

Nucleation Site Density in Pool Boiling of Binary Mixtures: Effect of Surface Micro-roughness and Surface and Liquid Physical Properties

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The nucleation site density (N/A) during pool boiling of binary mixtures has been experimentally examined in terms of the heating surface physical properties, the liquid physical properties, the vapour liquid equilibria and the surface micro-roughness. The liquid mixtures used were acetone – carbon tetrachloride and *n*-hexane – carbon tetrachloride. A parameter R_a , to characterize the micro-roughness of the heating surface was defined as the average value of the peaks and valleys on the surface. The study shows that while at low R_a values the physical properties of the liquid and the binary composition are not as important as the surface roughness; at higher R_a values the liquid properties and composition are also important. A correlation based on the present data has been proposed.

La densité de sites de nucléation (N/A) lors de l'ébullition de mélanges binaires a été étudiée expérimentalement en termes de propriétés physiques de surface de chauffe, de propriétés physiques liquides, d'équilibres liquide-vapeur et de micro-rugosité de surface. On a utilisé comme mélanges liquides un mélange d'acétone et tétrachlorure de carbone et un mélange de *n*-hexane et tétrachlorure de carbone. Un paramètre R_a pour caractériser la micro-rugosité de la surface de chauffe a été défini comme étant la valeur moyenne des sommets et vallées sur la surface. L'étude montre que si à de faibles valeurs de R_a les propriétés du liquide et de la composition binaire ne sont pas aussi importantes que la rugosité de la surface, à de fortes valeurs de R_a les propriétés du liquide et la composition sont aussi importants. On propose une corrélation s'appuyant sur ces données.

Keywords: surface micro-roughness, nucleation site density, binary mixtures, diffusion, pool boiling.

Nucleate boiling, by definition, is characterized by the formation of vapour bubbles at certain preferred locations known as "nucleation sites", when the heating surface is maintained at a temperature above the saturation temperature of the liquid with which it is in contact. This "nucleation site density", the number of such sites per unit area, is one of the key parameters in nucleate boiling. The boiling heat flux depends to a large extent on the nucleation site density in nucleate boiling. It is therefore not surprising to find that over the years a large amount of work has been directed towards increasing the number of such sites by polishing, etching, sintering and using coatings of various types on the heating surface, the primary purpose being to increase the heat flux for a given wall superheat (excess temperature). Moreover, it is generally accepted that the nucleation site density depends not only on the physical properties and the finish (micro-roughness) of the heating surface, but also the liquid physical properties and the wall superheat.

Nucleate pool boiling of binary mixtures, when compared to pure liquids, is characterized by a reduction in the nucleation site density (N/A), the bubble departure diameter and the heat flux. This is because: (a) there is a preferential evaporation of the more volatile component, (b) the mass diffusion of the more volatile component to the microlayer (the liquid layer trapped under a growing bubble) is much slower than the rate of bubble growth, (c) there is often a non-linear variation in the mixture physical properties with composition and (d) the effect of composition on bubble nucleation itself. In most of the earlier work on binary mixtures, the investigators have correlated the heat transfer coefficient during the pool boiling of a binary mixture in

terms of an 'ideal' heat transfer coefficient and a 'reduction factor'. This 'reduction factor' in general, is a function of the physical properties of the two liquids used, the vapour-liquid equilibria and the slope of the vapour pressure curve. This 'reduction factor' causes slower bubble growth rates, smaller bubble diameters and higher bubble frequencies. Much of this work has been summarized by Thome and Shock (1984), Kadhun et al. (1994) and Celata et al. (1994). The present study focuses on an experimental investigation on the nucleation site density in the pool boiling of binary mixtures.

One of the best known correlations for nucleate pool boiling heat transfer of pure liquids, proposed by Rohsenow (1952), related the heat flux with excess temperature, liquid properties and a parameter C_{sf} . The parameter C_{sf} is a constant whose values for pure liquids are tabulated for various surface-liquid combinations using experimental data. The parameter C_{sf} apparently takes into account the contact angle, the surface micro-roughness and their interaction in determining the nucleation site density. Vachon et al. (1968a) analyzed Rohsenow's (1952) correlation using literature boiling data and concluded that the boiling heat flux for a given excess temperature is dependent on the surface preparation (surface micro-roughness) as well as the surface – liquid combination. A number of experimental studies and analyses have been reported in the literature on the nucleation site density in the pool boiling of pure liquids. Most of these pertain only to specific heating surfaces and liquids and therefore the conclusions of these studies cannot be generalised. Yamagata et al. (1955), related the nucleation site density to the heat flux and the excess temperature for water boiling on a horizontal brass surface. Kurihara and Myers (1960) found that the nucleation site density increased with increase in surface micro-roughness for a given wall superheat. Their data matched the correlation proposed by Yamagata et al.

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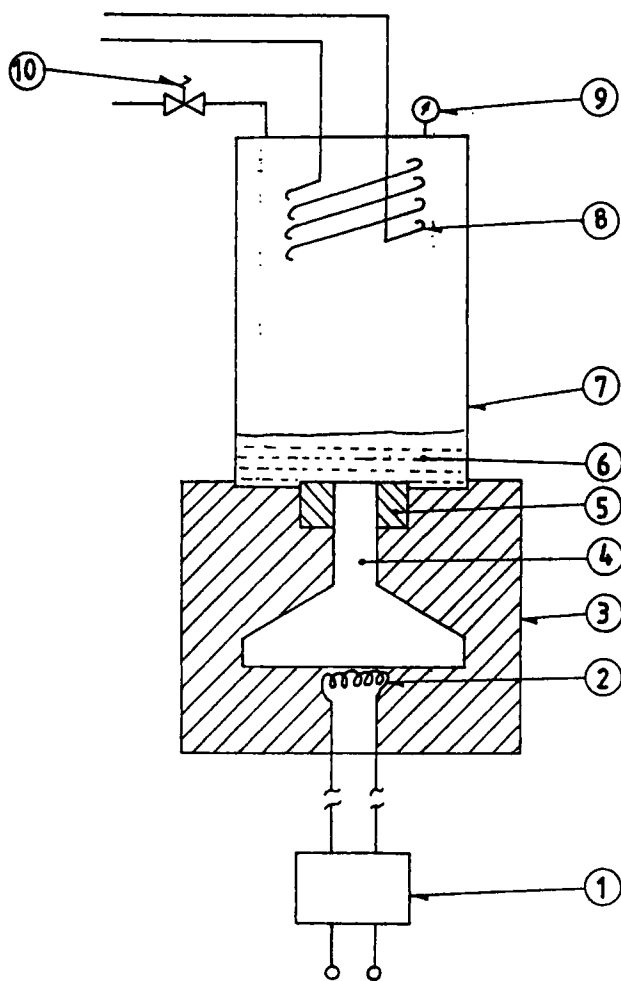


Figure 1 — Schematic of experimental set-up: 1. Dimmerstat, 2. Plate heater, 3. Insulation, 4. Heating block, 5. Teflon hollow cylinder, 6. Liquid, 7. Double walled glass vessel, 8. Cooling coil, 9. Pressure gauge, 10. Relief valve.

