of natural convective heat transfer. At Rayleigh numbers greater than about 2×10^4 a new pattern of temperature rise appeared (Fig. 1b). The centre temperature still fell to the steady plateau, but this was terminated more sharply than at lower Rayleigh numbers and the temperature rose to a shallow maximum, and then fell again before rising smoothly to the vessel temperature. Under the most extreme conditions used three successive maxima and minima were recorded. That these effects were not induced by the method of expanding the gases was shown by altering the direction of the side-arm and by expanding the gases simultaneously through two side-arms: these changes did not affect the pattern of temperature variation observed. Some details of the movement of the gases in this region of mainly convective heat transfer were obtained using the 1100-cm^3 vessel with four thermocouples. These showed a brief period of conductive heating of the gas near the wall, establishing steep temperature gradients, followed by a downward movement of the colder gas near the centre of the vessel. This draws down warmer gas from the top of the vessel, so causing the rapid rise of temperature recorded on the centre thermocouple. The later cooling is caused by the downward movement of gas from regions in the upper half of the vessel, but not directly above centre. The technique available does not yield sufficient detail to permit the estimation of heat-transfer coefficients for these transient processes.

From these experiments it has been possible to identify the value of Rayleigh number below which there is purely conductive heat transfer in this situation of a spherical body of gas in contact with a vessel of much greater heat capacity. This value is extremely close to that obtained ($Ra = 600$) as the condition at which convection is first of significance in reacting gases in spherical vessels (2). In these circumstances the gas temperature is greater than that of the vessel for exothermic reactions, the temperature distribution across the vessel is approximately parabolic and changes only slowly with time in comparison with adiabatic expansions.

We wish to thank Mr. T. A. B. Wesley for undertaking the construction of the thermocouples used in this work.

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FILM BOILING OF A MIXTURE ON A HORIZONTAL PLATE

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NOMENCLATURE

g, acceleration of gravity;
g_c, conversion factor;
h, boiling heat-transfer coefficient;
k_v, thermal conductivity of vapor;
L', enthalpy change of vaporization plus sensible heat;
Q/A, heat flux;
ΔT, temperature difference driving force;
x, y, mol fraction Freon-113 in liquid, vapor;
μ_v, viscosity of vapor;
ρ_L, ρ_v, density of liquid, vapor;
σ, surface tension.

THEORETICAL analyses exist for film boiling of pure liquids on horizontal cylinders [1] and horizontal flat plates [2, 3]. Experimental data [1, 4, 5] with pure liquids substantiate the equations. The only published data for a mixture in film boiling are the data of Dunskus and Westwater [6] who were examining the effects of trace amounts of high molecular weight, nonvolatile additives on the heat transfer to *iso*propanol. As little as 0·1 mol% of one additive, Igepal CO-880, caused the heat-transfer coefficient to increase by about 300 per cent. No data exist for miscible mixtures of two volatile components.

The goal of the new research was to obtain accurate film-boiling data for a miscible mixture of volatile liquids. It was of interest to show whether the results would agree with predictions based on the use of weighted physical properties in the existing equations for pure liquids.

The flat-plate geometry was chosen because the film

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boiling theory has been well worked out for this geometry. Zuber proposed that the situation existing at the minimum heat flux is Taylor hydrodynamic instability. He derived several expressions for the minimum heat flux in film boiling, the variation being due to different assumptions as to the bubble distribution pattern. Two of Zuber's expressions are as follows:

$$\min (Q/A)_1 = \pi L'\rho_v/24[\sigma g_c g(\rho_L - \rho_v)/(\rho_L + \rho_v)^2]^{\frac{1}{4}} \quad (1)$$

$$\min (Q/A)_2 = [0{\cdot}4\pi\sqrt{(2)}/(3)^{0{\cdot}25}](Q/A)_1. \quad (2)$$

Berenson also used Taylor instability and derived expressions for the heat-transfer coefficient, the minimum flux and the temperature difference at the minimum flux during film boiling. His expression for the heat-transfer coefficient is

$$h = 0{\cdot}425\left\{\frac{k_v^3\rho_v(\rho_L - \rho_v)L'g}{\mu_v\Delta T[g_c\sigma/g(\rho_L - \rho_v)]^{\frac{1}{2}}}\right\}^{\frac{1}{4}}. \quad (3)$$

The value 0·425 is an empirical constant, selected to fit CCl_4 and also pentane. All these equations were derived for pure liquids.

