

the larger particles to dry under free liquid condition. With solutions other than Lignosol it is possible that the falling-rate period will become significant, but this will have to be determined experimentally for each individual case.

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NOTATION

a = $E(1/R_1 - 1/R_2)$;
 c = solution concentration, lb. solids/lb. solution
 c_i = solution concentration in i th group, lb. solids/lb. solution
 C_p = heat capacity of particles, B.t.u./lb. ($^{\circ}$ F.)
 C_p = heat capacity of water vapor, B.t.u./lb. ($^{\circ}$ F.)
 D = particle diameter, ft.
 D_i = average particle diameter in i th group, ft.
 D_v = mean volume diameter, ft.
 D_{vs} = mean Sauter diameter, μ
 E = $(dm/d\theta)(C_p/4\pi k_f)$
 h = heat transfer coefficient, B.t.u./hr. (sq.ft.) ($^{\circ}$ F.)
 H = air humidity, lb. water/lb. dry air
 H_{hb} = air humidity calculated from heat balance, lb. water/lb. dry air
 H_{mb} = air humidity calculated from material balance, lb. water/lb. dry air
 k_f = thermal conductivity of gas film surrounding the drop, B.t.u./hr. (ft.) ($^{\circ}$ F.)

m = evaporation, lb.
 n = rate of particles, No./hr.
 Q = heat transferred to droplets, B.t.u.
 R_1 = radius of particle, ft.
 R_2 = radius of outer limit of gas film surrounding the drop, ft.
 t = temperature of air, $^{\circ}$ F.
 t_w = temperature of particles, $^{\circ}$ F.
 X = moisture content, lb. water/lb. dry Lignosol
 X_{hb} = moisture content calculated from heat balance, lb. water/lb. dry Lignosol
 X_{mb} = moisture content calculated from material balance, lb. water/lb. dry Lignosol
 x = distance from nozzle, ft.
 Z = total rate of dry Lignosol, lb./hr.
 Z_i = rate of dry Lignosol in particles in i th group, lb./hr.
 Δm = amount of water evaporated
 $\Delta\theta$ = time increment
 θ = time, hr.
 λ_w = latent heat of evaporation at the particle temperature, B.t.u./lb.
 ρ_s = density of particles, lb./cu.ft.
 ρ_{si} = average density of particles in i th group, lb./cu.ft.
 N_{Gr} = Grashof number
 N_{Nu} = Nusselt number
 $N_{Nu'}$ = modified Nusselt number
 N_{Pr} = Prandtl number
 N_{Re} = Reynolds number
 N_{Sc} = Schmidt number

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The Effect of Dynamic Surface Tension on Nucleate Boiling Coefficients

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A study was made of the effects the surface tension of aqueous solutions of surface-active agents had on the bubble-formation characteristics of these solutions. Bubbles were formed by two processes: by passing air through a horizontal orifice submerged in the solution and by nucleate boiling at an electrically heated surface. The experimental measurements taken with the air-bubble system were used to determine the nature of the surface-tension effect on air-bubble formation. This knowledge was then applied to the interpretation of boiling measurements made with the same solutions. The boiling coefficients of the Tergitol-water solution were found to vary inversely with dynamic surface tension. Boiling coefficients for Aerosol-water solutions, however, varied in a manner which could not be related to surface-tension effects.

Numerous investigators have studied the role of surface tension in boiling

and have reported widely contradictory results. Westwater (10) summarizes the results in terms of the value as-

signed to the exponent n in an assumed exponential relationship, $h \propto 1/\sigma^n$; the range of values he reports is 2.5 to -1.275. As Westwater points out, there