(1955), Gaertner and Westwater (1960) and Rallis and Jawurek (1964) found that the heat flux was approximately proportional to the number of nucleation sites. Gaertner (1963) used the experimental data of Gaertner and Westwater (1960) and found that the spatial distribution of the nucleation sites obeyed the Poisson distribution. This was later confirmed by Sultan and Judd (1978) and by Wang and Dhir (1993). Gaertner (1965) obtained nucleation site density using water on a polished copper surface and related the nucleation site density to the wall temperature and liquid properties. Brown (1967), Mikic and Rohsenow (1969) and Bier et al. (1978) related the nucleation site density to the corresponding cavity mouth radius using a power law relationship. Cornwell and Brown (1978) correlated the nucleation site density to the wall superheat. Kocamustafaogullari and Ishii (1983) proposed a correlation for the nucleation site density in terms of the heat transfer coefficient, the liquid properties, the bubble departure diameter and the wall superheat using the data of Kurihara and Myers (1960) and Gaertner and Westwater (1960). Yang and Kim (1988) developed an analytical expression for the nucleation site density as a product of two probability density functions, one each for the cavity mouth radius and the cone angle. The correlation compared well with their own data of water boiling on a polished stainless steel surface.

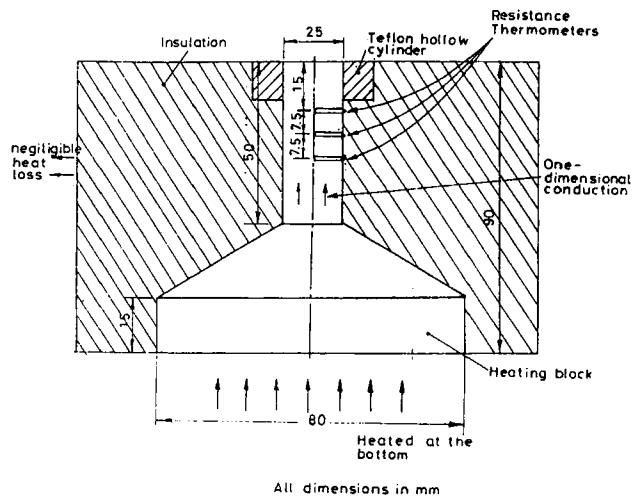


Figure 2 — Dimensions of the heating block.

Barthau (1992) obtained nucleation site density data of R-114 on a horizontal polished copper tube and found that the heat transfer contribution of an individual nucleation site decreased with increase in the pressure or the heat flux. Wang and Dhir (1993) conducted experiments using distilled water on a polished copper surface with the surface at three different angles from the horizontal. They found that an increase in the wettability decreased the fraction of the cavities that nucleate. They proposed a correlation for the nucleation site density in terms of the cavity mouth diameter and the contact angle. Kolev (1995) analyzed literature water boiling data obtained on different surfaces and surface finishes. He concluded that contact angle was perhaps the single most important factor in determining the nucleation site density.

While a large number of studies have been reported on the nucleation site density in pool boiling of pure liquids as can be seen from the review above, there are very few studies on the nucleation site density in the pool boiling of binary mixtures. Van Stralen and Sluyter (1969) experimentally measured the nucleation site density for water–methyl ethyl ketone and water–*n*-pentanol mixtures on a horizontal platinum wire. The nucleation site density for the binary mixtures at any composition was found to be always lower than for the pure liquids at any heat flux or excess temperature. Eddington and Kenning (1979) studied the effect of contact angle on the nucleation site density for ethanol–water mixtures. Two heating surfaces, specifically brass and copper were used. Their investigation showed that the contact angle increases as ethanol composition in the mixture decreases and the larger contact angles lead to large nucleation site densities. Hui (1983) and Hui and Thome (1985) measured the nucleation site density for two azeotropic mixtures: ethanol–water and ethanol–benzene at atmospheric pressure using a vertical brass disc as the heating surface. The number of nucleation sites was determined in an inscribed 19 mm diameter circle photographically. Their results show that the nucleation site density increases with ethanol composition in the ethanol–water mixture while for the ethanol–benzene mixture a maximum and a minimum in the nucleation site density was observed to the left and right respectively of the azeotropic composition. This non-linear variation was attributed to the ‘vapour spreading phenomenon’ observed by Thome et al. (1982).

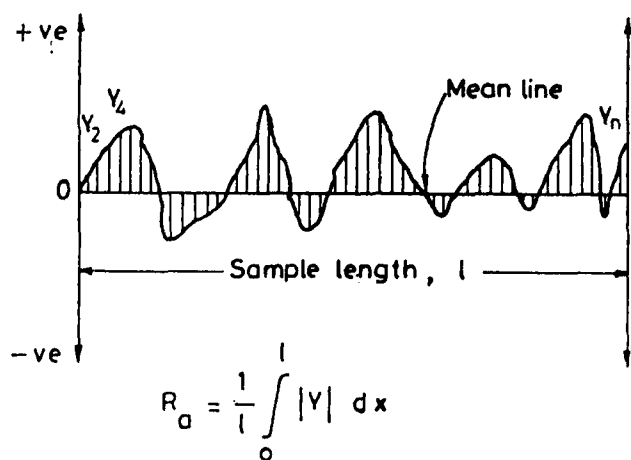


Figure 3 — Definition of R_a and the instantaneous output of the profilometer.

