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AN EXPERIMENTAL STUDY OF CONVECTION
INITIATION IN DEEP POOLS

by

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ABSTRACT

This experimental study is concerned with the initiation of convection in deep pools (penetration depth is small compared with the fluid depth) and shallow pools (penetration depth is comparable to the fluid depth). Fluids covering a wide range of Prandtl number were either cooled from above through a gas-liquid surface where both surface tension and density forces were expected to contribute to the convection, or heated from below where only the density forces were expected. The study of these two systems simultaneously allowed identification and separation of density and surface tension effects. The results were then used to evaluate current models of convection initiation driven by either density forces, surface tension forces or a combination of both over a wide range of experimental conditions.
INTRODUCTION

The reliable prediction of conditions at the onset of convection in deep pools is of importance to the engineer in designing commercial equipment and to the mathematician and physicist in understanding natural phenomena. Theoretical models predicting this time-dependent physical situation have yet to be developed to the degree where they agree with experimental observation, as is the case for the well-developed example of the onset of density-driven convection from the time-independent, linear gradient. One reason for this could be that the principles successfully used in the linear gradient case are not being interpreted correctly when applied to the deep pool analysis. This lack of understanding appears to stem from an inadequate scope of deep-pool data upon which to base a reliable deep-pool model.

Convection initiation at a fluid-fluid interface can be affected by both surface tension and density forces (Berg and Morig, 1969), which is in contrast to convection initiation at a fluid-solid surface where density forces can be expected to govern the situation. Consequently, the onset of convection driven by density forces can be investigated in a solid-fluid system, while comparison between conditions of convection initiation at a fluid-fluid interface (in this study, a gas-liquid surface) and at a solid-fluid interface can reveal any surface tension influence on initiation at the fluid-fluid interface.

Deep-pool, density-driven convection initiation has been analyzed mathematically by the Quasi-Stationary-Assumption (QSA) Theory developed by Morton (1957), Lick (1964) and Currie (1967) and by the Amplification Theory developed by Foster (1968), Mahler and Schechter (1970) and Gresho and Sani (1971). The QSA model approximates the deep-pool transient density profile by two linear segments (Figure 1) so that the depth (\( \ell \)) contains
Fig. 1. Effective Thermal Depths ($\xi$) for a Step Change in Surface Temperature (Case A) and a Linear Change in Surface Temperature with Time (Case B), Using the Segment Approximation Where: $\xi = 2 \int_0^\infty \left( \frac{T - T_b}{T_s - T_b} \right) dz$
all of the density change. The stability of the system is found by assuming that the growth of the disturbance is fast compared with the rate of change of the density profile with time, i.e., invoking the quasi-static assumption, and proceeding with the stability analysis in a manner similar to the time-independent, linear-gradient problem. The complicating factor in the deep-pool example is that the bulk fluid is considered to affect the stability of the density boundary layer. The critical Rayleigh number, $Ra_\ell$, based on the effective thermal boundary layer depth ($\ell$) is predicted to be in the range of 5 to 32, depending on the boundary conditions. These values are independent of the Prandtl number, $Pr$.

The authors of the Amplification Theory have compared the growth time scales of the initial disturbance with the time scale of the changing density profile and have observed that the quasi-static assumption is invalid for deep-pool density convection. As an alternate approach, they postulated a form of time-dependent initial disturbance in the fluid which was present even before the establishment of the density profile. The equations of motion were integrated to determine the critical conditions (expressed as a time-dependent Rayleigh number, $Ra_t$) necessary for the amplitude of the most preferred wavelength of the initial disturbance to be amplified by a certain factor. The critical amplification factor was necessarily found from experimental observation, since the amplification for observable convection could not be specified theoretically. Consequently, the critical Rayleigh number is predicted to be dependent on the initial conditions of the fluid, which one could expect to change with the site of the fluid. The calculations further show that $Ra_t$ should be dependent on $Pr$ for any given amplification factor.

Experimentally $Ra_t$ and $Ra_\ell$ have been observed to be much greater than predicted by the QSA model (i.e., by factors of 10 to 100). $Ra_\ell$ is normally greater than $Ra_t$ by a factor of 3 to 11, depending upon the boundary conditions (Fig. 1).
Foster (1965) and Spangenberg and Rowland (1961) reported data for the onset of convection produced by evaporation in deep pools of water. Using Amplification Theory, the data could be correlated with a value of $Ra_t$ of 255, and amplification factors in the range of $10^1$ to $10^2$. Foster (1969) heated water from below and found $Ra_t$ in the range of 1000 to 1200, which corresponded to an amplification factor of $10^3$ to $10^4$. Onat and Grigull (1970) heated a number of fluids covering a wide range of Pr and reported $Ra_t$ in the range of 80 to 120, which corresponds to an amplification factor of approximately $10^1$. Several authors have absorbed gases into water and observed the onset of convection. Blair and Quinn (1969) absorbed ammonia, ethyl ether, and sulfur dioxide through a contaminated aqueous surface. $Ra_t$ was reported as 320, corresponding to amplification factors from $10^5$ to $10^6$. Mahler and Schechter (1970) absorbed sulfur dioxide into water with surfaces free of or supporting soluble or insoluble surfactants. The results gave $Ra_t$ in the range of 120 to 300, which corresponded to an amplification factor of $10^3$. Burger (1970) absorbed sulfur dioxide and ethyl ether into water and found that $Ra_t$ of 324 and 280, respectively, would describe the results. One important observation relating to these various studies is that the range in amplification factors corresponding to observable convection suggests that Amplification Theory is not a reliable method for predicting onset conditions.

Deep-pool, surface-tension-driven convection initiation has been analyzed theoretically using an approach similar to the QSA theory. Vidal and Acrivos (1968) extended the earlier work of Pearson (1958), who had investigated the time-independent, linear-gradient, surface-tension-driven problem. The critical value of the Marangoni number $M_t$, based on penetration depth for the deep-pool case was predicted to be 4.0. Brian and Ross (1972) added a stabilizing effect due to the Gibbs adsorption phenomenon at a surface to the Vidal and Acrivos analysis and found that the critical Marangoni number was increased by a factor of approximately 1000 for some strongly adsorbing surfaces.
This latter development predicted values of $M_t$ closer to the experimentally observed values which have been reported by Vivian et al. (1971), and Clark and King (1970).

The broad objective of this paper is to report experimental conditions at the onset of convection at free and fixed surfaces, and in deep and shallow pools, and to analyze these results to give a deeper understanding of the mechanisms of density-driven and surface-tension-driven instability.

**EXPERIMENTATION**

Adverse density and surface tension gradients were produced by either heating the fluid from below or cooling the fluid from above. Heat transfer experiments were chosen rather than mass transfer experiments because of the ease of experimentation, ease of detecting onset of convection by monitoring the liquid surface temperature, and the availability of a wider range of Pr (0.7 to 8500) than in Sc, which would allow investigation of the increase in $Ra_t$ with decreasing Pr which Amplification Theory predicts will occur in this range of Pr.

The design strategy was to produce a one-dimensional temperature field in the fluid, so as to conform with the model used in the theoretical analysis and so as to eliminate horizontal gradients along the liquid surface, which had been found in preliminary studies to have a severe destabilizing effect. This strategy was implemented by using planar heating or cooling surfaces and designing each heat transfer layer with parallel edges and uniform thermal properties, even when the layer was composed of more than one material. Edge effects were designed to be far from the fluid surface.

