2]^{1/4}} \right\} \quad (8)$$

where h_{fg} is the latent heat of evaporation, g the gravitational acceleration, ρ_L and ρ_v the liquid and vapor density, and σ the surface tension.

Equations (3), (7) and (8) are fitted to the heat transfer coefficients measured for the five mixtures and compiled in Table 2 to obtain an empirical correlation. Physical properties of mixtures needed in Eq.(8) were calculated as follows. The liquid density was obtained from Yen and Woods correlation⁽¹⁶⁾ that is based on the corresponding state principle. The vapor density was calculated assuming each component of the vapor mixture as ideal gas. The latent heat of evaporation was determined by applying the mixing rule on mass base to pure components. The surface tension of aqueous mixtures was obtained using the Tamura equation⁽¹⁷⁾, while that of organic mixtures was calculated using the Macleod and Sugden equation^{(18),(19)} that is based on the parachor. Data fitting finally yields an empirical equation to predict the reduction of heat transfer coefficient in mixtures as,

$$\frac{\alpha}{\alpha_{id}} = \frac{1}{1 + \left[1 - \exp \left\{ - \frac{60q}{\rho_v h_{fg} [\sigma g (\rho_L - \rho_v) / \rho_v^2]^{1/4}} \right\} \right] (\Delta T_{bp}/\Delta T_{id})} \quad (9)$$

$$\Delta T_{id} = X_1 \Delta T_1 + (1 - X_1) \Delta T_2$$

$$\alpha_{id} = \frac{q}{\Delta T_{id}} = \frac{1}{X_1/\alpha_1 + (1 - X_1)/\alpha_2}$$

If we use the value $K=0.13$ for CHF of pure components, F given by Eq.(9) varies from 0.075 to 1.0 against the variation of heat flux from 1% of q_{CHF} to q_{CHF} . Thus, Eq.(9) approximately satisfies the

limiting conditions imposed on a liquid temperature rise of Eq.(5).

Figure 4 shows a comparison of the measured data with the predictions from the above correlation. Heat transfer coefficients calculated from Eq.(1) for pure components and azeotrope are used in evaluating α_{id} and ΔT_{id} . All measured data including those in the partial boiling at low heat flux were used in the comparison, naturally except for the data for pure components and azeotropes. The data that are correlated within $\pm 20\%$ accuracy lines are 96.4% for methanol/water, 84.9% for ethanol/water and 95.0% for ethanol/butanol. Although not shown here, all the data of methanol/ethanol fall within $\pm 15\%$ lines and 77.3% data of methanol/benzene are correlated within $\pm 20\%$. A fairly uniform distribution of the data points as seen in Fig. 4 indicates that the new correlation reasonably predicts heat transfer coefficients for a wide range of heat flux as well as the whole range of mixture concentration.

5.4 Use of correlation for pure components

In using Eqs.(4) and (9), heat transfer coefficients of pure components and azeotropes are needed as the anchoring data to evaluate the ideal coefficient, α_{id} . Hence the accuracy of predicted coefficients for a mixture may be greatly dependent on how to estimate such anchoring data for the mixture. Figure 5 shows a similar comparison as in Fig. 4 when Stephan and Abdelsalam correlation⁽²⁰⁾ is employed for calculating α_{id} . Contrary to expectation the accuracy is not so degraded in Fig. 5. In particular the data gathering for each mixture is at much the same degree as in the case of Fig. 4. Thus Eq.(9) is verified to well account for the mixture effect on the reduction of heat transfer coefficients in mixtures.

6. Conclusions

Heat transfer coefficients in nucleate boiling of five mixtures saturated at atmospheric pressure were measured on a flat plate of 40 mm in diameter. Data

Fig. 4 Comparison of measured and predicted heat transfer coefficients when the measured coefficients for pure components and azeotropes are used as anchoring data

Fig. 5 Comparison of measured and predicted heat transfer coefficients when Stephan and Abdelsalam correlation is used to estimate the anchoring data.

show reduction of heat transfer coefficients below interpolated values between pure components. Predictive methods of the reduction is discussed and the following conclusions are reached.

(1) Taking into account a decrease in effective temperature difference due to liquid temperature rise around bubbles, an empirical correlation (9) was obtained to predict heat transfer reduction in boiling of mixtures.

(2) In developing the correlation, a liquid temperature rise is assumed to change between nearly zero at the boiling inception and the maximum at the critical heat flux point that is limited by the flash temperature difference. Thus the obtained correlation is a modification of Thome correlation with introduction of the effect of heat flux, resulting in covering a wide range of nucleate boiling.

(3) The correlating equation (9) succeeds to reproduce the present data within $\pm 20\%$ accuracy in a wide range of heat flux from about 15% of the critical heat flux to almost the critical flux.

(4) Even when Stephan and Abdelsalam correlation is used to evaluate necessary heat transfer coefficients of pure components or azeotropes, prediction accuracy of the correlation (9) is not degraded.

As a future work, more extensive examination of the newly developed correlation remains to verify its accuracy and validity.

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