Although considerable analyses of the experimental data have been reported by the previous investigators, no general correlation exists to estimate the nucleation site density in the pool boiling of binary mixtures in terms of the surface micro-roughness, physical properties and vapour-liquid equilibria data of the binary mixtures. Further, the effect of surface micro-roughness on the bubble nucleation characteristics of binary mixtures is not well established. In the present study, the dependence of nucleation site density on the liquid physical properties and mass diffusional effects and on the metrological and thermo-physical properties of the heating surface has been experimentally examined.

Experimental

A schematic of the experimental set-up used in the present study is shown in Figure 1. Two different heating sur-

faces, namely aluminium and stainless steel were used in the present study. The dimensions of the heating block are shown in Figure 2. The test section consisted of a double walled glass cylinder with the annular space under vacuum. The glass vessel is held tightly between two stainless steel slabs. The bottom slab holds the heating surface (which is a 25 mm circular surface) through a Teflon annular cylinder. Therefore, there is no direct contact between the heating block and the bottom slab. The block is heated electrically. The current supplied and the voltage across are adjusted using a dimmerstat connected to a power source. Temperatures are measured at three points along the axis of the block using platinum resistance thermometers. Using these, the surface temperature was evaluated by extrapolation. The temperature of the liquid was also measured using a platinum resistance thermometer. The thermometer consists of a long cylinder at the end of which is a ceramic bulb that houses the platinum sensor. The micro-roughness of the heating surface was quantitatively defined using the centre line average, R_a . The centre line average R_a is defined as the average of the absolute values of the peaks and the valleys on the surface (see Figure 3). The R_a value has been extensively used in boiling literature to characterize micro-roughness. It is also a standard for measurement of surface roughness in metrology practice as per ISO 4287/1:1984. Before each run, the surface of the block was polished using a particular grade of emery paper and the micro-roughness, R_a measured using a profilometer called the Perthometer. The R_a values obtained after polishing the heating surface with different grades of emery paper are summarized in Table 1. The binary mixtures used were acetone-carbon tetrachloride and *n*-hexane-carbon tetrachloride. The compositions chosen and the physical properties of the binary mixtures are given in Table 2. These mixtures are readily miscible with each other, do not form azeotropes and there is a significant difference in the boiling points of the pure liquids. All the experiments were carried out at atmospheric pressure and

TABLE 1
Summary of Surface Characteristics and Binary Systems Used in the Present Study

Surface material	Surface characteristics	Binary system
Stainless steel	$R_a = 0.2 \mu\text{m}$ (polished with 1/0 grade emery paper)	acetone-carbon tetrachloride (53.5, 22.9 and 8.3 mass% acetone)
Aluminium	$R_a = 0.52 \mu\text{m}$ (polished with 4/0 grade emery paper)	
	$R_a = 0.89 \mu\text{m}$ (polished with 3/0 grade emery paper)	
	$R_a = 1.17 \mu\text{m}$ (polished with 2/0 grade emery paper)	
Stainless steel	$R_a = 0.2 \mu\text{m}$ (polished with 1/0 grade emery paper)	<i>n</i> -hexane-carbon tetrachloride (67.5, 29.35 and 7.67 mass% <i>n</i> -hexane)
Aluminium	$R_a = 0.52 \mu\text{m}$ (polished with 4/0 grade emery paper)	
	$R_a = 0.89 \mu\text{m}$ (polished with 3/0 grade emery paper)	
	$R_a = 1.17 \mu\text{m}$ (polished with 2/0 grade emery paper)	

TABLE 2
Physical Properties of Binary Mixtures Used in Present Study

x	y	ρ_l kg/m ³	λ_x 10 ⁻³ J/kg	C_p J/kg.K	μ_x 10 ³ kg/m.s	k W/m.K	σ_x 10 ³ N/m	α_x 10 ⁸ m ² /s	D_{AB} x10 ⁹ m ² /s
acetone-carbon tetrachloride system									
0.534	0.600	980	293	1632	0.403	0.120	20.33	7.50	3.2
0.229	0.373	1225	226	1225	0.511	0.107	21.17	7.12	2.37
0.083	0.221	1392	203	1035	0.546	0.100	21.70	6.93	1.68
<i>n</i> -hexane-carbon tetrachloride system									
0.675	0.720	761	291	1950	0.302	0.100	14.67	6.74	3.16
0.293	0.353	1046	234	1373	0.422	0.096	16.6	6.78	2.37
0.076	0.114	1327	199	1041	0.491	0.095	18.79	6.90	1.75

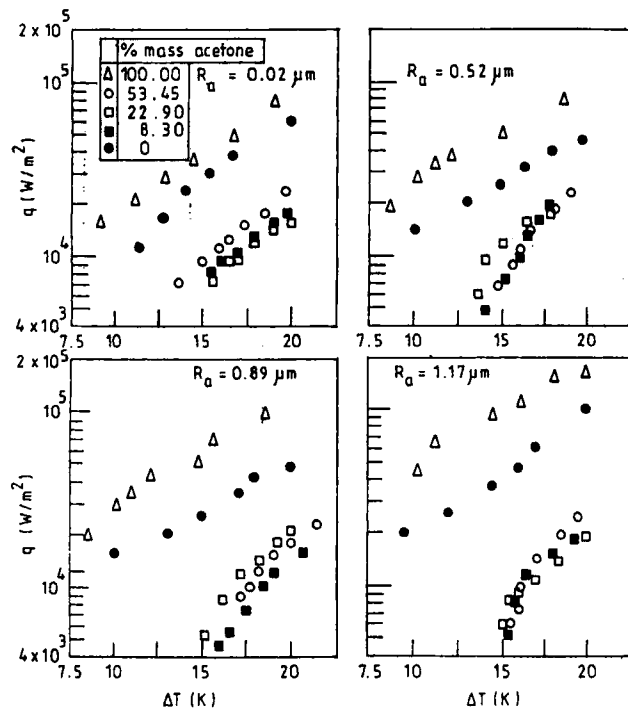


Figure 4—Heat flux versus excess temperature—acetone-carbon tetrachloride system.

standard gravity conditions. About 150 mL of the mixture was degassed and boiled in each run. The voltage across the heater was set using the dimmerstat. The temperatures along the axis of the heating block, the liquid temperature, the voltage and current were measured. A steady state was assumed to exist if the temperatures do not change by more than 0.1 K in two minutes. After nucleate boiling begins, the nucleation site density was measured by photographing the heating surface using a high speed still camera. The flash duration was 1/25,000 s. At each heat flux three photographs were taken to ensure both transient and permanent nucleation sites were accounted for. This procedure was repeated for the other surface finishes and repeated again with the other heating block (surface). From the photographs the number of nucleation sites was obtained by visual counting following the technique of Hui and Thome (1985). The uncertainty in counting the bubbles using this technique is $\pm 10\%$.