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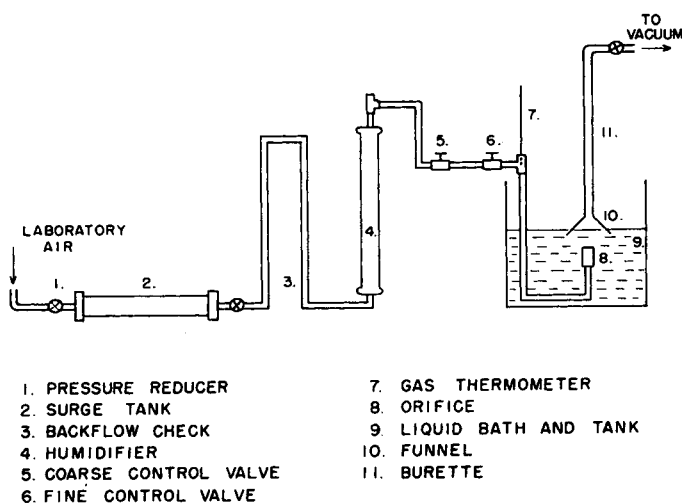


Fig. 1. Diagram of bubble apparatus.

are theoretical grounds for considering surface tension to be an important variable in boiling, and these have led numerous investigators to attempt quantitative measurements of the relationship. Experimental evidence gathered to date, however, has not proved the existence of such an effect.

The commonest experimental approach has been to add a surface-active agent to a boiling solution and from the results to establish an exponential relationship between the measured boiling coefficients and the static surface tension of the solution. It is known that the surface tension of a solution of a surface-active agent is dependent on the history of the system. For example, DuNouy (3) reports equilibrium values for aqueous sodium oleate solutions which are only 40% of the measured dynamic values. The time necessary to achieve equilibrium was between 2 and 10 min., depending on concentration. Tests with rabbit serum required up to 2 hr. for equilibrium to be established. Weiser (9) shows large differences between static and dynamic surface tensions for solutions of sodium oleate, saponin, heptylic acid, and lauryl sulfonic acid; dynamic determinations were made by the capillary-wave method and static surface tensions by the capillary-tube method.

Because the formation of bubbles in a boiling system is an extremely rapid process, sometimes requiring only a few milliseconds, it would seem that if surface tension has any effect on boiling solutions it would be the dynamic surface tension which would apply and not the static surface tension. Several methods have been reported for the measurement of dynamic surface tension (8). These involve measurements of such diverse quantities as the oscillation rate of a falling drop, the distance between nodes in a vibrating

liquid jet issuing from an elliptic orifice, and the cone angle produced by two impinging jets. However the total lack of geometric similarity between these systems and a boiling system makes it unlikely that they would be of use in predicting the dynamic surface tension of a boiling system.

In the present investigation (4) an attempt was made to measure the surface tension of aqueous solutions of surface-active agents under nonboiling conditions which nevertheless approximated the boiling process. These results were used to interpret the effect of surface tension on boiling coefficients under actual boiling conditions.

The nonboiling studies were based on the work of Benzing and Myers (2), who, studying factors affecting gas-bubble formation at horizontal circular orifices, found that for a number of gas-liquid systems the bubble volumes could be predicted with considerable accuracy by

$$\frac{D}{d} = 1.82 \left(\frac{\sigma}{\rho_L g d^3} \right)^{0.25} \quad (1)$$

This was based on an equation derived by writing a force balance on a bubble which was just at the point of breaking off. The theoretical expression however contains an exponent of 0.33 on the right-hand side instead of 0.25. The data obtained under a variety of conditions fitted this expression, including results obtained with an aqueous solution containing 1.0 wt. % of the commercial preparation Drene. The surface tension employed in Equation (1) for this solution was a static value. Conversely, results obtained with a 0.1 wt. % solution of Drene in water deviated considerably from the correlation. The conclusion drawn from these experiments was that at a concentration of 1.0 wt. % there was sufficient surface-active agent present in the water to achieve orientation in the vapor-liquid

interface in the time available during bubble formation. Thus the measured static surface tension of the solution was approximately the same as the dynamic surface tension, but the 0.1 wt. % solution was apparently so dilute that a longer orientation time was required than was available. The static surface tension in this case, therefore, was not equivalent to the dynamic surface tension, and the use of the static value in the correlation resulted in substantial deviations. The measured values of the static surface tension were 30.6 and 26.0 dynes/cm. for the 0.1 and 1.0 wt. % solutions respectively.

