

FIG. 6. The locus of the position of the triple points for $\lambda_0 = 2$, and for $L = 0.2, 0.1, 0.05$, and 0.01 .

plane between these two curves in which the most unstable modes are oscillatory. The width of this region appears to diminish with decreasing σ and thus may not be discernible in these figures for lower values of σ , but nevertheless it exists.

The position of the solid curves in both Figs. 4 and 5 represents an absolute lower bound for the instability region in the first quadrant of the R - Rs plane. This is to say that any basic state configuration for which both R and Rs fall above that curve is linearly unstable. Thus the location of this curve is valuable as a general stability criteria for double diffusive convection.

Figure 6 shows the various envelope curves formed by the loci of the triple points for four Lewis numbers. Two significant features may be observed in this figure. The first is that all curves shown originate from a single point on the ordinate. This is the location of the critical point for $\Delta S = 0$. The second is that the inclination of each curve to the abscissa appears to increase with decreasing values of L . It may be concluded on the basis of this figure that the region in which the monotone unstable modes are dominant diminishes with decreasing L for any one value of Rs . However, this does not imply an equivalent increase in the stability region since, as was shown earlier, the size of that region is a function of σ . Depending on the value of σ there could be a substantial

region below each of these curves in which the most unstable modes are oscillatory. It was also found that all curves shown in Fig. 6 collapse onto a single curve in the first quadrant of the R - Rs plane when plotted on an R - (Rs/L) scale (i.e. on a redefined Rs scale to $Rs' = \beta g \Delta S d^3 / \kappa_s \nu$).

4. CONCLUSIONS

The analysis of the previous section revealed that the three stability and instability regions in the first quadrant of the R - Rs plane may be altered substantially by varying either σ or L . It was found that the region in which the oscillatory modes are found to possess maximum growth rates, in that portion of the plane, may diminish in size with increasing L . In fact, the results indicated that in the limit $L \rightarrow \infty$ only monotone modes possess maximum growth rates. This is a situation in which the instability is manifested by steady convective motion. This is also the case where mass diffusion has the dominant influence. Furthermore, it was found that for fluids possessing vanishingly small Prandtl numbers, again, only monotone modes have maximum growth rates for moderate values of both R and Rs . In this case, again, the instability is manifested by steady convection.

The analysis above lead also to the establishment of a simple general criterion for the onset of penetrative double diffusive convection. This was manifested by a single curve in the first quadrant of the R - Rs plane for all values of σ and L . The area above the curve comprises of a region of instability through monotone modes while below it the basic state configuration is either unstable through oscillatory modes or stable. There exists one such curve for every value of λ_0 .

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The relation between the rewetting temperature and the liquid-solid contact angle

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INTRODUCTION

DETERMINATION of the rewetting or minimum film boiling temperature is of great importance in reactor safety analysis during the reflooding phase of a hypothetical loss of coolant

accident (LOCA). This temperature separates the high temperature region of a fuel pin where inefficient film boiling or vapor cooling takes place, from the lower temperature region, where more efficient transition boiling occurs. As the minimum film boiling temperature is the boundary between transition and film boiling its knowledge is required in the application of transition and film boiling correlations.

Two principal mechanisms are commonly proposed in the literature for the rewetting phenomenon.

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NOMENCLATURE

a	parameter, equation (1)	γ	surface tension
B	constant, equation (2)	θ	contact angle.
T	temperature		
T^*	temperature corresponding to zero contact angle	Subscripts	
V_L	molecular volume.	L	liquid
		V	vapor.
Greek symbols			
α	parameter, equation (1)		

(a) Hydrodynamic instability: the separation of the liquid-vapor interface is possible so long as the liquid forces do not exceed the vapor forces. When a force unbalance occurs, the liquid forces overcome vapor ones, the vapor film collapses and rewetting takes place, e.g. refs. [1-3].

(b) Thermodynamic mechanism: it is assumed that a liquid phase cannot exist beyond the maximum liquid superheat temperature, which is specific to each liquid. Therefore, a surface heated to a temperature above this value, disables liquid-solid contact, e.g. refs. [4-6].

Several authors considered the wetting ability of liquids [7-9]. For a moving liquid it was shown that its wetting characteristics are strongly connected with the adsorption potential of its vapor on the surface [9]. For a static configuration a rather qualitative relation between temperature and the liquid-solid contact angle developed in ref. [10], was used in ref. [9] to point at the possibility of analyzing the wetting mechanism.