The liquid container was made from a solid chosen to have similar thermal properties to the test liquids, because of the criteria discussed above. The thermal response of a specimen of Plexiglas showed a thermal conductivity and thermal diffusivity in the range of many organic liquids, and so Plexiglas was
used for the liquid container. The corrosive properties of liquids on Plexiglas and their availability, in addition to the above thermal property considerations, narrowed the choice of liquids to Methanol (Pr=7.7), n-Decane (Pr=13.3), n-Undecane (Pr=17), n-Butanol (Pr=43), n-Hexanol (Pr=64), n-Octanol (Pr=108), Silicone Oils 50 cs (Pr=450), 100 cs (Pr=890), and 1000 cs (Pr=8500). Although air (Pr=0.7) did not satisfy the thermal property criterion, it was used to approximate low-Pr responses.

Because Amplification Theory predicted that the convection onset would be sensitive to the initial conditions of the fluid, the experiments were carried out on a vibration-free table constructed for the purpose of carrying out precision laser experiments, and located at the Lawrence Berkeley Laboratory. This environment was felt to have a very low vibration level.

Greater detail on experimental technique is given elsewhere (Davenport and King, 1972).

Convection Initiation at a Fixed Surface

Relationship Between $Ra_t$ and $Pr$ for the Deep Pool Case. The apparatus to determine $Ra_t$ as a function of $Pr$, for the case where the fluid depth and width were large enough so as not to influence the results, consisted of a Plexiglas cylinder (9 cm i.d., 4.5 cm height) attached to a 750-watt plate heater with an isothermal heating surface. The assembly was fitted to a leveling table so that the heating surface was horizontal. A 1 mil copper-Constantan thermocouple was placed on the surface of the isothermal heating plate, being insulated from it by a thin sheet of mica. The temperature response was recorded over ranges of 50 $\mu$V to 2 mV.

A run was begun by allowing the test liquid to come to thermal equilibrium with the surroundings, as indicated by the change in thermocouple response. A DC heating power supply was adjusted to give an initial high power input, followed by a constant power level to produce a linear increase of the heating
surface temperature with time. Fluid convection initiation was observed by the first deviation from linearity of the surface temperature, as shown in Figure 2.

For a particular fluid, the conditions at the onset of convection were identified by the temperature elevation, \( \Delta T_c \), and the period of conduction, \( t_c \), prior to the onset of convection. Foster (1969) and Onat and Grigull (1970) correlated these critical conditions for the fixed-surface example using the time-dependent Rayleigh number, \( R_{a_t} \), where

\[
R_{a_t} = \left( \frac{\rho g \beta \Delta T (at)^{3/2}}{\mu a} \right) = \left( \frac{\rho g \beta a^{1/2}}{\mu} \right) \Delta T_c^{3/2}
\]  

(1)

For any given fluid in which the physical properties do not vary significantly, the first group in the Rayleigh number in Eq. 1 and the Prandtl number will both be constant. Thus, the experimental conditions can be varied easily by changing the fluid viscosity and the heating rate, which in turn varies the conditions \( \Delta T_c \) and \( t_c \).

The experimental results are plotted in Figures 3 and 4. In Figure 3 the slopes of the lines are \(-3/2\), confirming that \( R_{a_t} \) for a given fluid is independent of heating rate, as would be expected for density-driven convection. The values of the physical properties used to evaluate \( R_{a_t} \) and \( Pr \) for obtaining Figure 4 from Figure 3 are tabulated by Davenport and King (1972).

For air (\( Pr=0.7 \)), even though the temperature profile was two-dimensional near the walls because of the difference in thermal properties between air and Plexiglas, the region in the center of the plate was thought to be nearly one-dimensional. A thin thermocouple was suspended about 1mm above the surface, and this detected the onset of convection much better than the thermocouple on the air-solid surface.

Predicted relationships between \( R_{a_t} \) and \( Pr \) calculated by Foster (1968) using Amplification Theory with constant amplification factors of \( 10^1 \) and \( 10^8 \) are also given in Figure 4. The data show that the apparent amplification factor corresponding
Fig. 2. Response of the Metal-Liquid Surface Temperature During a Fixed-Surface Run.
Fig. 3. Experimentally Determined Conditions for the Onset of Convection for a Linear Temperature Decay at a Fixed Surface in a Deep Pool.

- Methanol, • n-Decane, ▲ n-Butanol, △ n-Octanol, ● Silicone Oil 50 cs., ▲ Silicone Oil 1000 cs.

Bulk liquid temperature at 25.0 ± 0.2°C.
Fig. 4. Experimental Values of Ra_t versus Pr for a Linear Temperature Decay at a Fixed Surface in a Deep Pool.

- Experimental Data
- Foster's (1968) theoretical curve for constant Amplification Factor of $10^1$ and $10^8$. 
to onset decreases as $Pr$ increases. Foster (1969) published experimental results for heating water in a similar apparatus to that used in this report. With $Pr$ of 6.6, he correlated his results with an amplification factor of $10^3$ to $10^5$, which agrees well with the present results.

Onat and Grigull (1970) report values of $Ra_t$ which are lower by approximately a factor of five than the present results, over a range of $Pr$ from 7 to 4400. Their experiments were carried out in similar manner to the present experiments and those of Foster (1969). The lower values of $Ra_t$ could be due to high residual velocity resulting from a short period of rest between runs. Such an effect was noted in our preliminary investigations. In the current experiments the quietest liquid state was used to correspond with the infinitesimal-disturbance concept of the QSA model. Consequently, rest periods between runs were up to 1-1/2 hours for a run lasting 20 seconds to 150 seconds. Runs with longer conduction times before instability were left longer to rest between runs.

Effect of Wall Spacing. As the ratio of fluid width ($D$) to depth ($H$) decreases, an influence of the walls on the incipient convection can be expected. The problem of convection initiation in narrow slots has been examined for the linear-profile, time-independent case, but not for the deep-pool case. Heitz and Westwater (1971) have observed that $Ra_H$ in the time-independent case increased for $D/H$ less than 2.0.

Experiments in the present study observed the effect of $D^2/\alpha t$, which is similar to $D/H$, on $Ra_t$ in the range where the walls were expected to affect the onset conditions on the basis of the time-independent study. Experimental data were obtained by temporarily placing a Plexiglas block with a well-finished face on the heating surface of the large vessel used for the $Ra_t$-vs-$Pr$ experiments. Holes of 5.7 mm and 8.7 mm were drilled into the block to contain the fluid. The results are shown in Figure 5. It can be seen that wall effects are negligible for n-octanol ($Pr=108$) and n-decane ($Pr=13$), provided $\frac{\alpha t}{D^2} < 0.1$. For greater values of $\frac{\alpha t}{D^2}$, $Ra_t$ rises sharply.
Fig. 5. Experimental values of $R_a$ versus width factor $(at/D^2)$ for the fixed-surface, deep-pool case.

n-Octanol:  
- $D = 8.9$ cm  
- $D = 0.87$ cm

n-Decane:  
- $D = 8.9$ cm  
- $D = 0.57$ cm
Effect of a Vibrating Heating Surface on $\text{Ra}_t$. The influence of environmental vibration on convective instability is anticipated for Amplification Theory (Foster, 1969; Gresho and Sani, 1971) and analyses of finite-amplitude disturbances (Schulter et al., 1965; Krishnamurti, 1968). Under different conditions of vibration, the disturbances which initiate convection can be expected to vary in size and nature. The expected result is that as the site of the experiment changes, so does the level of vibration and hence the onset time as reflected by $\text{Ra}_t$. The utility of laboratory experiments to predict $\text{Ra}_t$ in non-laboratory situations is therefore suspect.