Further evidence supporting this interpretation was obtained by studying the effect of bubble-formation rate on bubble volume. The volume of air bubbles in water was approximately twice the volume of air bubbles in 1.0 wt. % Drene solution in water, which was in accordance with the predicted effects of surface tension in Equation (1). The results obtained with a 0.1 wt. % solution of Drene in water were intermediate. At high rates of bubble formation, when very little time was available for surface concentration, the bubble volumes approached those measured for pure water. However at low bubble-formation rates the surface had more time to approach equilibrium, and the bubble volumes approached those measured for the 1.0 wt. % Drene solution.

From these experiments it was concluded that there may be concentrations of surface-active agent for which, under certain circumstances, the dynamic surface tension is equal to the static surface tension. It appears that this condition is approached by increasing the concentration of the agent or by decreasing the bubble-formation rate.

The information described above was used in the present study by performing a series of air-bubble experiments at various air rates and solution concentrations to determine the values of the dynamic surface tension. Boiling experiments were then conducted under conditions as nearly like the air-bubble studies as possible. The measured boiling coefficients from the heat transfer experiments were then correlated with the dynamic surface-tension measurements from the air-bubble experiments.

EXPERIMENTAL

Air-bubble and boiling investigations were performed with aqueous solutions of detergents known as Tergitol and Aerosol. Tergitol is a water-soluble yellow liquid containing 25% sodium tetradecyl sulfate in water; Aerosol-22 is an anionic agent containing *n*-octadecyl tetrasodium 1,2 dicarboxyethyl sulfosuccinamate and is soluble in both hot and cold water.

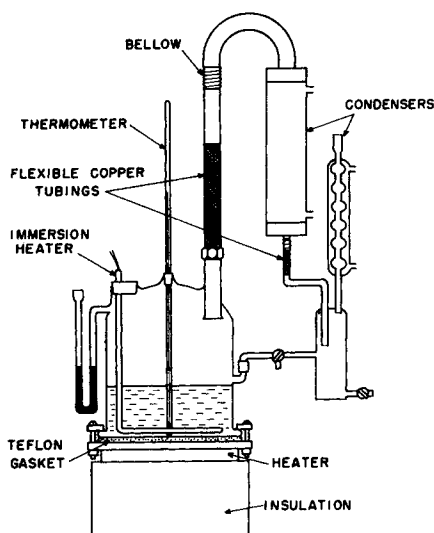


Fig. 2. Boiling apparatus.

Air-Bubble Experiments

The investigation of air-bubble volumes was done on an apparatus essentially the same as that described by Benzing and Myers (2) (Figure 1). Air passed through a pressure-reducing valve, discharging at approximately 5 lb./sq. in. gauge into a surge tank constructed from a 14-in. section of 2-in. pipe. This was followed by passage through a humidifier, constructed of 12 in. of 2-in. pipe. The flow rate was controlled by two needle valves in series. The orifice employed was made from brass bar stock and had a throat $\frac{1}{2}$ in. long with an I.D. of 0.357 cm. The orifice was immersed 3 in. beneath the surface of the liquid, which was contained in a cylindrical glass jar 1 ft. in diameter and 1 ft. high. The bubble-collection system consisted of an inverted funnel connected by rubber tubing to a 50-ml. burette, which was connected through a valve to an aspirator.

All data in this phase of the project were taken at room temperature, which ranged from 72° to 76°F. A platinum ring tensiometer was used to measure the static surface tension of the solution. Solutions were prepared on a volumetric basis and concentrations recorded as