Based on the physical process which takes place when a liquid droplet contacts a very hot surface, e.g. ref. [12], a zero contact angle is suggested here as the angle the corresponding temperature of which is an upper bound on the rewetting temperature. An improved version of the contact angle-temperature relation developed by Adamson [11] is then used to find upper bounds on the temperature for water on two non-metallic surfaces. It is shown also that other upper bounds on the rewetting temperature can be obtained by a linear extrapolation of known contact angle-temperature curves to zero contact angle. Using this method, bounds on the rewetting temperature are presented for water on several polymer surfaces.

THEORY

The rewetting temperature is generally considered to correspond to a state at which the liquid constitutes large contact angles with the solid surface, i.e. when the liquid has nearly point contact with the surface. In the sequel we offer a different point of view, leading to the association of the rewetting temperature with small liquid-solid contact angles.

Let us consider the process taking place when a liquid droplet comes into contact with a surface, as described by the series of photographs in Figs. 3-13 of ref. [12]. Upon impact the droplet forms a large contact angle with the surface, having still a nearly spherical shape and a very small contact area. Then, its base begins to expand causing it to take a hat shape, with a decreasing contact angle. In the meantime vapor is generated beneath the droplet until a vapor film is formed and liquid-surface contact terminates. At this stage the droplet spread to its maximum expansion capability, liquid-solid adhesion forces do not exist anymore, and the droplet starts to gather on a vapor cushion as a result of surface tension forces, and finally leaves the surface. Thus, the complete separation of the droplet from the wall starts when its base is at maximum spreading (i.e. at a very small contact angle, $\theta \approx 0^\circ$) and not at nearly point contact ($\theta \approx 180^\circ$).

It is suggested here that the threshold between wetting and non-wetting is the stage at which complete separation of the droplet from the surface begins. The contact angle decreases with temperature up to the maximum expansion limit of the droplet, at very low contact angles, when its base area is large enough for a substantial amount of heat to flow from the solid to the liquid, thereby generating the disjoining vapor film. This mechanism explains also the observed change in heat transfer removal upon droplet separation. Such a sharp transition would not be possible for a gradual separation of a droplet forming a bell shape with an increasing contact angle.

Since the rewetting temperature constitutes a sharp threshold between a state of wetting and non-wetting, it is suggested in light of the above physical description that this temperature corresponds to a state slightly after the droplet reaches its maximum expansion limit, at the beginning of its separation from the hot surface by the vapor film, which occurs at very low contact angles. This is the reason why it is postulated here that the temperature which corresponds to zero contact angle constitutes an upper bound on the rewetting temperature.

An explicit relation is derived now for the dependence of the liquid-solid contact angle, θ , on the surface temperature, T . Adamson [11] obtained the following implicit relation:

$$\frac{d \cos \theta}{dT} = (\cos \theta - 1) \left(\frac{a}{\alpha - a} \frac{1}{T^* - T} - \frac{d \ln(\gamma_{LV} V_L)}{dT} \right) \quad (1)$$

where T is the temperature, T^* the temperature which corresponds to a zero contact angle, V_L the molecular volume of the liquid, γ_{LV} the surface tension of the liquid against its own vapor, and a and α are parameters which appear in Adamson's adsorption isotherm.

Relation (1) is rather inconvenient to use, since it requires the knowledge of $d \cos \theta / dT$ over the whole range of relevant temperatures if one wishes to plot θ vs T . A more practical relation can be derived by integrating equation (1), which gives

$$\cos \theta = 1 - \frac{B}{\gamma_{LV} V_L} (T^* - T)^{a(\alpha - a)} \quad (2)$$

where B is a constant. The evaluation of B and T^* may be obtained by curve fitting to experimental results. The parameters γ_{LV} , V_L , a and α are known properties for a given system. Equation (2) can be used in principle to predict the behavior of the contact angle and to trace the function $\theta(T)$ in the whole range by a limited number of data points (the minimum is obviously two points, which will result in two equations for the two unknowns B and T^*).