Experimental data were obtained by mounting the apparatus used to determine $\text{Ra}_t$ vs. $\text{Pr}$ on top of a vertical sinusoidal vibrator (Davenport, and King, 1972). The frequency ($f$) range was 10 to 10,000 cps and the amplitude ($A$) range was $10^{-7}$ to $10^{-1}$ inches, but these variables were limited in their independence so that $A^2 f^3$ varied by approximately 1000 at most. Different bulk fluid motions were produced by the upper surface of the liquid being either flush with the vessel lid or free of it. The two different wave forms produced in the liquid with free and fixed upper surfaces did not produce results which were significantly different from each other. The exception came at low frequencies and high amplitudes, where the liquid surface was broken for the free surface case, which lowered $\text{Ra}_t$ significantly.

The results showed that as the forcing vibration decreased in frequency and increased in amplitude the onset of convection occurred earlier. The greatest destabilizing effect occurred at approximately 60 cps and 0.01 inches amplitude, where the value of $\text{Ra}_t$ was lowered by 40% from the value obtained with a very low vibration environment.

The conclusion from the results is that vigorous vibration lowers the onset time as predicted, but that under moderate conditions $\text{Ra}_t$ will probably change by no more than 15% as a result of variation in environmental vibration level. This is to be compared with the random variation of about 10% in $\text{Ra}_t$ which occurred for the heating experiments carried out on the vibration-free table, where the vibration level was constant.
The stability of a vertically oscillating horizontal layer of fluid supporting an adverse linear density gradient has been analyzed by Gresho and Sani (1970). They predict a decrease in Rayleigh number at low frequencies and large amplitudes, which is only significant for very thin layers of fluid (high Froude number). The experimental conditions discussed above are well out of this range, and no observable decrease would be anticipated.
Effect of the Shape of the Density Profile in the Fluid on \( \text{Ra}_t \). Amplification Theory (Foster, 1968) predicts that \( \text{Ra}_t \) will be dependent on the nature of the surface-temperature-vs.-time relationship. \( \text{Ra}_t \) for a step change was calculated to be about three times lower than that for a linear time decay for the same amplification factor.

The experimentation used to investigate these predictions was similar to that used to determine \( \text{Ra}_t \) vs. \( \text{Pr} \), with the exception that the heating at the surface in this case was produced by a thermoelectric heat pump rather than by a plate heater. The variation of the heated surface temperature with time was controlled by monitoring the current to the heat pump. In this way curve shapes ranging from a step function to a linear time increase were produced.

The correlating parameter used to describe the shape of the surface-temperature-vs.-time profile was

\[
S = \frac{\text{Area under the surface temperature curve up to } (\Delta T_c, t_c)}{\text{Area given by } \Delta T_c \text{ times } t_c} = \frac{\int_0^{t_c} (T_S - T_b)dt}{\Delta T_c \cdot t_c}
\]

\( S \) is 1.0 for a step change and 0.5 for a linear change with time. This was the simplest parameter that could describe the surface temperature curve. It has the drawback that several curve shapes can be described by a given value of \( S \).

The results for n-octanol have been plotted in Figure 6. They show that \( \text{Ra}_t \) decreases as \( S \) increases in the range 0.5<\( S \)<0.9. Similar dependence of \( \text{Ra}_t \) on \( S \) has been found for n-undecane, n-butanol, n-hexanol, and n-octanol at a free surface; these results have been reported in Davenport and King (1972).
Fig. 6. $R_{a_t}$ versus Shape Factor ($S$) for n-Octanol at a Fixed Surface in a Deep Pool.
Effect of Fluid Depth on \( Ra_t \). The predicted effect of fluid depth on convection initiation for the QSA Theory was given by Currie (1967). It indicates that \( Ra_t \) should increase as the ratio of effective density-layer depth (\( l \) in Figure 1) to fluid depth increases. The two asymptotic conditions were the time-dependent, deep-pool case and the time-independent, shallow-pool case.

The transition from the deep-pool to the shallow-pool mechanism was observed experimentally by varying the fluid depth and the heating rate. The fluid depth was changed by temporarily placing, but not glueing, a smaller diameter Plexiglas annulus on the heating surface in the vessel used for the \( Ra_t \) vs. \( Pr \) determination. A large aluminum cylinder, 5 cm high and slightly less than 9 cm wide, was placed on top of the annulus to keep the temperature of the upper surface of the liquid constant. A thermocouple attached to the base of the metal cylinder showed that the upper surface temperature varied by less than 5% of the total temperature drop across the fluid.

The correlating parameter to show the change in critical Rayleigh number, based on either fluid depth, \( Ra_H \), or penetration depth, \( Ra_t \), at shallow depths and constant \( Pr \) is \( \frac{at}{H^2} \). The results have been plotted on Figures 7 and 8 for three fluids and for liquid depths as small as 2.05 mm. The transition from the deep-pool mechanism to the shallow-pool mechanism is clearly seen in Figure 7. The shallow pool data converge on an asymptote which is independent of \( Pr \) and heating rate, and dependent on fluid depth (\( Ra_H = 1700 \)). The deep pool asymptotes are independent of \( H \) but are dependent on \( Pr \), as reported in Figure 4.

There are no previously published shallow-pool experimental results for constant temperature at the upper surface. However, some shallow-pool experimental results for an adiabatic upper surface have been reported by Soberman (1958). His values of \( Ra_H \) are below the current results and lower than what was theoretically predicted by Currie (1967) using the QSA approach. Foster (1969) reported values for convection onset in shallow pools with
Fig. 7. Experimental Values of Ra, versus Depth Factor (at/H²) Showing the Transition from Deep- to Shallow-Pool Conditions for the Fixed-Surface Case.

X Methanol, Pr = 7.6, Butanol, Pr = 43, Δ Silicone Oil 50 cs, Pr = 450. † Foster's (1969) datum point for water.
Fig. 8. Experimental Values of Ra_H versus Depth Factor (at/H^2) for the Shallow-Pool Data.

X Methanol Pr = 7.6
● Butanol Pr = 43
◇ Silicone Oil 50 cs Pr = 465
a linear increase in the lower surface temperature with time. The liquid at the upper surface was open to the air and so was closer to an insulated surface than to an isothermal surface. The critical Rayleigh numbers are estimated as $Ra_t = 10,800 - 12,700$ at $\frac{H^2}{L} = 1.8$, as shown in Figure 7.

Convection Initiation at a Free Surface

Deep-Pool Convection Initiation. Previous experimentation with free surfaces has used evaporation to provide cooling at the liquid surface, with and without air blown over the surface. The disadvantages of that approach include two-dimensional temperature profiles on the liquid surface, variable disturbance level if an air draught is used, a limited operating range if no air draught is used, and possible wave formation if a mechanical system is employed to start the evaporation. These disadvantages are inherent in evaporation systems and can be overcome by cooling with a heat sink over the liquid surface, provided the resistance to heat transfer below the cold surface can be made one-dimensional.