The apparatus used for the film boiling studies was a modification of the equipment used by Hosler and Westwater [4] in earlier film boiling studies of pure liquids. The heat-transfer surface was an 8-in. square, 3-in. thick, horizontal, aluminum plate whose bottom surface was extended by 342 vertical, stainless steel rods, of $\frac{1}{4}$-in diameter and 8-in length. City gas was burned beneath the plate. A copper boiler-housing fastened to the top of the plate contained the boiling liquid. The four sides and the top of the boiler had windows; film boiling was confirmed always by visual observation. A vapor loop had provisions to condense the vapor, reheat the condensate to the saturation temperature, meter and returned it to the boiler. Thermocouples in the aluminum block permitted the surface temperature to be computed by extrapolation. The uncertainty in the surface temperature is believed not to exceed ±3 degF. Liquid samples were removed from the boiler by means of a hypodermic needle in one side window of the housing. Samples were taken also from the condensed vapor. Analyses were carried out by refractive index measurements. The heat-transfer rate was calculated from the boil-up rate, the vapor composition and the latent heats of vaporization of the pure components. This rate was reproducible within ±3 per cent. The heat of mixing of a 50:50 mixture was found to be 0·9 Btu/lb, a negligible value.

The mixture carbon tetrachloride–Freon 113 (CCl_2FCClF_2) was used for the tests. The physical properties of the pure components are known. For the mixtures the surface tensions were measured with a ring tensiometer. The results are shown in Fig. 1. Vapor–liquid equilibrium data at 760 mm were determined with a Fowler–Norris still [7] having a pressure controller. The equilibrium data are shown in Fig. 1.

Measurements in the flat-plate test apparatus indicated that the boiling liquid in it and vapor leaving it had equal temperatures and had equilibrium compositions. Of course this does not mean that the thin vapor film adjacent to the hot aluminium plate was at the liquid temperature nor at the vapor composition.

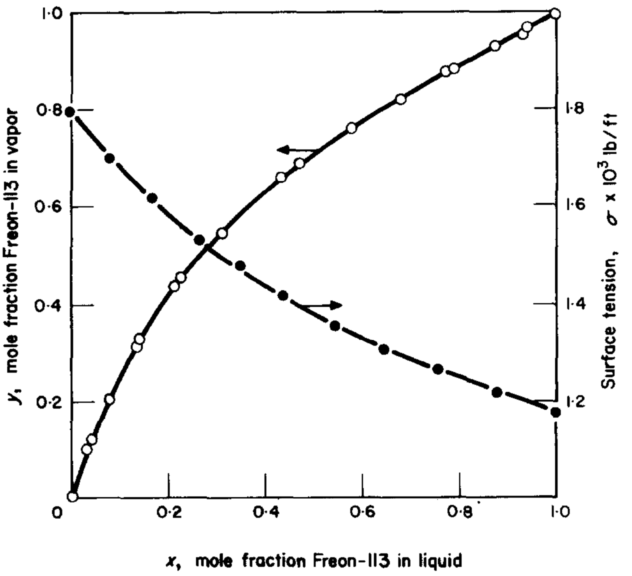

FIG. 1. Vapor–liquid equilibrium data at 760 mm, and surface tension data at 25°C, for mixtures of CCl_4 and Freon-113.

A typical run was made by heating the boiler plate to approximately 550°F before introducing any liquid into the boiler. Liquid to a depth of 1 in was then added, and film boiling was immediately established. When the system reached an equilibrium, the thermocouple temperatures were recorded, measurements of the condensate composition and flow rate were taken, and a liquid sample was withdrawn from the boiler and analysed. The boiling temperature was determined from the liquid composition and use of the experimental boiling point diagram. This temperature was subtracted from the surface temperature to get the boiling ΔT. All runs were carried out at atmospheric pressure.

The heat-transfer data for four liquid compositions are given in Fig. 2. Seven compositions are included in Fig. 3. The boiling curves in Fig. 2 show that pure CCl_4 gives lower heat fluxes than pure Freon-113 at all values of ΔT. The difference between these two curves is about 18 per cent. However, the various mixtures generally do not give intermediate values. In fact, many of the mixtures give heat fluxes higher than those for either pure component at the same ΔT. This is shown clearly in the cross-plot, Fig. 3. Note that the addition of about 1 per cent of the second component to either pure component causes a very real increase in the heat flux—in some cases by as much as 20 per cent, which is significantly greater than the possible experimental errors.