Results

The variation of heat flux with excess temperature for the four surface finishes for each binary composition of the two

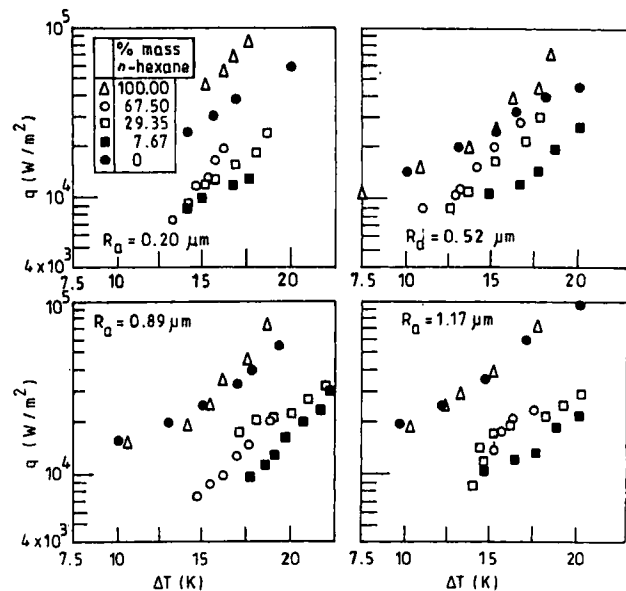


Figure 5—Heat flux versus excess temperature—*n*-hexane-carbon tetrachloride system.

systems is shown in Figures 4 and 5. It can be seen that the heat flux, for a given excess temperature and a particular binary mixture, increases, then decreases and increases again with increasing R_a . A similar trend is found when the nucleation site density is plotted against the heat flux (Figures 6 and 7). At low R_a values, the nucleation site density is very sensitive to the heat flux. On the other hand, at higher R_a values the nucleation site density is less sensitive to the heat flux but the activation of the sites occurs at lower heat fluxes.

CORRELATION OF THE DATA

From the experimental investigations, analysis of previous investigations and the data obtained in the present study, it is seen that the nucleation site density is a function of the heating surface physical properties, its micro-roughness, the liquid physical properties, the vapour liquid equilibria and the wall superheat. Benjamin (1996) studied the pool boiling phenomena of various pure liquids and found that the nucleation site density (and the heat flux) for a given liquid and excess temperature, decreases and then increases with surface micro-roughness. A similar trend is discernible in the data of Gilmour (1958), Berenson (1962), Vachon et al. (1968b), Raad and Myers (1971) and Nishio and Chandratilleke (1989). To account for the decreasing-

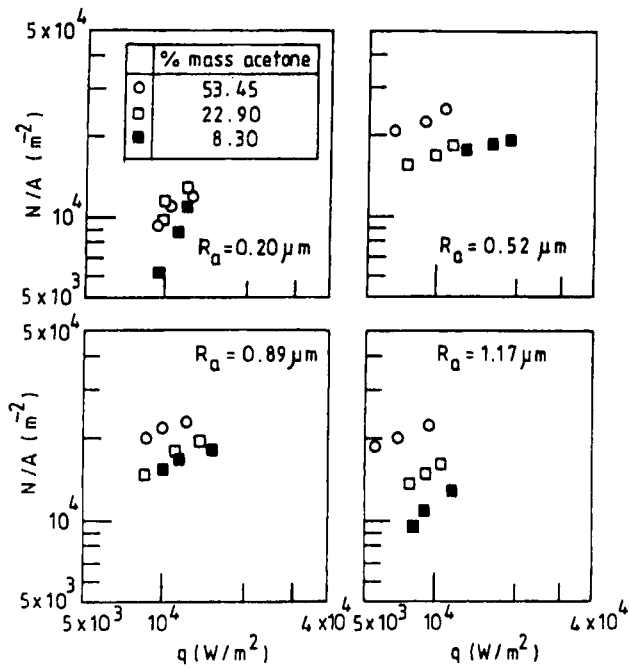


Figure 6 — Heat flux versus (N/A) — acetone-carbon tetrachloride system.

increasing trend of the nucleation site density with surface micro-roughness, Benjamin (1996) defined a dimensionless surface roughness parameter θ as:

$$\theta = A + B \left(\frac{R_a P}{\sigma} \right) + C \left(\frac{R_a P}{\sigma} \right)^2 \dots \dots \dots (1)$$

A, B and C are constants that have to be evaluated using experimental data. For pure liquids these were evaluated as 14.5, -4.5 and 0.4 respectively by Benjamin (1996). In contrast to pure liquids, the nucleation site density data obtained in the present study shows an increase, decrease and then a slight increase with R_a . While this is the exact opposite of the behaviour of pure liquids, a similar polynomial is required to describe the trend. The same polynomial as in Equation (1) is used so that $1/\theta$ satisfies the trend obtained for binary mixtures. The surface tension used is that of the mixture.

Further, because of the preferential evaporation of the more volatile component during the boiling of a binary mixture, there is mass diffusion of the more volatile component from the bulk towards the heating surface. Hence, a part of the temperature driving force is utilised in overcoming the resistance caused by this mass diffusion. In the present study, the actual and apparent temperature driving forces are related as:

$$\Delta T = (T_w - T_s) \left(1 - \left[|y-x| \sqrt{D_{AB}/\alpha} \right]^{0.5} \right) \dots \dots \dots (2)$$

In Equation (2), $|y-x|$ is the driving force for mass diffusion. Since mass diffusion is the slower (and hence the rate controlling) process, $(D_{AB}/\alpha_{mix})^{0.5}$ is a measure of the resistance to heat transfer. Equation (2) therefore quantifies the actual driving force in the boiling of binary mixtures. Similar expressions have been developed (or used) by many investigators in the literature and these have been summarized by