The apparatus having this one-dimensional character is shown in Figure 9. A cooler with an isothermal cold plate (diameter 50 mm) rested on foam supports approximately 1.5 mm thick, which in turn rested on top of the walls of a hollow Plexiglas cylinder (36.5 mm i.d., 7 mm wall thickness, 36.5 mm depth). These dimensions were found experimentally to remove the edge effects associated with heat transfer across an air gap away from the liquid surface. The foam was the best compromise for a material with thermal properties similar to air and yet with the mechanical strength to support the cooler and isolate the air gap between the cooler and the liquid from external air convection currents. The liquid surface was raised to the level of the top of the Plexiglas walls so that there was no meniscus, in keeping with the planar surface strategy. A thermocouple (1 mil, copper-Constantan) supported from below the liquid by
Fig. 9. Schematic Diagram of Apparatus Used to Determine Convection Conditions of a Gas-Liquid Surface.
glass leads (1.6 mm diameter), lay on the liquid surface to record its temperature. The thermocouple was set in the plane of the top of the walls, and the liquid was raised until the thermocouple was in the plane of the liquid surface.

The cooler was required to provide an isothermal cold plate with a temperature which could be controlled and varied. The two cooling systems chosen were a thermoelectric cooler and an enclosed cold-liquid spray chamber. The spray chamber had the advantage of being able to produce a variety of temperature profiles on the liquid surface, from a linear time decay through to a step change, whereas the thermoelectric cooler was limited to a linear decay in surface temperature. However, the sprayer had the disadvantage of introducing variable vibrations into the system from the spraying mechanism. The thermoelectric cooler having no moving parts was essentially vibration-free, and it was used to represent the quietest possible conditions for free-surface convection.

The temperature response of the gas-liquid surface showed a much sharper transition at the onset of convection than did the fixed-surface response. The temperature gradient immediately after the onset of convection reversed sign, and the temperature oscillated from that point onward. The difference between the fixed- and free-surface response lay in the high thermal capacity and conductivity of the metal and the low thermal capacity of the air relative to the liquid phase. Thus the metal surface dominated the temperature response in the fixed surface case, and the liquid phase dominated in the free surface case.

The thermoelectric cooler was used to produce free-surface results for comparison with the fixed-surface experiments. The liquids were selected with the constraint that they be non-volatile, in addition to the thermal and corrosive compatibility conditions discussed before. n-Decane (Pr=13.3), n-butanol (Pr=43), n-octanol (Pr=108), and 100 cs silicone oil (Pr=900) were tested. The results for n-decane have been plotted in Figure 10 for both the free and fixed surfaces. Results for the
Fig. 10. Experimental Conditions of Onset of Convection in n-Decane (Pr = 13) for a Linear Surface-Temperature Decay in a Deep Pool Using the Thermoelectric Cooler.

X Fixed-Surface Data, • Free-Surface Data,
--Free-Surface Predictions of Mahler (1970) Assuming Same Amplification Factor as for Fixed-Surface Results.
other liquids were similar (Davenport and King, 1972) except under certain conditions where the free-surface results were slightly lower than the fixed-surface results. These conditions were for high driving force, where the character of the cooling surface was suspected to have some two-dimensional character. This was partially verified by rebuilding the thermal insulation system which was used to keep the thermoelectric cooler cold surface isothermal and observing that the onset times at high driving forces increased with the improved cooling surface. The other condition under which the free-surface results were slightly lower was at long conduction times, where it was suspected that the difference in thermal properties between the Plexiglas and the liquid and heat losses through the edge of the apparatus became significant enough to produce two-dimensional character into the surface temperature. Preliminary investigation showed that two-dimensional gradients in surface temperature could lower the onset time appreciably.

The results in Figure 10 and for the other liquids show that the convection is density-driven, since the slope is $-3/2$, as opposed to the slope of $-1/2$ which is predicted for surface-tension-driven convection. Furthermore, these results show that convection initiation is independent of the shear conditions at the heating or cooling boundary, in a vertical, one-dimensional, deep-pool density field. This is in contradistinction to Amplification and QSA theoretical predictions, where the difference between the fixed- and free-surface Rayleigh numbers is a factor of 2 to 3.5. Even when the free-surface data fell below the fixed-surface data in the present work, they were always well above the limits expected from either of these theories (see Figure 10).

Effect of Shape Factor and Surface Waves on $Ra_l$. The spray cooler was used to produce shapes of surface-temperature-vs.-time curves on the free surface ranging from a step change to a linear change with time. These results (see Davenport and King, 1972) had a similar shape to the fixed-surface results plotted in
Figure 6. The influence on $Ra_t$ of the surface waves from the spray cooler can be seen in Figure 11, where the results of the spray cooler and thermoelectric cooler have been compared for a linear decrease of surface temperature with time.

Comparison with Previous Free-Surface Results. Foster (1965) identified his experiments where water was evaporated from stagnant pools as a case of linear decay of surface temperature with time ($S=0.5$). Vidal and Acrivos (1968) observed that evaporation from stagnant pools could be better described by constant flux at the evaporation surface. The surface-temperature history with constant flux would correspond to $S=0.67$, which agrees closely with Foster's reported record of $T_s$ vs. $t$. Foster's results of $Ra_t=255$ for evaporation of water ($Pr=6.6$) with $S=0.67$ agree well with the present results for n-decane ($Pr=13.3$) using the spray cooler (Davenport and King, 1972). The fact that his result compares better with the spray-cooler results than with the thermoelectric cooler suggests the possibility of surface-wave influence in Foster's experiments.

Clark and King (1970) published results for evaporation of carbon disulfide from tridecane solvent. They observed possible density-driven convection at high $CS_2$ mole fraction corresponding to $Ra_t=285$, $S=0.62$, for thermal-driven convection at $Pr=27$. This compares favorably with the spray-cooler results for n-butanol ($Pr=43$) and n-undecane ($Pr=17$), suggesting the importance of surface waves or other destabilizing influences in the horizontally flowing liquid.

Ball and Himmelblau (1967) identified density-driven convection for the absorption of carbon dioxide into a laminar flowing aqueous stream. The mass-transfer-driven Rayleigh number, $Ra_t$, was reported as 785 with $Sc=300$, but the thermal-driven Rayleigh number was not reported. Since the absorption was exothermic, the thermal gradient would tend to stabilize the system, which may account for the high value of $Ra_t$ when compared with the present results where no stabilizing influence was present.
Fig. 11. Comparison of Values of \( \text{Ra} \) at a Free Surface Determined with the Spray Cooler (\( \times \)) and the Thermoelectric Cooler (\( \circ \)), for a Linear Surface-Temperature Decay (\( S = 0.5 \)).
Shallow-Pool Convection Initiation. The shallow-pool apparatus consisted of a Plexiglas annulus attached to a copper cylinder. The annulus was 36.5 mm i.d., 7 mm wall thickness, and the height varied from 2.06 mm to 11.9 mm. The copper cylinder was 5 cm in diameter and about 4 cm long. The glass thermocouple leads were vertical and were introduced through the copper base rather than through the Plexiglas walls, because the Plexiglas was too short. The copper block was used to keep the lower liquid surface temperature constant so that the data could be compared with the fixed-surface, deep-pool results.

Both the spray cooler and the thermoelectric cooler were used to determine the onset conditions in shallow pools. The surface temperature either decreased linearly with time, so that the results could be compared with the fixed-surface results, or the shape factor, $S$, was varied so as to show its influence in shallow-pool convection initiation. n-Butanol and n-octanol were used as liquids.

