Int. J. Heat Mass Transfer. Vol. 17, pp. 77–83. Pergamon Press 1974. Printed in Great Britain

NATURAL CONVECTION FROM TIME-DEPENDENT PROFILES AT A GAS–LIQUID SURFACE

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(*Received* 17 *December* 1972 *and in revised form as two separate papers* 25 *May* 1973)

Abstract—This experimental study is concerned with the initiation of natural convection at gas–liquid interfaces. Fluids covering a wide range of Prandtl number were cooled from above in a vertical one-dimensional temperature field. The results showed that the conditions at the onset of observable convection at a gas–liquid interface and at a solid–liquid interface were the same, if surface waves, meniscus and surface temperature gradient effects were eliminated from the experiment. Under these special conditions, there was an absence of surface tension destabilization, even though the Marangoni number was three orders of magnitude above its predicted critical value.

NOMENCLATURE

g,	gravity constant [cm/s^2];
H,	fluid depth [cm];
l,	penetration depth $= \int_0^H \left(\frac{T - T_s}{\Delta T}\right) dz$ [cm];
M_H,	Marangoni number based on fluid depth = $\Delta\sigma H/\mu\alpha$;
M_t,	Marangoni number based on conduction time $= \frac{\Delta\sigma t^{\frac{1}{2}}}{\mu\alpha^{\frac{1}{2}}}$;
Pr,	Prandtl number = $\mu/\rho\alpha$;
Ra_l,	Rayleigh number based on penetration depth $l = \frac{\Delta\rho g l^3}{\mu\alpha}$;
Ra_t,	Rayleigh number based on conduction time $= \frac{\Delta\rho g\alpha^{\frac{1}{2}}t^{\frac{3}{2}}}{\mu} = \frac{\rho g\beta\Delta T\alpha^{\frac{1}{2}}t^{\frac{3}{2}}}{\mu}$;
S,	surface temperature shape factor;
t_c,	conduction time prior to convection onset [s];
T,	temperature in fluid [°C];
T_s,	surface temperature [°C];
ΔT,	temperature difference between bulk and surface conditions [°C].

Greek letters

α,	thermal diffusivity [cm^2/s];
β,	coefficient of thermal expansion [°C^{-1}];
μ,	viscosity [P];
$\Delta\sigma$,	difference in surface tension between surface liquid and bulk liquid conditions [dynes/cm];
ρ,	density [g/cm^3];
$\Delta\rho$,	difference in density between surface liquid and bulk liquid [g/cm^3].

Subscripts

c,	condition at onset of convection.

INTRODUCTION

CONVECTION at a gas–liquid interface can be driven by both surface tension and density forces [1–3]. Although linear stability theory can predict the conditions at the onset of convection for a linear temperature gradient through the fluid, and for low values of the ratio of the liquid phase resistance to gas phase resistance [2], instability conditions for the more commonly met case of the non-linear, time-dependent profile are less understood. The experimentally observed conditions at convection onset are normally orders of magnitude above those predicted by linear stability theory.

The discrepancy between linear stability theory and observation for density-driven convection initiation from a time-dependent profile at a liquid–solid surface has been interpreted in terms of both the growth rates of the incipient convection and the magnitude of convection required to affect a heat or mass transfer experiment [4]. Application of this growth rate interpretation to convection at gas–liquid interfaces can potentially provide a description of the density-driven part of the process, but some modifications may be necessary to account for the surface tension influences. The purpose of this work is to provide experimental results and an analysis comparing and contrasting convection initiation results at a gas–liquid surface with results for a solid–liquid interface.

THEORY

The conditions at the onset of density-driven convection can be correlated through the time-dependent Rayleigh number Ra_t where

$$Ra_t = \frac{\rho g \beta \Delta T \alpha^{\frac{1}{2}} t^{\frac{3}{2}}}{\mu}. \tag{1}$$

Ra_t has been observed to be dependent upon the thermal boundary conditions and the depth of the fluid; however a modified Rayleigh number

$$Ra_l = \frac{\rho g \beta \Delta T l^3}{\mu \alpha} \tag{1a}$$

successfully correlates the effects of changes in these conditions [4]. A parameter (S), used to reflect the thermal boundary condition for semi-infinite fluids, describes the surface temperature (T_s) vs time relationship as

$$S = \frac{\int_0^t T_s(\theta)\,\mathrm{d}\theta}{T_s t}. \tag{2}$$

The conditions at the onset of surface tension-driven convection can be interpreted [5] through the time-dependent Marangoni number

$$M_t = \frac{\Delta \sigma t^{\frac{1}{2}}}{\mu \alpha^{\frac{1}{2}}}. \tag{3}$$