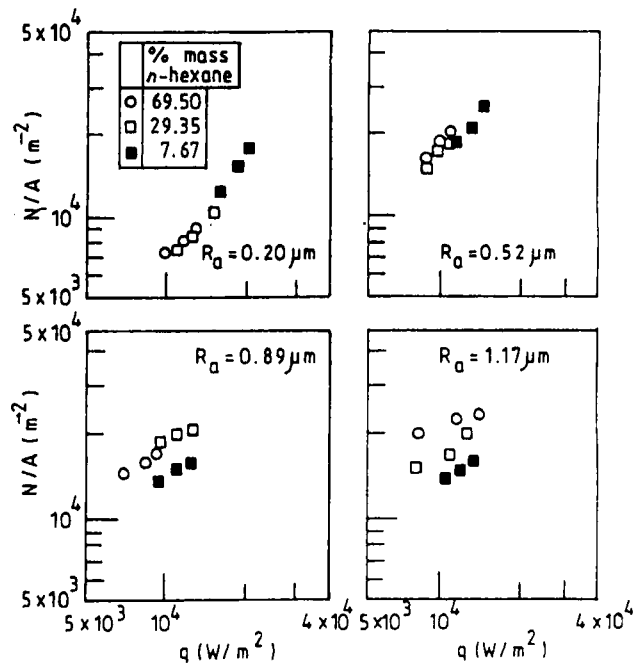


Figure 7 — Heat flux versus (N/A) — n-hexane-carbon tetrachloride system.

Thome and Shock (1984) and Celata et al. (1994) among others.

The nucleation site density in the present study is correlated using the following parameters: a surface-liquid interaction parameter γ (which is the ratio of the physical properties of the heating surface to the liquid), the dimensionless surface roughness θ (as defined by Equation (1)), the Prandtl number of the liquid mixture, a properties-profiles parameter ξ (which accounts for the variation of the physical properties of the binary mixture and the heating surface profile) and the apparent wall superheat. By regression the following equation is obtained:

$$\frac{N}{A} = 5 \times 10^{-5} (\xi)^{-0.25} (Pr)^{1.63} (\gamma)(\theta)^{0.4} (\Delta T)^3 \dots \dots \dots (3)$$

where:

$$\xi = \frac{\rho_l R_a^2 g}{\sigma_l} \dots \dots \dots (4)$$

$$Pr = \frac{C_{pl} \mu_l}{k_l} \dots \dots \dots (5)$$

$$\gamma = \left(\frac{k_w \rho_w C_{pw}}{k_l \rho_l C_{pl}} \right)^{0.5} \dots \dots \dots (6)$$

$$\theta = 14.5 - 4.5 \left(\frac{R_a P}{\sigma_l} \right) + 0.4 \left(\frac{R_a P}{\sigma_l} \right)^2 \dots \dots \dots (7)$$

All the liquid physical properties are of the binary mixture at the composition under consideration. The proposed nucleation site density correlation fits the present data within ± 30

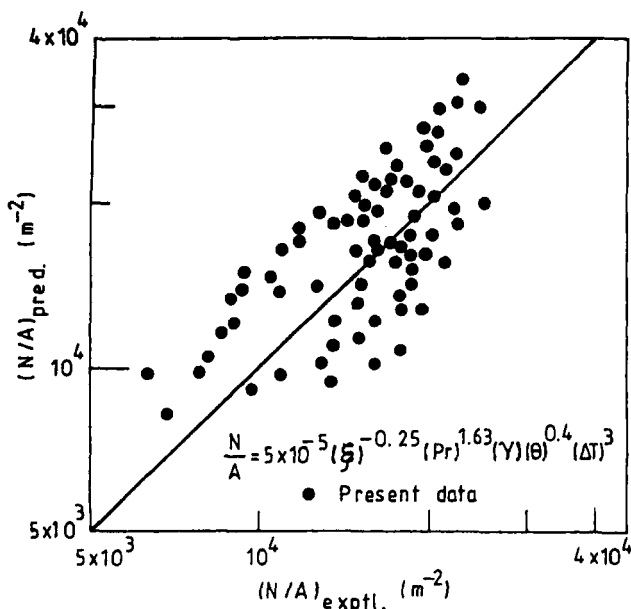


Figure 8 — Performance of the correlation.

%, has a standard deviation of 27.8% and is shown in Figure 8. A summary of the properties of the heating surface and the binary liquids used to obtain the correlation is given in Table 2. The nucleation site density data of binary mixtures reported in the literature were either obtained on thin wires (Van Stralen, 1959 and Van Stralen and Sluyter, 1969) or were reported as a function of cavity mouth radius and mass fractions (Eddington and Kenning, 1979 and Hui and Thome, 1985). It was therefore not possible to use the literature data to further validate the correlation. The range of parameters used in developing the correlation are:

$$\begin{aligned}
 5.3 < Pr < 6.1 & \quad 21.5 < \gamma < 61.6 \\
 0.19 \mu\text{m} < R_a < 1.2 \mu\text{m} \\
 14.6 \times 10^{-3} \text{ N/m} < \sigma_1 < 21.8 \times 10^{-3} \text{ N/m} & \quad 1.8 < \theta < 10.6 \\
 5 \text{ K} < T_w - T_s < 25 \text{ K}
 \end{aligned}$$

Discussion

Webb (1994) defined a necessary condition for boiling nucleation of a pure liquid on a surface as:

$$T_w - T_s = \frac{qr_c}{k_l} - \frac{2\sigma_l}{mr_c} \dots \dots \dots (8)$$

where r_c is the cavity radius and m is the slope of the vapour pressure curve. In Equation (8), r_c can be assumed to be R_a . As a binary liquid vaporises on a hot surface, there is mass transfer of the more volatile component to the liquid layer under the bubble, since the liquid layer is depleted of the more volatile component due to bubble growth. Therefore an additional term which incorporates this mass transfer effect has to be included in Equation (8) for binary mixtures:

$$T_w - T_s = \frac{qR_a}{k_l} + \frac{2\sigma_l}{mR_a} - \frac{nN_A R_a}{D_{AB}} \dots \dots \dots (9)$$

The additional term is obtained from Fick's law of diffusion in the same way as the first term in Webb's (1994) equation

(Equation (8)) is obtained from the Fourier's law of heat conduction. Here n is the slope of the bubble point curve, D_{AB} is the mass diffusivity and N_A is the mass flux of the more volatile component to the microlayer. The mass transfer term is negative because it reduces the temperature driving force.