The free-surface, shallow-pool data obtained with the thermoelectric cooler have been plotted in Figure 12. The spray-cooler data were similar. The results show that there is no significant difference between the fixed-and the free-surface data in the range of $\frac{\alpha t}{H^2}$ used in these experiments (compare Figure 8). The conclusion is that the mechanism for convection initiation at these two surfaces in the shallow-pool case is the same, and therefore that convection onset under these experimental conditions for free-surface shallow pools is density-driven. This is true even for thin liquid films (2 mm thick) and high values of Marangoni number, $M_T$ up to 10,500 and $M_H$ up to 5,200.

The comparison between the spray cooler and thermoelectric cooler results (Figure 12) shows that the influence of the vibrations is not important in shallow-pool convection initiation. This is probably due to the size of waves in this experimental apparatus being reduced by the shallow depth of liquid.
Fig. 12. Experimental Values of Ra, at a Free Surface as a Function of Depth Factor $H(\alpha t/H^2)$ for n-Butanol Using the Thermoelectric Cooler.

Fluid Depth: ○ 5.0 mm, □ 3.3 mm, ● 2.16 mm.
Spray Cooler Data for $0.5 < S < 0.6$: △ n-Butanol,
□ n-Octanol; -- -- Boundaries of Spray Cooler Data for $0.5 < S < 0.8$. 
DISCUSSION OF RESULTS

Evaluation of Models Describing the Onset of Time-Dependent, Density-Driven Convection in Deep Pools

The purpose of this section is to evaluate the strengths and weaknesses of the QSA and Amplification theories for describing density-driven convection initiation in a wide, deep pool. The shallow-pool, time-independent model has been well established theoretically and experimentally and can be used as a reference for judging the validity of the marginal-stability assumption when applied to non-linear density profiles. Mathematical comparison between the QSA and Amplification theories by Robinson (1965), Foster (1968), and Gresho and Sani (1971) has highlighted features of both theories and rejected the QSA approach as invalid for deep-pool analysis. Such a comparison is suspect since the form of the developing disturbance has not been established and therefore has to be assumed. This assumption is the basis for the "mathematical" comparison. The analysis in this section compares the predictions of the two theories with experimental evidence. Such a comparison has no assumed basis as for the mathematical comparison, and would therefore appear to be more practical in its evaluation.

The equations of motion, linearized around zero fluid velocity and the conduction profile $\frac{\partial T}{\partial z}$, are shown below (refer to Chandreskhar, 1962).

\[
\frac{\partial^2 T}{\partial z^2} = \frac{\partial T}{\partial t} \tag{2}
\]

\[
\left(\frac{\partial}{\partial t} - \nu^2 \right) \Theta = -\sqrt{Ra} \nu \frac{\partial T}{\partial z} \tag{3}
\]

\[
\left(\frac{1}{Pr} \frac{\partial}{\partial t}\right) \nu^2 \nu = -\sqrt{Ra} \nu^2 \Theta \tag{4}
\]
The assumption of marginal stability in the QSA approach causes the time derivatives to be eliminated from these above equations.

\( \text{Ra}_t \) Independent of Heating Rate. Experimental results reported herein and the results of Foster (1965, 1969), Blair and Quinn (1969) and Onat and Grigull (1970) verify this prediction of both QSA and Amplification theories.

Variation of \( \text{Ra}_l \) with the Shape of the Density Profile of the Liquid. Currie (1967), using QSA theory, predicted that \( \text{Ra}_l \), where \( l \) is the thermal depth in Figure 1, was independent of the shape of the density profile in the fluid. Foster (1968), using Amplification theory, studied the cases of a step change in the surface temperature (Case A), and a linear change of surface temperature with time (Case B). From Figures 3 and 5 of his paper, the ratio of the time-dependent Rayleigh number, \( \text{Ra}_t \), at the large-Pr asymptote and an amplification factor of \( 10^8 \) for the two cases was evaluated as 3.0. The ratio of effective thermal layer depth (\( l \)) to penetration depth (\( \sqrt{\alpha t} \)) was different for each case, as seen in Figure 1, but after \( \text{Ra}_t \) was transformed to \( \text{Ra}_l \), the values of \( \text{Ra}_l \) for each case at high \( Pr \) were approximately equal.

The experimental data measuring the effect of shape of density profile on \( \text{Ra}_l \) have been reported for n-octanol, a high-Pr fluid, in Figure 6, and for other fluids by Davenport and King (1972). Values of \( \text{Ra}_l \) were calculated from these data by approximating the ratio of \( l \) to \( \sqrt{\alpha t} \) as a function of density-profile shape in the fluid, which in turn is dependent upon the surface-temperature-vs.-time relationship and the depth of the fluid (see Davenport and King, 1972). The surface-temperature-vs.-time curve can be described to a first approximation by the shape factor \( S \), defined earlier. In order to calculate the above length ratio as a function of \( S \), the temperature profile in the fluid has to be expressed as a function of \( S \). An observation of the mathematical equations involved shows that this is
not possible for the general case. However, the family of curves described by \( T_s = kt^{n/2} \) can be used to find values of \( S \) and values of the length ratio for this family of curves. This relation can be used generally to give the value of the length ratio for any \( T_s \)-vs.-\( t \) function, if it is assumed that the values of \( S \) fall within a narrow range about the values calculated using the \( T_s = kn^{n/2} \) family of curves. The analysis was based on this assumption, since there did not appear to be any other reasonable alternative to find a general relationship between the length ratio and \( S \).

\[ Ra_t \text{-vs.-} S \] data for the fixed and free surfaces are replotted as \( Ra_x \) versus \( S \) on Figure 13. For \( Pr = 108 \) the spray-cooler free-surface results are essentially the same as the thermoelectric cooler free-surface and fixed-surface results, as was shown in Figure 11. The results show that \( Ra_x \) is essentially independent of \( S \) at this high value of \( Pr \), as predicted from both theories. The scatter is probably due to the assumed generalized relation between the length ratio and \( S \). The scatter encountered for the linear time decay of \( T_s (S = 0.5) \) is shown for comparison.

**Variation of \( Ra \) with the Ratio of Thermal Depth to Fluid Depth.** Currie (1967) used QSA theory to calculate \( Ra_H \) as a function of \( \ell/H \) for a fluid bounded by two perfectly conducting surfaces. His results are replotted in the form \( Ra_x \) vs. \( \frac{\alpha t}{H^2} \) in Figure 14.

There are no calculations published showing \( Ra_t \) as a function of \( \frac{\alpha t}{H^2} \) using Amplification theory. However, weaknesses of this theory for shallow pools have been discussed in Davenport and King (1972).

The fixed-surface experimental values of \( Ra_t \) were transformed into \( Ra_x \) using the relationship between the length ratio \( \ell/\sqrt{\alpha t} \) and \( \frac{\alpha t}{H^2} \) for a linear surface temperature change with time with a finite fluid depth (Davenport and King, 1972). \( Ra_x \) vs. \( \frac{\alpha t}{H^2} \) is plotted in Figure 15. The asymptotic value at high values of \( \frac{\alpha t}{H^2} \) (very shallow pools) is \( Pr \)-independent and is in the range of \( Ra_x = 1700 \), which is the time-independent solution for a linear gradient with fixed, perfectly conducting surfaces. This is also the solution for \( Ra_x \) where the marginal-stability assumption has been shown to be valid (Chandreskhar, 1962). Figure 14
Fig. 13. Experimental Values of $Ra_e$ for n-Octanol ($Pr = 108$) as a Function of the Surface-Temperature Shape Factor $S$ in the Deep-Pool Case.