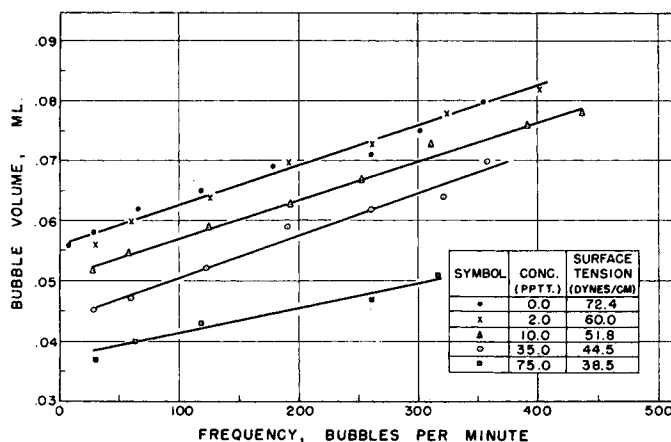


Fig. 4. Volume-frequency-concentration plot for Tergitol solution at 72° to 76°F.

volumetric parts of surface-active agent per ten thousand parts of water.

Boiling Experiments

The boiling data were taken with an apparatus (Figure 2) built by Kurihara (5) and described by Kurihara and Myers (6). The heater was cut from a cylinder of copper to a diameter of 3 in. Seven fins on the bottom of the block were heated by nichrome ribbon wound on a mica core and inserted between the fins. A stainless steel skirt (5-in. O.D.) was silver-soldered to the top of the block, and the copper and stainless steel were turned down till the skirt had a thickness of $\frac{1}{16}$ in. No boiling was observed from the stainless steel skirt. Thermocouples of No. 30 copper-constantan, located as shown in Figure 3, were used to measure temperatures in the heater block and on the skirt. Liquid temperatures were measured with a mercury thermometer whose bulb was $\frac{1}{2}$ in. above the center point of the heater surface. All thermocouples and the thermometer were checked against an N.B.S. calibrated thermometer and were considered to be accurate within $\pm 0.1^\circ\text{F}$.

The liquid was confined in an 8-in.-diameter inverted glass bell jar, 11 in. high, which was bolted to the heater

skirt and sealed by a Teflon gasket. An auxiliary heater inserted in a 6-mm. glass tube encircled the periphery of the heater to compensate for minor heat losses in the upper part of the apparatus. The external heater circuit included a 7-kv.-amp. variac rated at 20 amp. and 270 v.; two wattmeters, a high-range meter with 4,000 w. minimum and a low-range meter with 500 w. maximum; a 300-v. voltmeter; and a 25-amp. ammeter. Table 1 shows a sample of the experimental boiling data and results. All boiling data were taken at an approximately constant heat input of 400 B.t.u./hr. with heat flux from the heater surface varying from 7,460 to 8,040 B.t.u./ (hr.)(sq. ft.), depending on heat loss. Estimated bubble-formation rates were between 800 and 1,000 bubbles/min. for pure water. Upon the addition of the detergents these rates increased to values which could not be estimated visually.

In the Tergitol-water runs there were approximately twenty active nuclei on the heater surface when pure water was boiled and approximately seventy nuclei when the concentration of Tergitol was 75 p.p.t.; for the Aerosol-water runs there were thirty bubble columns with pure water and about 200 when the concentration was 7.5 p.p.t.

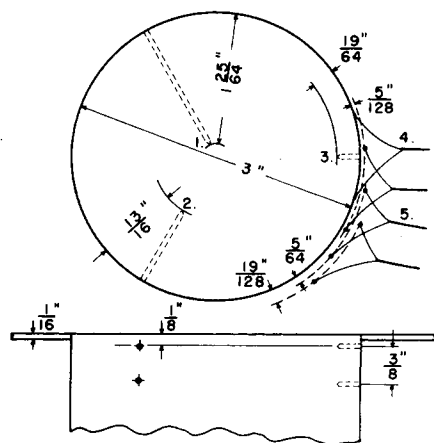


Fig. 3. Locations of thermocouples.