The data obtained in the present study are shown in Figures 4 and 5 as a plot of the heat flux against the wall superheat for the two binary systems studied (including the respective pure liquid constituents) at different R_a values. From Figures 4 and 5 it can be seen that at the lower R_a values used, i.e. $0.2 \mu\text{m}$ and $0.52 \mu\text{m}$, all the data points are clustered together irrespective of the composition. This indicates that at low R_a values the nucleation site density is not determined by the physical and transport properties of the liquid or by the vapour liquid equilibrium. In other words, at low R_a values a binary mixture behaves as a single component liquid. On the other hand at larger R_a values the data are far apart.

The experimental results of Zeugin et al. (1975) show that the microlayer does not entirely evaporate in the boiling of binary mixtures. This is due to the rise in the boiling point of the microlayer because of the preferential evaporation of the more volatile component. At low R_a values (small cavities) the amount of unevaporated microlayer (which is depleted of the more volatile component) left behind in the cavities along with the trapped vapour (which is essential for nucleation to occur again) is negligible. Therefore, after bubble departure when fresh liquid fills the cavities for nucleation to occur again, the composition difference between the liquid in the cavities and the bulk liquid is also negligible. Hence, mass diffusion of the more volatile component to the cavities is negligible and Equation (9) reduces to that of a pure liquid (Equation (8)). A binary mixture therefore behaves like a pure liquid at low R_a values and the bubble growth is heat transfer controlled. However, in the case of pure liquids the heat flux is of the order of 10^5 W/m^2 , while for binary mixtures it is an order of magnitude less. Therefore as R_a increases, the magnitude of the increase of the first term is significant in comparison to the decrease of the second term, unlike in pure liquids. Therefore, the nucleation site density and the heat flux increases with R_a till N_A becomes significant. This can be seen in Figure 9 which shows the raw experimental data as also in Figure 10 which shows the same trends. The curves in Figure 10 are generated from the correlation (Equation (3) for the case of binary mixtures) and from the correlation proposed earlier by Benjamin and Balakrishnan (1997).

Once the amount of unevaporated microlayer left behind in the cavities becomes sufficiently large in quantity, even after fresh liquid fills the cavities (for nucleation to occur again), the concentration difference between the liquid in the cavities and the bulk is no longer negligible. Hence, there is increased mass diffusion of the more volatile component to the cavities (significant N_A) and the temperature driving force (Equation (9)) is reduced. Furthermore, as R_a increases, $2\sigma_l/R_a$ becomes less significant. The increased mass diffusion to the nucleation sites and the reduction in $2\sigma_l/R_a$ reduces the temperature driving force which in turn gives rise to lower nucleation site densities. Therefore, after reaching a maximum, the nucleation site density decreases with increasing surface micro-roughness. The greater the amount of the liquid left behind in the cavities, the greater will be the mass diffusion flux (N_A) (see Figure 10).

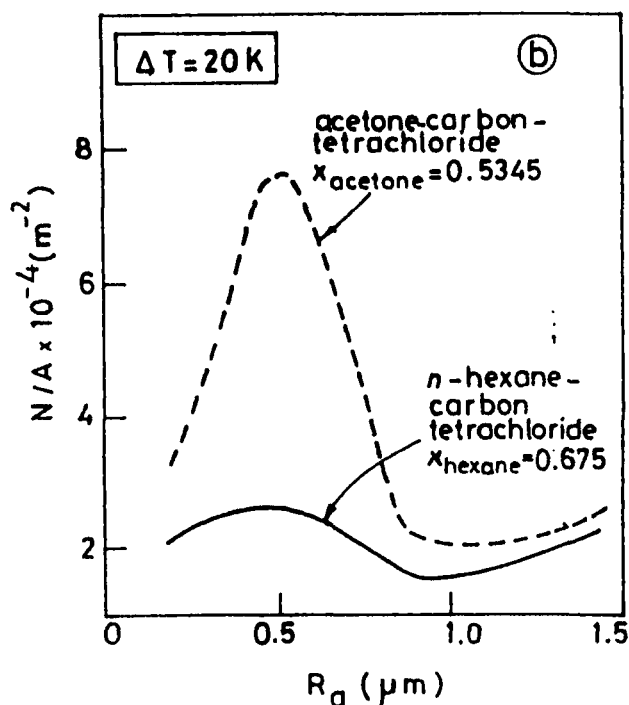
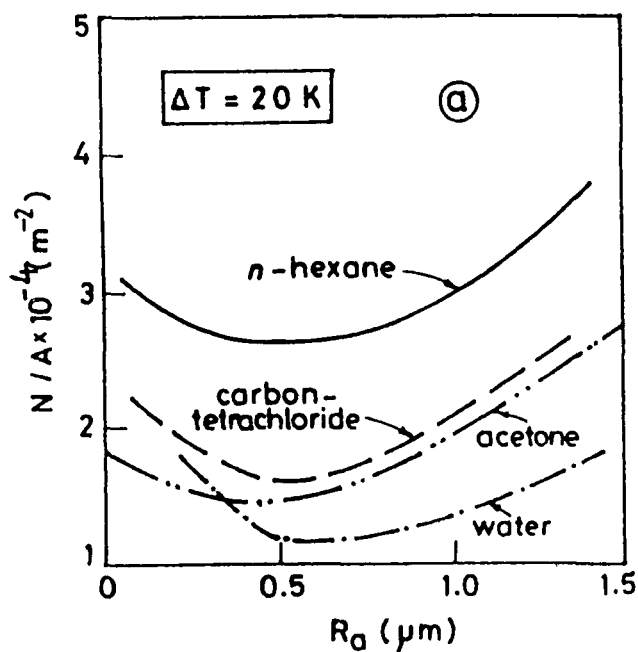


Figure 9 — (N/A) versus R_a — curves generated from correlation: (a) pure liquids; (b) binary mixtures.