In Fig. 2, the predicted Q/A vs. ΔT is shown, as calculated from Berenson's equation (3), for pure CCl_4. The prediction for CCl_4 is very close at the higher ΔT values, but it is low by 20 per cent at ΔT = 180°F. For pure Freon, the predicted curve intersects the data at ΔT = 200°F, and at ΔT = 325°F, the prediction is high by 10 per cent. For most applications, these errors are acceptable, therefore the Berenson equation is useable for the pure components in the ΔT range of about 175–350°F.

The left end of each experimental line in Fig. 2 corresponds to the minimum heat flux. A slight decrease in the ΔT, for each test, caused a breakdown of film boiling (visually apparent) and a sharp rise in the heat flux. The observed minimum heat fluxes for pure CCl_4 and pure Freon-113 fell

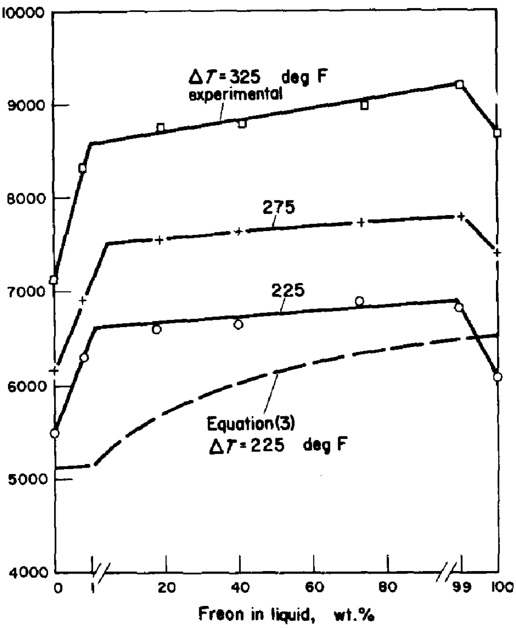

Fig. 3. Effect of composition on heat flux. The dotted line shows the theoretical prediction using weighted physical properties. Note the change in scale on the abscissa at 1 and 99 per cent.

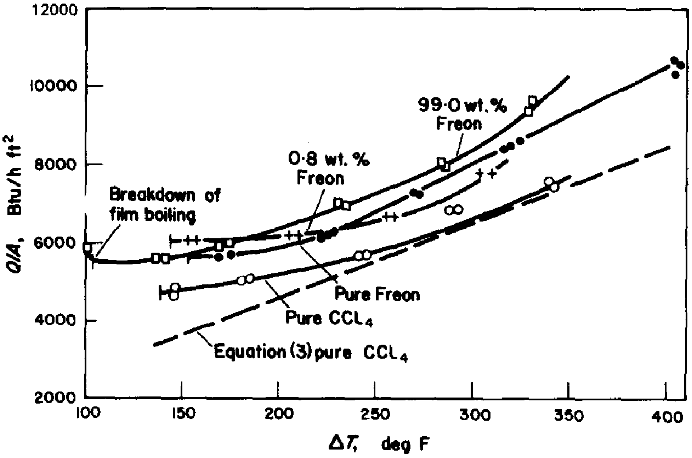

Fig. 2. Film-boiling curves for mixtures of CCl_4 and Freon-113 on a horizontal plate at atmospheric pressure.

about half way between Zuber's equations (1) and (2). These minimum fluxes occurred at ΔT values which were 6 degF higher (for CCl_4) and 43 degF higher (for Freon-113) than the ΔT values predicted by a theoretical equation given in Berenson's paper. The result is that Berenson's equation is low by about 35 per cent when used to predict the minimum heat flux for pure Freon-113.

May one interpolate between results for pure components to predict the results for mixtures? Figure 2 shows clearly that such a procedure is not valid. Nor is it valid to use weighted physical properties. The dotted line in Fig. 3 represents equation (3), using weighted physical properties for the liquid (at its saturation temperature and composition) and the vapor (at the same composition as the liquid but at the mean temperature of the vapor film). The theoretical predictions are wrong in shape; they do not predict large effects for small composition changes at compositions near the end points. The marked effect of the addition of a small amount of a second component to a pure system suggests that mass diffusion plays an important role in the heat-transfer process. Apparently, the diffusion problem will have to be coupled with the present theory to predict satisfactorily the film boiling behavior of miscible liquid mixtures.

ACKNOWLEDGEMENT

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