EXPERIMENTATION

Deep-pool convection initiation

Previous experiments with free surfaces have used evaporation to provide cooling at the liquid surface, with or without air blown over the surface. The disadvantages of that approach include the development of 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 surface 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 through the gas phase 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 used in this work was chosen to have this one-dimensional character, and is shown in Fig. 1. A cooler with an isothermal cold plate (dia. 50 mm) rested on polystyrene foam supports approximately 1·5 mm thick, which in turn was atop 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 uneven heat transfer across an air gap away from the liquid surface or preferential conduction along the vessel walls. 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 for one-dimensional heat transfer. A thermocouple (1 mil, copper–Constantan), supported from below the liquid by glass leads (1·6 mm dia), 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 temperature response of this thermocouple showed a sharp transition at the onset of convection.

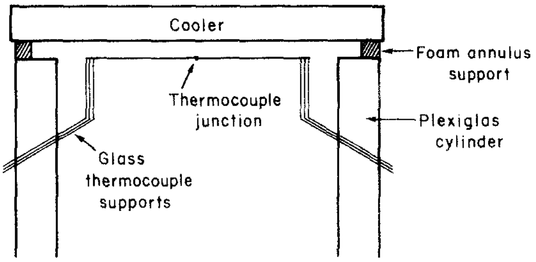

FIG. 1. Schematic diagram of apparatus used to determine convection conditions at a gas–liquid surface.

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 [4]. 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 was essentially vibration-free since it has no moving parts, and it was used to represent the quietest possible conditions for free-surface convection.

The liquids were selected with the constraint that they be non-corrosive and non-volatile and have similar thermal properties to the plexiglas. n-Decane ($Pr = 13{\cdot}3$), n-butanol ($Pr = 43$), n-octanol ($Pr = 108$), and 100 cs silicone oil ($Pr = 900$) were tested. The results for n-decane with the thermoelectric cooler, plotted as ΔT_c vs t_c at the onset of convection, are shown in Fig. 2 and compared with the convection initiation data at a solid–liquid surface obtained in the previous study [4]. Results for the other liquids were similar [6] except under certain conditions where the free-surface results were slightly lower than the fixed-surface results. These conditions were for high ΔT driving force, where 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 onset times for 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/or heat losses through the edge of the apparatus became significant enough to produce two-dimensional character in the surface temperature. Preliminary investigation showed that two-dimensional gradients in surface temperature could lower the onset time considerably.

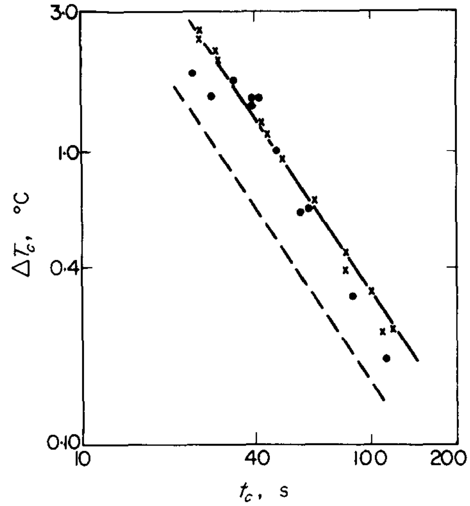

FIG. 2. Experimental conditions for onset of convection in n-decane (Pr = 13) for a linear surface-temperature decay in a deep pool using the thermoelectric cooler × Fixed-surface data [4], ● Free-surface data, -- Free-surface predictions of [7], assuming same amplification factor as for fixed-surface results.

The results in Fig. 2 and for the other liquids show that the convection is density-driven, since the slope is $-\frac{3}{2}$, as opposed to the slope of $-\frac{1}{2}$ which is predicted for surface-tension-driven convection [3]. Furthermore, these results show that convection initiation is independent of the shear conditions at the heating or cooling boundary (i.e. fixed vs free surface), in a vertical, one-dimensional, deep-pool density field. This is in contradistinction to Amplification Theory [7] and Linear Stability Theory [6], which predict that the free-surface Rayleigh number should lie below that for a fixed surface by a factor of 2–3·5. Even when the free-surface data fell below the fixed-surface data in the present work, they were always well above the free-surface predictions from either of these theories (see Fig. 2).