The analysis of Sterling and Tichacek (1961) support this hypothesis. Sterling and Tichacek (1961) suggested that the decrease in the rate of nucleation (as well as the nucleation sites) was caused by the clogging of the cavities by the less volatile component due to the preferential evaporation of the more volatile component. This decrease is also in accordance with the analysis of Calus and Rice (1972) who stated that for higher diffusion rates of the more volatile component, there will be a higher resistance to heat transfer. Hence with further increase in R_a , the nucleation

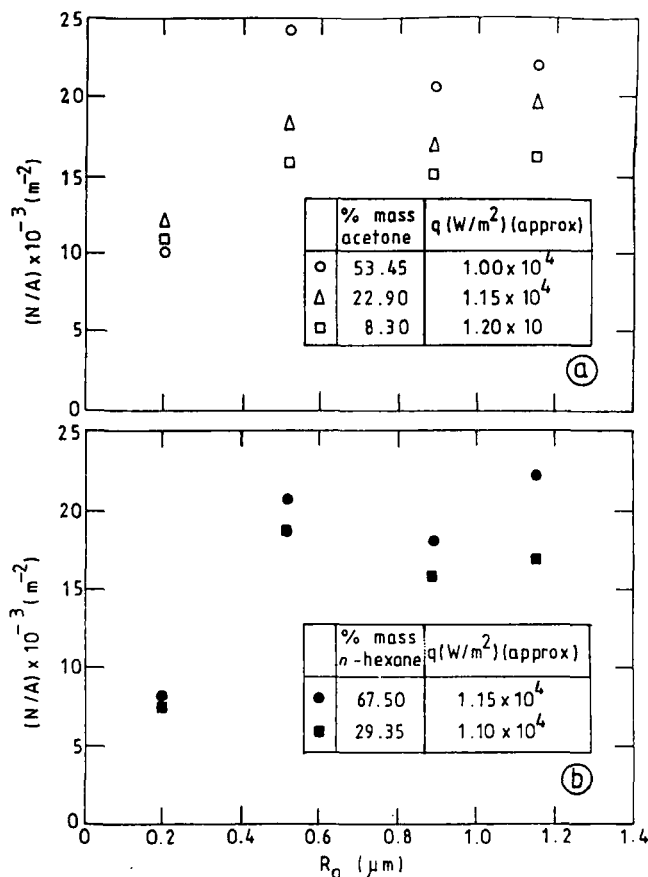


Figure 10 — (N/A) versus R_a : (a) acetone-carbon tetrachloride system; (b) *n*-hexane-carbon tetrachloride system.

site density continues to decrease and then reaches a minimum. After the minimum, the nucleation site density increases again. This is probably because each cavity becomes so large that two or more bubbles can nucleate from the cavity. In each of these smaller nucleation sites, the liquid left behind becomes negligible once again. This increases the temperature driving force and leads to an increase in the nucleation site density.

The dependence of the nucleation site density on the mass fraction of the more volatile component for the two systems studied is shown in Figures 10 and 11. It is seen that for both the systems, the nucleation site density decreases and then increases with increasing mass fraction of the more volatile component in the binary mixture. When a low boiling liquid is added to a higher boiling liquid, the nucleation site density decreases initially because of the presence of the second liquid (which is more volatile). When a binary mixture evaporates, there is preferential evaporation, with the vapour richer in the more volatile component. Therefore, during bubble growth, the more volatile component diffuses from the bulk towards the bubble impeding heat transfer, leading to a reduction in the temperature driving force and thereby decreasing the nucleation site density. As the composition of the more volatile component in the liquid increases further, the nucleation site density increases. This is because as x continues to increase, $|y-x|$ decreases and the value of $1-|y-x|(D_{AB}/\alpha_1)^{0.5}$ increases, leading to an increase in the nucleation site density. This is in agreement with the analysis of Stephan and Körner (1969) who concluded that as $|y-x|$ decreases, heat transfer to the bubble increases (leading to an increase in the nucleation site density).

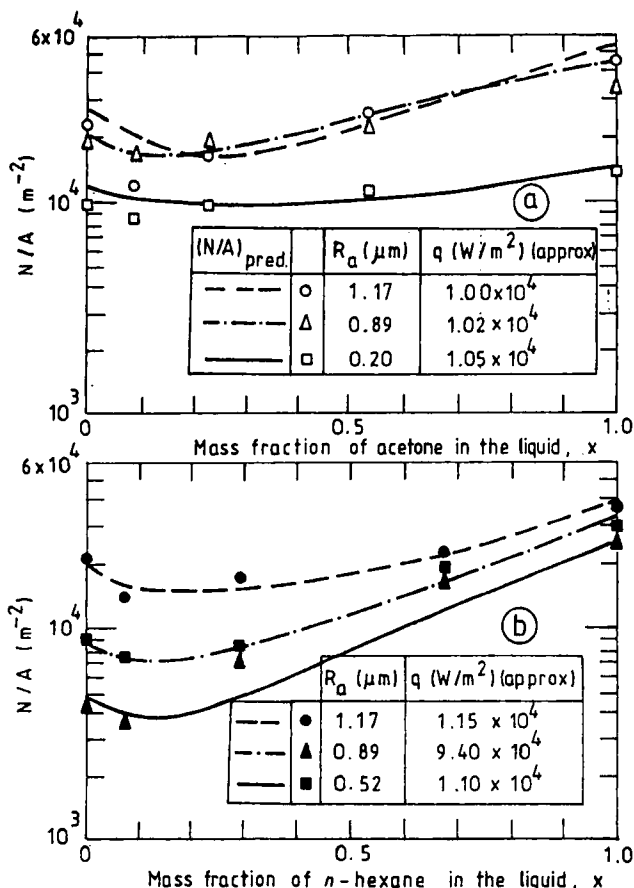


Figure 11 — (N/A) versus mass fraction of the more volatile component: (a) acetone-carbon tetrachloride system; (b) *n*-hexane-carbon tetrachloride system.