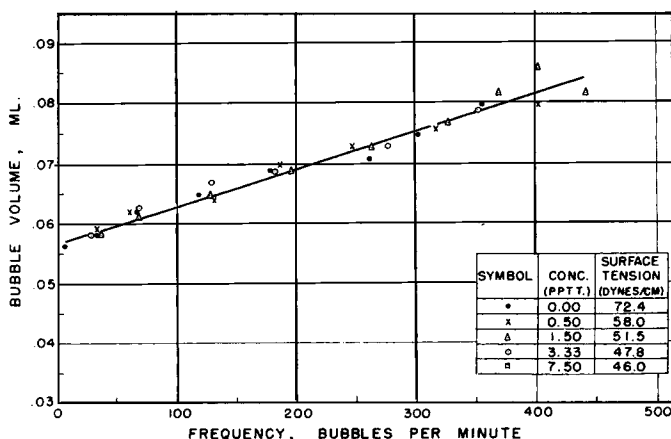


Fig. 5. Volume-frequency-concentration plot for Aerosol solution at 72° to 76°F.

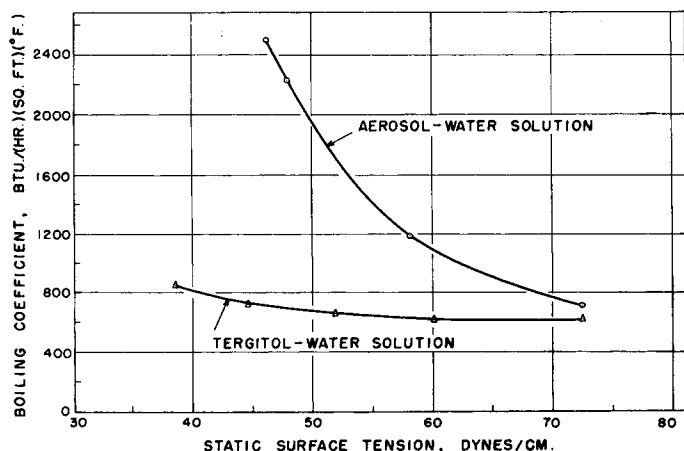


Fig. 6. Boiling coefficient vs. static surface tension.

The heat flux was determined by subtracting the skirt loss from the power input. A description of the details of the experimental measurements, including the method of determining average surface temperature, is given by Kurihara and Myers (6).

The boiling surface was prepared before each set of runs by systematic polishing with No. 2 emery paper. The differences in the boiling coefficient for pure water (716 for the Aerosol runs to 627 for the Tergitol runs) may be attributed to slight differences in the surface roughness (25.0 μ in. root mean square for the Aerosol runs and 22.6 μ in. root mean square for the Tergitol runs). The roughness was measured in the manner described by Kurihara and Myers (6).

The boiling surface was examined for fouling after each run. No visible deposits were left on the surface; however a slight increase of root mean square microroughness was observed (25.0 to 26.9 μ in. in one set of Aerosol runs and 25.0 to 26.4 μ in. in another set). This commonly occurs when pure water is boiled on a copper surface and is to be expected with aqueous solutions. It seems probable however that most of this change occurred during the 2 hr. of boiling which always preceded the taking of data.

Air-Bubble Results

The results of air-bubble measurements made in solutions of Tergitol are

shown in Figure 4. It is apparent that over the entire range of bubble frequencies employed there is a significant effect of detergent concentration, and thus of surface tension, on the measured bubble volumes. Measurements could not be taken at rates greater than 500 bubbles/min. in pure water because the bubbles were no longer uniform in volume or shape beyond this point. The limiting rate was even lower in solutions of detergent. There does not appear to be any major effect of bubble rate on the relative behavior of solutions of different concentrations, contrary to the observations of Benzing and Myers (2). With the exception of the most concentrated solution, the lines representing the various solutions appear to have nearly the same slope.

The results of air-bubble studies with solutions of Aerosol in water are shown in Figure 5. It may be seen that none of the solutions appeared to be affected by the presence of the detergent, as the bubble volumes were in all cases the same as for pure water. Thus the dynamic surface tension in this case is the same as the surface tension of pure water and, as far as surface tension can affect boiling, it would seem that the boiling coefficients of

these solutions should be the same as for water.