- Fixed Surface
- X Free Surface (data taken with spray cooler)
- Scatter for Linear Decay of Surface Temperature Using Thermoelectric Cooler
Fig. 14. Experimental Values of Ra, as a Function of the Depth Factor (at/H^2) for the Fixed-Surface Case, with Linear Decay of Surface Temperature.

- n-Butanol, Pr = 43
- Silicone Oil 50 cs, Pr = 465
- Silicone Oil 1000 cs, Pr = 8500
- Methanol, Pr = 7.6
shows that, for a fluid with an infinite value of Pr, \( \text{Ra}_t \) is relatively independent of \( \frac{at}{H^2} \). This suggests that the marginal-stability assumption is valid over all pool depths for infinite-Pr fluids, if the length scale in the equations of motion is taken as \( \lambda \), as defined in Figure 1. It would not follow that the marginal-stability assumption is true for all Pr, since \( \text{Ra}_t \) is found experimentally to be Pr-dependent, contrary to the QSA-theory predictions using the marginal-stability assumption.

Despite this apparent validity of the marginal-stability assumption for high-Pr fluids, it can be seen from Figure 14 that QSA results of Currie (1967) underestimate the deep-pool experimental results by almost a factor of 40. The QSA theory predicts that the presence of the bulk fluid in the deep-pool case causes the thermal layer to be less stable, which from the experimental results appears to be incorrect.

The numerical relationship between the length ratio \( \lambda/\sqrt{\alpha t} \) and \( \frac{at}{H^2} \) becomes independent of S at high values of \( \frac{at}{H^2} \). This fact corresponds with the experimental results with the spray cooler for shallow-pool convection (see Davenport and King, 1972), where the values of \( \text{Ra}_t \) at high values of \( \frac{at}{H^2} \) appeared to be independent of S for the range 0.5 < S < 0.8. This may also explain why Foster's (1969) estimated shallow-pool datum point for water with an adiabatic non-heating surface falls close to the results of present work where the non-heating surface temperature was held constant (see Figure 7). The temperature profiles in each case would be different, but the pools are shallow enough so that this difference is not important.

Influence of Prandtl Number on \( \text{Ra}_t \). Directly through the quasi-stationary assumption the QSA approach predicts the critical Rayleigh number \( \text{Ra}_\lambda \) or \( \text{Ra}_t \) to be independent of Pr. Amplification theory assumes the time-dependent terms in the equation of motion to have equal weight with other terms. Consequently, the amplification time is a function of the Prandtl number, with two asymptotic solutions. At low Pr the term \( \frac{1}{Pr} \frac{\partial}{\partial t} \) in the
equations of motion becomes increasingly important, so that in the limit of very low \( Pr \) the equations of motion are made dimensionless through the group \( Ra_t \cdot Pr \). At high values of \( Pr \), the converse is true and only \( Ra_t \) is needed to non-dimensionalize the equations of motion. Consequently, the convection onset conditions would be expected to be correlated by \( Ra_t = \text{constant} \) and \( Ra_t \cdot Pr = \text{constant} \) at extreme values of \( Pr \). The significant difference between these two asymptotes is that \( Ra_t \cdot Pr \) has no viscosity term in it while \( Ra_t \) does. Thus, constant \( Ra_t \cdot Pr \) shows the region where only conduction is the important stabilizing force, and constant \( Ra_t \) shows the region where both conduction and viscosity are important stabilizing forces.

The experimental fixed-surface results plotted on Figure 4 show that \( Ra_t \), for a linear change of \( T_s - vs. - t \), increases as \( Pr \) decreases, which is in qualitative agreement with Amplification Theory, but not QSA Theory. As is shown in Figure 15, these data are fitted well with the two asymptotes discussed above, but are poorly fitted with a constant amplification factor \( w \) (see Figure 4); for the two asymptotes \( Ra_t = 350 \ (w=10^2) \) and \( Ra_t \cdot Pr = 7200 \ (w=10^{20}) \).

Effect of Rigidity of the Heat Transfer Surface on \( Ra_t \). Both QSA and Amplification theories use the velocity perturbation \( (v) \) expansion form \( v = g(x,y,t) \cdot f(z) \), where \( f(z) \) is the vertical component and is a continuous function of \( z \) through the fluid, satisfying the shear boundary conditions at the upper and lower fluid surfaces. The function for \( f(z) \) at a free surface will differ from that at a fixed surface because of the differences in rigidity between the two boundaries. It is from this reasoning that both theories predict that \( Ra_t \), or \( Ra_t' \), at a fixed surface will be higher than that at a free surface. This prediction has been shown to be valid for the time-independent, linear-gradient case by Berg and Palmer (1971) and Silveston (1958) in experiments made with different shear conditions.
Fig. 15. Experimental Values of $Ra_t$ for a Linear Surface Temperature Decay at a Fixed Surface as a Function of $Pr$ Showing the Two Experimentally Fitted Asymptotes.
The experimental evidence in this paper shows that with deep-pool, penetration-type density profiles and with low external vibration, the degree of rigidity at the heat-transfer boundary has no influence on the value of $Ra_c$ for convection initiation (Figure 10). These results, together with the interferometric photos of Onat and Grigull (1970), indicate that the first motion occurs away from the surface in the bulk of the fluid, and more specifically at the bulk edge of the density boundary layer. This initial motion is apparently not strong enough to penetrate the density boundary layer and "feel" the degree of rigidity of the heat transfer surface. Both the QSA and Amplification theories incorrectly assume a form of initial motion which is distributed throughout the density boundary layer and influenced by the rigidity of the heat transfer surface.

This conclusion differs from the conclusion and experimental results of Mahler and Schechter (1970). They observed that the onset time at a contaminated aqueous surface, defined as a fixed surface, was almost double that at a non-contaminated surface, defined as a free surface. A similar conclusion was reached by Blair and Quinn (1969). Sulfur dioxide was absorbed into water to produce the unstable density gradient in both studies. Blair and Quinn observed that when a wire mesh was held across the contaminated aqueous surface the onset times were the same as those without the wire mesh, after the absorption rate had been corrected for the reduction in transfer area by the wire mesh. They concluded that this showed that a contaminated aqueous surface acted like a fixed surface. When the water surface was partially covered with natural contaminant (insoluble surfactant) and partially covered with oleic acid (soluble surfactant), the onset time under the contaminant was greater, and the forms of motion under the two portions of surface were different; this was attributed to differences in the lateral rigidity of the surface. Thus, Blair and Quinn concluded that the less rigid the surface, the earlier the onset time. Mahler and Schechter extended this work by observing the effect of different types of surfactants.
In view of the present findings that $Ra_t$ for a free-surface under low external vibration conditions is the same as that for a fixed surface, it may be that there was a side effect in the experiments of Blair and Quinn (1969) and Mahler and Schechter (1970) which lowered the onset time as the surface rigidity was decreased. Surface waves at a gas-liquid interface can lower the onset times. To demonstrate this in the present work several liquids were tested with the spray-cooler apparatus to observe the influence of surface-wave action on a free surface. The values of $Ra_t$ for a linear time decay of surface temperature, $S = 0.5$, using the spray cooler were compared with the results using the thermoelectric cooler, as was shown in Figure 11. The influence of surface waves appeared greatest for the less viscous fluids, probably because of the increased surface deformation and the decreased depth of the density boundary layer at onset in less viscous fluids. The spray cooler generated surface waves and produced onset times in n-decane which were approximately 50% of those produced by the vibration-free thermoelectric cooler.