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 [6] were similar to the fixed-surface results reported in the previous paper [4]. Ra_t varied approximately linearly with the shape factor S, and was a factor of three higher for the linear

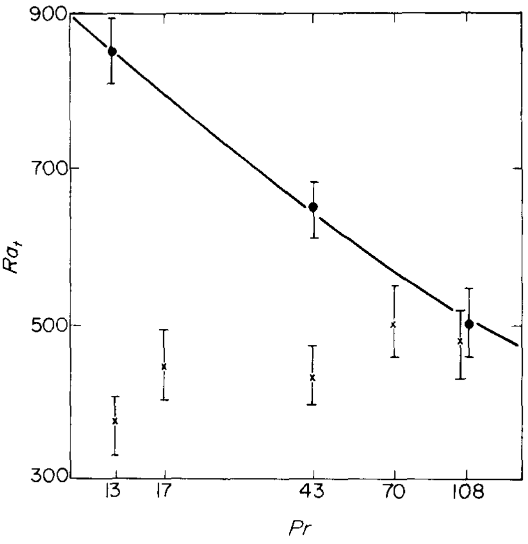

FIG. 3. Comparison of values of Ra_t at free surface determined with the spray cooler (×) and the thermoelectric cooler (●), for a linear surface-temperature decay (S = 0·5).

change in surface temperature than for the step change.

The influence of the surface waves from the spray cooler on the onset of convection can be seen in Fig. 3, 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, deep-pool results

Foster [8] identified his experiments where water was evaporated from stagnant pools as a case of linear decay of surface temperature with time ($S = 0{\cdot}5$). Vidal and Acrivos [9] 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 $S = 0{\cdot}67$, which agrees closely with Foster's reported record of T_s vs t. Foster's results of $Ra_t = 225$ for evaporation of water ($Pr = 6{\cdot}6$) with $S = 0{\cdot}67$ agree well with the present results for n-decane ($Pr = 13{\cdot}3$) using the spray cooler. 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 [5] published results for evaporation of carbon disulfide from tridecane solvent. They observed possible density-driven convection at high CS_2 mole fraction, corresponding to $Ra_t = 285$ and $S = 0{\cdot}62$, 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 [10] identified density-driven convection for the absorption of carbon dioxide into a laminar flowing aqueous stream. The Rayleigh number, Ra_c, based upon the composition gradient was reported as 785 with $Sc = 300$, but the Rayleigh number based upon the temperature gradient 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 (Fig. 3) where no stabilizing influence was present.

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, shallow-pool results reported elsewhere [4].

Both the spray cooler and the thermoelectric cooler were used to determine the onset conditions in shallow pools. Either the surface temperature 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 are given in Fig. 4. The spray-cooler data were similar [6]. The fixed-surface data from the previous study have been plotted with the gas-liquid data for comparison. The results show that there is no significant difference between the fixed- and the free-surface data in the range of $\alpha t/H^2$ used in these experiments. 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, rather than surface-tension-driven. This is true even for thin liquid films (2 mm thick) and high resultant values of Marangoni number, M_t up to 10500 and M_H up to 5200.

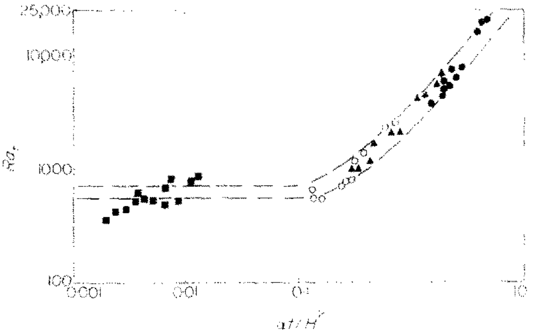

FIG. 4. Experimental values of Ra_t at a free surface as a function of depth factor ($\alpha t/H^2$) for n-butanol. The linear decay of surface temperature with time was produced with a thermoelectric cooler.

Fluid depth: ■ 36·5 mm ▲ 3·3 mm
○ 5·0 mm ● 2·16 mm
---- Limits of fixed surface results from [4].