The temperature T^* which appears in equation (2) is the desired upper bound on the rewetting temperature, according to the aforementioned proposal. Though of most interest is probably the rewetting temperature of water coming into contact with metal surfaces, it seems that there are no suitable data for carrying out an analysis such as was previously outlined. Therefore, we find T^* by fitting equation (2) with

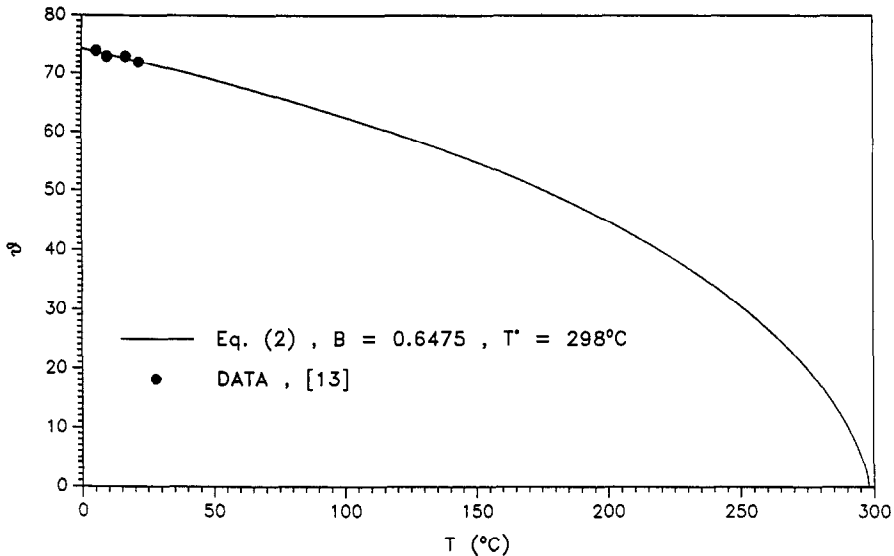


FIG. 1. Variation of the contact angle with temperature for water on pyrolytic-carbon-silicon alloy.

experimental data for water on two polymer surfaces. The relation between $\gamma_{LV}V_L$ and the temperature for water is [13]

$$\gamma_{LV}V_L = 1366.7 - 2.735T(^{\circ}\text{C}). \quad (3)$$

Water on pyrolytic-carbon-silicon alloy

From ref. [13], $a = 0.067 \text{ \AA}^{-1}$ and $\alpha = 0.015 \text{ \AA}^{-1}$. From the same reference the experimental data given in Table 1 were used for a least squares fitting of equation (2). The following values were obtained: $T^* = 298^{\circ}\text{C}$ and $B = 0.6475$. The resulting variation of the contact angle with temperature is shown in Fig. 1.

Water on polyethylene

From ref. [13], $a = 0.463 \text{ \AA}^{-1}$ and $\alpha = 0.0074 \text{ \AA}^{-1}$. The experimental data of ref. [14] yield after least squares fitting of equation (2): $T^* = 208^{\circ}\text{C}$ and $B = 7.133$. The resulting variation of the contact angle with temperature is plotted in Fig. 2.

Another way to find an upper bound for the rewetting temperature is as follows. In the investigation of ref. [14],

several plots similar to Fig. 3 are presented for the variation of the contact angle with temperature. For these liquid-solid pairs a and α are not available, so that an analytical relation between θ and T cannot be constructed. However, useful information about T^* can still be obtained. By extrapolating linearly the curve until it intersects the T axis (at $\theta = 0^{\circ}$, see the dashed line in Fig. 3) one may determine 'an upper bound on the upper bound'. At low contact angles the θ - T curve is expected to decrease more steeply than that obtained by a linear extrapolation. Therefore, the values which are obtained for T^* by this method will be higher than those predicted by the use of equation (2). Table 2 summarizes the results of such extrapolations for water on several non-metallic surfaces.

DISCUSSION

For a static configuration surface wettability is suitably expressed by the contact angle. In this investigation an improved Adamson contact angle-temperature relation was used to predict an upper bound on the rewetting temperature.