In the sulfur dioxide mass-transfer experiments, both the viscosity and the density boundary layer thicknesses were in the range where the heat transfer results had shown that surface waves can be important. Surface waves could have been generated in those experiments either by building vibrations or by the mechanical action used to increase the sulfur dioxide pressure suddenly to approximate a step change, especially since the instability occurred between 10 to 20 seconds after this mechanical action. The magnitude of the surface waves would be expected to decrease as the surface rigidity increased. Thus, the presence of surfactants would lengthen onset times because they increase the surface rigidity and dampen the surface wave action.

The shallow-pool data for free and fixed surfaces where the temperature profiles were almost linear were expected to show an effect of difference in wall-shear conditions, as in the time-independent studies. This difference was not observed (compare Figures 8 and 12), possibly because of the uncertainty in the liquid depth in the free-surface experiments, or possibly because $\frac{\partial \theta}{\partial t}$ was not high enough to be in the truly time-dependent regime.
Effect of Vibration on $\text{Ra}_t$. Both the QSA (through the finite amplitude approach—Krishnamurti, 1968) and Amplification theories predict that a shorter onset time will occur for larger initial disturbances in the fluid. The results on Figure 14 indicate that the onset of convection on the vibration-free table for high $\text{Pr}$ fluids in either shallow or deep pools corresponds to the predictions of the linear-profile and an infinitesimal-amplitude disturbance approach. In this light, the time for amplification of the disturbance to an observable scale in a high $\text{Pr}$ fluid seems negligible compared to the time to reach marginal instability. Consequently, Amplification theory does not seem applicable in the high $\text{Pr}$ case. The lowering of $\text{Ra}_t$ by up to 40% in the vibrating high $\text{Pr}$ fluid would appear superficially to correspond with the increased destabilizing effect of a finite-amplitude disturbance. The interpretation of these results in terms of finite amplitude disturbances has been discussed elsewhere (Davenport, 1973).

The present study did not investigate the effect of vibration on low $\text{Pr}$ fluids beyond the observation that $\text{Ra}_t$ in methanol ($\text{Pr} = 6.6$) and $n$-octanol ($\text{Pr} = 108$) were decreased by approximately the same percentage under identical vibration conditions.

Comparison of Mass-Transfer Results with Heat-Transfer Results. Both QSA and Amplification theory predictions do not distinguish between adverse density gradients produced by heat or mass transfer. The relevant thermal or mass diffusivity and the coefficient of expansion due to a change in either temperature or composition in the definitions of $\text{Ra}_t$ and $\text{Pr}$ account for the nature of the density gradient.

The experimental results using gas absorption or desorption in aqueous systems have involved a step change in density at the gas-liquid surface, and either a clean or a contaminated aqueous surface. A summary of the results with contaminated surfaces which have been identified as density-driven convection is given in Table 1; as discussed above, these should be appropriate for comparison with the present heat-transfer results, since surface waves are minimized. Generally, the $\text{Ra}_t$ are higher than the heat-transfer, high-Pr value of $\text{Ra}_t$, which is 1400 (see Figure 13).
Table 1. \( R_{t} \), \( R_{f} \) Determined from Aqueous-Gas Absorption/Desorption Experiments Involving a Step Change in Surface Conditions and a Contaminated Aqueous Surface.

<table>
<thead>
<tr>
<th>Investigator</th>
<th>( R_{t} )</th>
<th>( R_{f} )</th>
<th>Gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blair &amp; Quinn (1969)</td>
<td>350</td>
<td>4000</td>
<td>Ethyl Ether - desorption</td>
</tr>
<tr>
<td>&quot; &quot; &quot; &quot;</td>
<td>320</td>
<td>3650</td>
<td>Ammonia - desorption</td>
</tr>
<tr>
<td>&quot; &quot; &quot; &quot;</td>
<td>300</td>
<td>3400</td>
<td>Sulfur Dioxide - absorption</td>
</tr>
<tr>
<td>&quot; &quot; &quot; &quot;</td>
<td>210</td>
<td>2400</td>
<td>Carbon Dioxide - absorption</td>
</tr>
<tr>
<td>Mahler &amp; Schechter (1970)</td>
<td>290</td>
<td>3300</td>
<td>Sulfur Dioxide - absorption</td>
</tr>
<tr>
<td>Plevan &amp; Quinn (1966)</td>
<td>140</td>
<td>1600</td>
<td>Carbon Dioxide - absorption</td>
</tr>
<tr>
<td>Burger (1970)</td>
<td>320</td>
<td>3650</td>
<td>Sulfur Dioxide - absorption</td>
</tr>
<tr>
<td>&quot; &quot;</td>
<td>270</td>
<td>3100</td>
<td>Ethyl Ether - desorption</td>
</tr>
</tbody>
</table>
This could be due to the difference in detecting the onset conditions. In the heat-transfer experiments the surface temperature was monitored, while in the mass-transfer experiments the surface flux was monitored.

Evaluation of Models Describing the Onset of Time-Dependent, Surface-Tension-Driven Convection in Deep Pools

The free-surface convection-initiation results (Figure 12) showed no surface-tension influence, even to high values of the Marangoni number \( M_t = 10,500, M_H = 5,200 \). This was in the absence of meniscus effects, surface-temperature gradients, and surface waves. This result is in contradistinction to the predictions of QSA theory developed by Vidal and Acrivos (1968), who proposed a Marangoni number \( M_t = 4 \) as the critical value in the deep-pool case. The Gibbs-adsorption modification of Brian and Ross (1972) should not change this deep-pool solution significantly for these experimental conditions.

This discrepancy between experiment and theory appears to be connected with the observation of the equivalence of convection initiation at free and fixed surfaces in deep pools. Recalling the earlier analysis, it appears that the incipient convection is confined to the bulk edge of the density boundary layer and consequently does not feel the rigidity of the transfer surface. Thus, there is apparently no shear variation at the surface due to the incipient convection and consequently no surface deformation. Provided that there is no surface temperature gradient, local surface disturbance or meniscus, there will be no surface-tension gradients along the surface, and hence surface-tension effects will be absent from the initiation mechanism.

Adherence to these experimental conditions appears to be the difference between the present deep-pool results and the
previously published work on convection initiation in thin films of evaporating, pure organic liquids. Berg et al. (1965) and Vidal and Acrivos (1968) found convection initiation at $Ra_H$ much below 1700 for evaporating films and so ascribed surface-tension forces as the driving force. Similar results to those occurred in our laboratory when a temperature gradient or a meniscus existed along the liquid surface, such as can be expected in evaporation experiments. When the surface gradient and meniscus were eliminated, the results were similar to the fixed-surface results. Mitchell and Quinn (1966) observed surface-tension-driven motion in thin films of liquid when a temperature gradient, produced by a point source, existed along the curved surface of the liquid. As in the case of the evaporating pools, the liquid had a meniscus. Thus, previously published results for thin-film, free-surface, thermal convection produced by evaporation did not conform to the theoretical models used to describe surface-tension-driven and density-driven convection, because of the presence of destabilizing effects other than the adverse temperature profile through the fluid.