The dimensionless surface roughness (θ) is defined in the form of a polynomial because this form alone can take into account the increasing-decreasing trend of the nucleation site density with increasing surface micro-roughness in the case of binary mixtures. A polynomial form of θ as a function of R_a alone can correlate the nucleation site density data, but there will be an additional dependence of the nucleation site density on the surface tension. The centre line average R_a and the surface tension together take into account the effect of the contact angle on the nucleation site density implicitly. Experimental contact angle data are not readily available. Further, the contact angle is specific to a particular surface material — surface finish — liquid combination. Changes in surface micro-roughness cause a lot of variation in the contact angle (Hong et al. (1994)). Further, the term ' $R_a P / \sigma_1$ ' comes from the force balance on a bubble. That is,

$$2\pi R\sigma = \pi R^2(P_v - P_l) \dots\dots\dots (10)$$

Therefore,

$$P_v - P_l = \frac{2\sigma}{R} \dots\dots\dots (11)$$

where R is the bubble radius. Therefore, in the present study ' $R_a P / \sigma_1$ ' was used to obtain a dimensionless surface roughness and correlate the data. The same expression of θ that was developed in an earlier study on pure liquids (Benjamin

and Balakrishnan, 1997) was used to correlate the nucleation site density data for binary mixtures also. Since for binary mixtures the nucleation site density increases and then decreases with R_a , the exponent of θ was of the opposite sign (-0.4 for pure liquids and 0.4 for binary mixtures). Although the nucleation site density increases, decreases and then reaches a minimum, the further increase of the nucleation site density with R_a was very marginal. Hence, the same form of θ correlated the data satisfactorily.

Ramilson et al. (1992), while seeking to correlate the peak heat flux in terms of the surface micro-roughness and contact angle, observed based on boiling data in the literature, that "smooth" (or "rough") and "wetted" (or "non-wetted") surfaces can be discussed only in connection with a surface — liquid combination. A "smooth" surface for one liquid can serve as a "rough" surface for another. This is analogous to a "rough" pipe being hydrodynamically "smooth". This means that the profilometer data (R_a values) alone are not sufficient to characterise the effect of surface micro-roughness on the nucleation site density and the heat flux. The liquid and surface physical properties also play a significant role in determining the nucleation characteristics and the heat transfer rate. Therefore, in the present study, a surface-liquid interaction parameter γ (the ratio of the thermal conductivity, density and specific heat of the surface to that of the liquid) was used in conjunction with the profilometer data (the R_a values for different surface finishes) in the correlation.

The surface-liquid interaction parameter γ is the ratio of the product of the thermal diffusivity of the solid to the liquid and the heat content of the solid to the liquid:

$$\gamma = \sqrt{\frac{k_w \rho_w C_{pw}}{k_l \rho_l C_{pl}}} = \sqrt{\frac{\alpha_w}{\alpha_l} \left(\frac{\rho_w C_{pw}}{\rho_l C_{pl}} \right)} \dots\dots\dots (12)$$

The parameter γ has been used by Sernas and Hooper (1969) and Ünal (1976) in their analysis of bubble growth during nucleate pool boiling. Further, Magrini and Nannei (1975), while proposing a correlation for the boiling heat transfer coefficient, observed that a parameter $(k_w \rho_w C_{pw})^{0.5}$ affected the heat transfer coefficient obtained. According to Kant and Weber (1994), the temperature of the heating surface during bubble growth is a function of γ and γ indirectly affects the bubble growth time. Therefore in addition to the profilometer output, the surface-liquid interaction parameter γ was used in this study to correlate the experimental data.

During bubble growth in nucleate boiling, there is convective flow of liquid away from the surface as liquid is being pushed away by growing vapour bubbles. Once the bubble departs from the heating surface, fresh liquid flows towards the heat transfer surface to take the place of the departed vapour bubbles. This was envisaged as "source" and "wake" flows respectively by Zuber (1963). Therefore, although there is no bulk flow, the viscosity of the liquid plays a significant role in pool boiling also. Han and Griffith (1965) analyzed pool boiling using distilled water as the boiling liquid and also developed a "bulk convection" model. They concluded that the viscosity of the boiled liquid determines the contact angle and the bubble departure diameter. The Prandtl number of the liquid (which takes the viscosity into account) was used by Rohsenow (1952) to develop his widely used correlation. He argued that although there is no net flow during pool boiling, the Prandtl number is important because the heat transfer (at a nucleation

site) takes place directly from the wall of the liquid. The Prandtl number of the liquid was also used by Nishikawa and Fujita (1990) to develop their bubble agitation model for pool boiling. Since the Prandtl number of the liquid is an important parameter which affects both the nucleation site density and the heat flux in pool boiling, it was used as a correlating parameter.

In the boiling of binary mixtures, the effect of the physical properties on the process of nucleation (and hence the nucleation site density) is often non-linear. Because of such non-linear property variations, the interaction of the heating surface with the boiled liquid (the binary mixture in this case) has to be accounted for explicitly over the entire range of compositions. For this reason a property – profile parameter ξ was defined and used. Although it is similar in form to the Bond number, it does not have the same physical meaning of the Bond number and is used here only to take into consideration the property variations and the surface profile.

Concluding remarks

The nucleation site density in the pool boiling of binary mixtures has been examined experimentally. The heating surface micro-roughness and the apparent temperature driving force were defined quantitatively in this study. The nucleation site density is found to depend on the heating surface properties, its micro-roughness, the liquid properties, the vapour-liquid equilibria and the wall superheat. A correlation has been developed in terms of these parameters. At low micro-roughness the binary mixture behaves as a pure liquid, but at higher values of micro-roughness mass transfer effects are significant.

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Nomenclature

A	= heating surface area, (m ²)
C_p	= specific heat, J/kg·K
C_{sf}	= constant in Rohsenow (1952) correlation, (–)
D_{AB}	= mass diffusivity, m ² /s
g	= acceleration due to gravity, m/s ²
k	= thermal conductivity, W/m·K
m	= slope of the vapour pressure curve, N/m ² ·K
n	= slope of the bubble point curve, (K)
N/A	= nucleation site density, (1/m ²)
P	= pressure, N/m ²
Pr	= Prandtl number, $Pr = C_p \mu / k$, (–)
q	= heat flux, W/m ²
R	= bubble radius, (m)
R_a	= centre line average, (m)
r_c	= cavity radius, (m)
T	= temperature, (K)
ΔT	= apparent excess temperature (wall superheat), (K)
x	= mass fraction of the more volatile component in the liquid phase, (–)
y	= mass fraction of the more volatile component in the vapour phase, (–)

Greek letters

α	= thermal diffusivity, m ² /s
γ	= surface-liquid interaction parameter defined in Equation (6), (–)
θ	= dimensionless surface roughness parameter defined in Equation (7), (–)
μ	= absolute viscosity, N·s/m ²
ξ	= properties-profile parameter defined in Equation (4), (–)
ρ	= density, kg/m ³
σ	= surface tension, N/m

Subscripts

l	= liquid
s	= saturation
v	= vapour
w	= heating surface (wall)

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