Boiling Results

The results of the boiling experiments are shown in Figures 6 and 7. As was expected from the air-bubble experiments with Tergitol solutions, there was a noticeable effect produced on the boiling coefficients. Addition of Tergitol up to 75 p. p. t. t. increased the boiling coefficient by about 50%. Although the air-bubble rates were below 500 bubbles/min. and the boiling rates generally above 1,000 bubbles/min., an estimate was made of the effect of surface tension on the boiling coefficients.

The first step in the calculation was to determine the values of the effective or dynamic surface tension from measurements of bubble volume by the use of Equation (1). The results of these calculations, based on bubble volumes taken from the straight lines of Figure 4, are given in Table 2 which also contains static readings for the same solutions.

If the heat transfer coefficient is assumed to follow a relation of the form $h\alpha(1/\sigma^n)$, then the exponent can be evaluated from a proportionality consisting of the ratios of heat transfer coefficients and dynamic surface tensions. This was done by using the data of Figure 6 and Table 2 with pure water as the reference state and produced the values shown in Table 3.

The heat flux for these runs was held between 7,400 and 7,710 B.t.u./ (hr.) (sq. ft.). Temperature-difference driving force ranged from 9.0 to 12.1 °F. The values of the exponent n ranged from 0.65 to 1.04, not a large range in view of the assumptions made. Although widely scattered values of the exponent have been reported by other authors, Bankoff (1) in a recent paper predicts a value of $n = 2/3$.

The results presented in Tables 1,

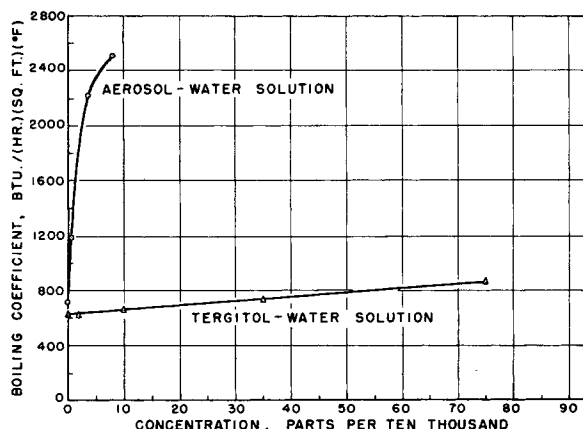


Fig. 7. Boiling coefficient vs. concentration.

TABLE 1. SAMPLE EXPERIMENTAL BOILING DATA AND RESULTS

System: Tergitol-water

Surface roughness: 22.6 μ in.

Concentration, at 72° to surface p.p.t.t. (by volume)	Static surface tension, 76° F., dynes/cm	Average surface temperature, °F.	Liquid temperature, °F.	Power input, B.t.u./hr.	Skirt loss, B.t.u./hr.	Heat flux q/A , B.t.u./ (sq. ft.) (hr.)	Film temperature drop, °F.	Boiling coefficient, B.t.u./ (hr.) (sq. ft.) (°F.)
0	72.4	223.5	211.6	399	33	7,640	11.9	627
2.0	60.0	223.2	211.1	399	31	7,510	12.1	621
10.0	51.8	222.4	211.1	399	32	7,490	11.3	663
35.0	44.5	221.2	211.1	392	29	7,400	10.1	732
75.0	38.5	220.1	211.1	403	25	7,710	9.0	856

2, and 3 are based on the dynamic surface tensions measured by air-bubble experiments at 72° to 76°F. When one uses this information to evaluate boiling performance at temperatures of 212° to 220°F., however, only the ratios of the surface-tension values at 72° to 76°F. are used. For this procedure to be valid, therefore, it is necessary only that the ratios not change with temperature. This point was investigated in the initial stages of the research by a series of measurements of the static surface tension of Tergitol solutions at temperatures up to 210°F. These values decreased by about 15% over the range of 70° to 210°F. However, the ratio of the static surface tension of a Tergitol solution to the static surface tension of water changed by amounts ranging only from 3 to 6%. Ratios of dynamic surface tension were assumed to have the same response to temperature change.

Preliminary experiments were made in which the air-bubble system was operated at 210°F. However, convection effects made it impossible to reproduce the results even when insulation was applied to the system.