The effects of surface waves and meniscuses have shown that deformed surfaces and surface-tension-gradient effects can lower the critical value of $Ra_t$. Furthermore, in cases where surface-tension effects have appeared to dominate the onset of convection -- i.e., Clark and King (1970), Brown and King (1969), Blair and Quinn (1969), Vivian et al. (1971) -- surface waves have been present and from the above discussion appear to be the important disturbance. These results have been analyzed by Davenport and King (1972), and it is observed that the critical Marangoni number in such cases is in the range of 3000 to 8500, and that the Marangoni number decreases as the surface wave action increases.

Ellis and Biddulph (1966) studied the effect of surface-tension forces on the amplitude of waves on a laminar-flowing water stream which was absorbing either acetone or methanol. They observed that the amplitude of the waves increased by an order of magnitude when the driving force for mass transfer
passed a critical value. The authors explained this phenomenon by postulating that the solute absorption across the wavy inter-
face caused sites of low surface tension, and consequently
high surface-tension gradients along the surface which caused
a rapid spreading of surface liquid. This resulted in exposure
of bulk liquid to the surface in the center of the site, which
increased the surface tension in the exposed region. This caused
an annulus of low surface tension fluid and a tendency to re-
verse direction of flow. The momentum associated with the
spreading and reversal of the low-surface-tension fluid re-
sulted in a surface ripple.

The above experimental observations suggest that surface
waves are important and necessary for deep-pool, surface-tension-
driven convection initiation. Convection onset involving density
forces and surface-tension forces for the case of the time-
independent, linear profile in the fluid appears to be in contrast
to this absolute need of surface motion. In the analysis of
this case, Pearson (1958) and Nield (1964) assumed that the
important disturbance for initiation occurs throughout the
fluid layer. Their predictions have been experimentally veri-
fied by Berg and Palmer (1971) and Koshmeider (1967). In a
similar manner, Zeren and Reynolds (1972) observed that the
onset of convection from a linear temperature gradient through
the fluid in a water-benzene system corresponded with their
calculations for the density-surface tension coupling action
in a fluid-fluid system.

The possible difference between the linear-profile time-
independent case, where experiment agrees with theory, and the
deep-pool time-dependent case is that in the time-independent case,
the bulk fluid motion produces immediate surface renewal and
thus immediate surface-tension influence, whereas in the time-
dependent case with a planar surface and no horizontal variation
in temperature profile through the fluid, the initial convection
is within the bulk, driven by density forces and does not
produce immediate surface renewal.
From the above observations it would appear that surface-tension-driven convection in the absence of density driving forces could only be observed when there is some form of initiator which will produce surface tension variation along the fluid-fluid surface. These might include meniscus action, surface waves, surface temperature or composition variations, and non-planar fluid-fluid boundaries. The absence of these initiators might explain why Zeren and Reynolds (1972) did not observe surface-tension-driven convection from an adverse-surface-tension linear profile in the water-benzene system when the Marangoni number was almost an order of magnitude above its calculated critical value.

CONCLUSIONS

1. Experiments have been carried out to ascertain the point of convection initiation during transient heating or cooling of deep pools of liquid, with a one-dimensional thermal gradient normal to the surface. Experiments covered fixed and free surfaces, and varied the Prandtl number, the fluid depth, the fluid width, the external vibration level and the shape of the surface temperature history. The results were used to test various predictions of the QSA and Amplification theories.

2. The effects of different surface-temperature histories and different fluid depths are satisfactorily correlated by a unique value of $Ra_{\xi}$, a Rayleigh group based upon the effective thermal boundary layer depth.

3. External vibration promotes instability, in directional accord with the predictions of Amplification theory and the finite-amplitude version of QSA theory.
4. For a low-vibration situation, decreasing the Prandtl number of the fluid increases the Rayleigh number for instability. The asymptotic Ra-Pr functionalities are in agreement with Amplification theory, but correspond to very different Amplification factors required for visible convection. Thus, Amplification theory cannot predict the observed Pr-functionality quantitatively. QSA theory predicts independence of Pr.

5. In the absence of surface waves and external vibration the conditions for instability in deep pools are the same for fixed and free surfaces. This result is in conflict with both QSA and Amplification theories and strongly suggests that the point of incipient convection is at the bulk-fluid edge of the thermal boundary layer.

6. In the absence of surface waves, meniscuses and gradients in surface temperature or composition, there is no surface-tension influence upon free-surface convection initiation in deep and shallow pools, even for Marangoni numbers more than three orders of magnitude above those predicted for instability by the QSA analysis. This relates to the absence of any effect of surface rigidity, and again indicates that the point of instability does not initially interact with the surface. Previous reported instances of surface-tension-driven instability in deep-pool situations may be attributed to surface waves and/or meniscuses.

7. Conclusions 5 and 6 differ from the reported observations of time-independent convection studies (i.e., Berg and Palmer, 1971) where destabilizing surface tension forces and free surface shear conditions have affected the conditions for density-driven convection initiation. The explanation for this should lie in the manner by which the destabilizing effect interacts with the unstable field in the fluid. Further identification of the destabilizing effects most commonly met in convection initiation phenomena is needed for a deeper understanding to explain the difference between time-dependent and time-independent cases.
ACKNOWLEDGMENT

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NOMENCLATURE

\( C_p \) heat capacity at constant pressure (cal/g°C)

\( D \) mass diffusivity (cm²/sec)

\( D \) fluid width (cm)

\( g \) gravity constant (cm/sec²)

\( H \) fluid depth (cm)

\( k \) thermal conductivity (cal/cm sec °C)

\( \ell \) density layer thickness using the segment approximation

\[
2 \int_{0}^{\infty} \left( \frac{T-T_b}{T_s-T_b} \right) \, dz \text{ (cm)}
\]

\( M_H \) Marangoni number based on fluid depth = \( \frac{\Delta \sigma H}{\mu a} \)

\( M_t \) Marangoni number based on conduction time = \( \frac{\Delta \sigma t^{1/2}}{\mu a^{1/2}} \)

\( Pr \) Prandtl number = \( \frac{C_p \mu}{k} \)

\( Ra_H \) Rayleigh number based on fluid depth \( H = \frac{\Delta \rho g H^3}{\mu a} \)

\( Ra_\ell \) Rayleigh number based on effective density layer thickness = \( \frac{\Delta \rho g \ell^3}{\mu a} \)

\( Ra_t \) Rayleigh number based on conduction time = \( \frac{\Delta \rho g a^{1/2} t^{3/2}}{\mu} \)

\( S \) surface temperature shape factor

\( Sc \) Schmidt number = \( \frac{\mu}{\rho D} \)
t  conduction time prior to convection onset (sec)
T  temperature (°C)
\( \Delta T \) temperature difference between bulk and surface conditions
v  velocity perturbation term
w  amplification factor
z  vertical coordinate (cm)

Greek Letters

\( \alpha \)  thermal diffusivity (cm\(^2\)/sec)
\( \mu \)  viscosity (poise)
\( \Delta \sigma \) difference in surface tension between surface liquid and bulk liquid conditions (dynes/cm)
\( \rho \)  density (g/cc)
\( \Delta \rho \) difference in density between surface liquid and bulk liquid (g/cc)
\( \theta \) temperature perturbation term

Mathematical Operators

\[ \nabla = \frac{\partial}{\partial x} + \frac{\partial}{\partial y} + \frac{\partial}{\partial z} \]
\[ \nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \]

Subscripts

b  bulk
c  condition at onset of convection
s  surface
REFERENCES

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