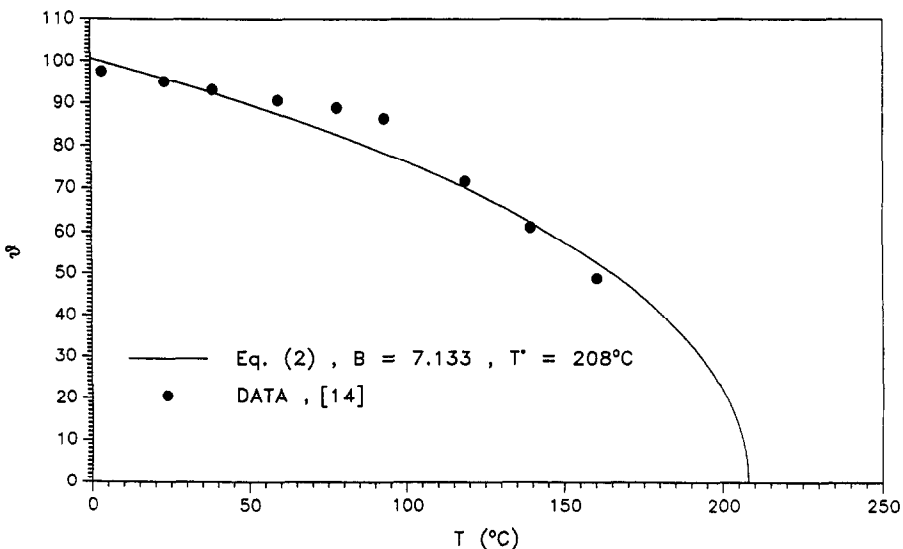


FIG. 2. Variation of the contact angle with temperature for water on polyethylene.

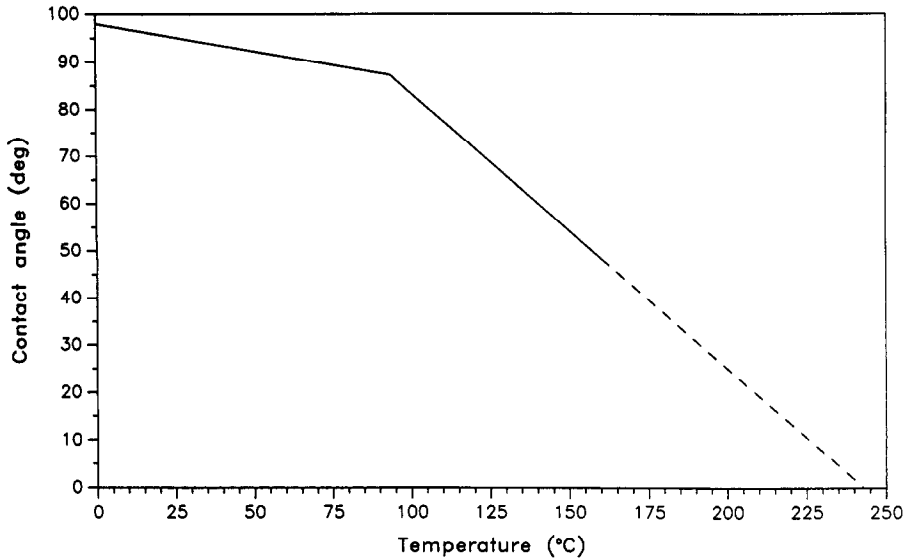


FIG. 3. Temperature dependence of the contact angle of water on polyethylene (the solid line is taken from ref. [14]).

The temperature which corresponds to zero contact angle was postulated to constitute an upper bound on the rewetting temperature. This proposal was based on observations on the behavior of a droplet coming into contact with a hot solid surface. The values for T^* which were obtained from either using equation (2) or by linearly extrapolating known

contact angle-temperature curves to $\theta = 0^\circ$ lie within the common acceptable range for the rewetting temperature of water, i.e. 140–350°C at atmospheric pressure. At present, only few experimental values for adsorption parameters and contact angle-temperature data are available for pertinent systems. These data are needed to establish the functional relation between θ and T and for evaluating T^* , the upper bound on the rewetting temperature. In particular there is need for such data for liquids on metal surfaces in order to employ the results in the important analysis of nuclear safety.

Table 1. The experimental data of contact angle, θ , at various temperatures, T , used in the least squares fitting of equation (2) for water on non-metallic surfaces

Water on pyrolytic-carbon-silicon alloy [13]	
θ	T (°C)
74	5.5
73	9.5
73	16.5
72	21.5

Water on polyethylene [14]	
θ	T (°C)
97.5	3.5
95.1	23.2
93.2	38.3
90.7	59.1
88.9	77.7
86.4	92.8
71.6	118.3
61.1	139.1
48.8	160.0

Table 2. The temperature, T^* , corresponding to zero contact angle for water on various solids

Solid	T^* (°C)
Polyethylene	243
Polystyrene	177
Polyacetal	290
Polycarbonate	213
Polyester	189
Fluoropolymer	302

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