The boiling results obtained with the Aerosol solutions were in direct contrast to what was expected. In Figure 5 it can be seen that the addition of the surface-active agent Aerosol had no effect on the volume of air bubbles. Thus it might be expected that it would have no effect on the boiling coefficients. An examination of Figures 6 and 7 shows, however, that the addition of 7.5 parts of Aerosol/10,000 parts of solution increases the boiling coefficient from 716 to 2,500 B.t.u./ (hr.) (sq. ft.) (°F.). In these runs the

heat transfer rate varied from 7,740 to 8,040 B.t.u./ (hr.) (sq. ft.) and the temperature-difference driving force from 10.8° to 3.2°F.

The presence of an effect in boiling but not in the air-bubble measurements clearly indicates that the effect of the detergent in this case is not related to the break-off dimensions of the bubbles in boiling. In fact, it would seem to have nothing to do with surface tension, since the dynamic surface tension of the solution is apparently unaffected by the presence of the detergent.

Lowery and Westwater (7) have suggested that large organic molecules in a solution may serve as nuclei for the formation of bubbles at smaller degrees of superheat than would be necessary at a liquid-solid interface. This explanation is attractive in view of the fact that the highest boiling coefficient [$h = 2,500$ B.t.u./ (hr.) (sq. ft.) (°F.)] occurred with a temperature-difference driving force of 3.2°F. At this superheat stable nucleate boiling is seldom attained, and the usual mechanism is one of natural convection.

CONCLUSIONS

It appears that the addition of surface-active agents to liquids may affect nucleate boiling coefficients in two quite different ways. First there may be a decrease in the dynamic surface tension with a consequent increase in the boiling coefficient. A method of studying this effect quantitatively has been proposed and tried with solutions of Tergitol in water. Results indicate that the exponent n in the relation $h\alpha(1/\sigma^n)$ varies between 0.65 and

1.04.

The second effect, which is much stronger, was found to occur in solutions of Aerosol and water. The presence of this detergent was found to have no effect on the dynamic surface tension. However a fourfold increase resulted in the boiling coefficient, which is, perhaps, due to the effect of the detergent on the number of active nuclei in the solution.

The authors suggest that a pluralistic explanation of the effects of detergents on the boiling behavior of solutions may be necessary to explain the extreme divergence of results obtained by previous workers in this field.

ACKNOWLEDGMENT

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NOTATION

d	= orifice diameter, cm.
D	= bubble diameter, cm.
g	= acceleration due to gravity, 980 cm./sec. ²
h	= boiling heat transfer coefficient, B.t.u./ (hr.) (sq. ft.) (°F.)
n	= constant in exponential relation $h\alpha(1/\sigma^n)$
q/A	= heat flux, B.t.u./ (hr.) (sq. ft.)
ΔT	= average surface temperature of heater minus bulk temperature of boiling liquid, °F.
ρ_L	= liquid density, g./cc.
σ	= surface tension, dynes/cm.

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TABLE 2. STATIC AND DYNAMIC SURFACE TENSIONS
TERGITOL-WATER SOLUTIONS

Concentration by volume, p.p.t.t.	Liquid temperature, °F.	Static surface tension, dynes/cm.	Dynamic surface tension, dynes/cm.			
			Air-bubble rates, bubbles/min.			
			30	100	200	300
0	72	72.4	72.4	72.4	72.4	72.4
2	73	60.0	72.4	72.4	72.4	72.4
10	75	51.8	65.5	65.7	66.0	66.6
35	75	44.5	56.3	58.0	60.2	61.9
75	73	38.5	48.6	47.6	47.4	47.2

TABLE 3. VALUES OF EXPONENT n FOR TERGITOL-WATER SYSTEM
[Equation $h\alpha(1/\sigma^n)$]

Solution concentration, p.p.t.t.	Air-bubble rates, bubbles/min.			
	30	100	200	300
10	0.65	0.68	0.70	0.78
35	0.66	0.74	0.89	1.04
75	0.79	0.76	0.76	0.75