DISCUSSION

The result that the observed conditions for convection onset at a gas–liquid interface and at a solid–liquid surface are the same is unexpected when the shear boundary conditions in the two cases are so different. With systems that are not dependent upon the convection–amplification process, i.e. for linear gradients in time-independent systems, the effect of the difference in shear boundary conditions has been observed experimentally [2, 11]. However, it appears that in time-dependent processes the location of the growth of the incipient convection is far enough removed from the interface so that the observable convection is dependent upon only the density driving force, and not the rigidity of the surface. This explanation does not preclude the importance of surface rigidity for well-developed convective flow, however. In the Schlieren studies of Onat and Grigull [12], the first convective motion can be observed at the fluid-side edge of the density boundary layer. This can be rationalized on the grounds that the density gradient at this site is weak enough to be rearranged by the disturbance motion and yet strong enough to sustain convection growth.

Surface waves in low-viscosity fluids, where present, were observed to have a significant effect on the time of observable convection onset. However, the lower values of Ra_t found if waves were present were still within the range where the growth rates of the incipient convection are beginning to increase rapidly [4, 8]. Whether the decrease in stability was due to increased motion from the waves or to surface tension effects could not be ascertained from the present data. However, Foster [8] showed that for convection on a water surface, which apparently was affected by waves, that Ra_t for convection onset was approximately constant, while M_t for onset varied by a factor of 2. This would suggest that surface waves are destabilizing, but does not rule out the possibility of surface-tension influences upon convection initiation with clean surfaces.

Wave action may be the explanation of why Ra_t for some gas absorption systems was found to increase as the surface contamination and thus rigidity increased. Blair and Quinn [3] and Mahler and Schechter [13] observed that Ra_t was higher by about a factor of two for an insoluble surfactant layer on an aqueous surface, than for a soluble surfactant. In light of the results shown in Fig. 2, surface waves were probably dampened by the more rigid insoluble surfactant to a greater extent than with the soluble surfactant.

Both the shallow- and deep-pool free-surface convection-initiation results (Fig. 4) showed no influence of surface-tension gradients upon convection initiation, even up to high values of the Marangoni number ($M_t = 10500$, $M_H = 5200$). This was in the absence of meniscus effects, gradients of temperature along the surface, and surface waves. This result is in contradistinction to the predictions of linear stability theory developed by Vidal and Acrivos [9], 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 [14] should not change their deep-pool solution significantly for these experimental conditions.

This discrepancy between experiment and existing theory may well be connected with the observation of the equivalence of convection initiation at free and fixed surfaces in deep pools. If the incipient convection is confined to the bulk-fluid edge of the density boundary layer, the rigidity of the surface is not felt and thus no apparent shear variation along the surface is produced during the growth period. Provided that there are no surface temperature gradients along the surface, no local surface disturbance and no meniscus, there should 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 results and previously published work on convection initiation in thin films of evaporating, pure organic liquids. Berg *et al.* [1] and Vidal and Acrivos [9] found convection initiation at Ra_H much below 1700 for evaporating films and so ascribed the surface-tension gradient 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-temperature gradient and meniscus were eliminated, the results were similar to the fixed-surface results. Mitchell and Quinn [15] 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 [5], Brown and King [16], Blair and Quinn [3], Brian *et al.* [17], surface waves have been present and from the above discussion appear to be the important disturbance.

Ellis and Biddulph [18] 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 interface 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 reverse direction of flow. The momentum associated with the spreading and reversal of the low-surface-tension fluid resulted in a surface ripple.

The stabilization observed in a benzene–water layer [19] heated from above and predicted to be unstable because of the surface tension gradient could have been due to an absence of surface disturbances along the water benzene interface. In this case, the density gradient was stable and could suppress bulk and surface wave motion. However, when the system was heated from below so that both the density and surface tension gradients were stable, the experimentally observed destabilization agreed with theoretical predictions. Surface waves could be present in the unstable density field.

CONCLUSIONS

The observable onset of convection at a gas–liquid surface appears experimentally to occur under the same conditions as at a liquid–solid surface, provided there are no meniscus, surface wave or surface temperature gradient effects present. This observation indicates that the site of growth of the incipient convection between first instability and observable size is sufficiently deep within the fluid so that there is negligible influence of the surface rigidity upon it.

Surface waves were found to destabilize the system by either promoting surface tension motion on clean surfaces or providing more energetic disturbances on both clean and contaminated surfaces. Meniscus action and surface temperature variations on clean surfaces were also observed to destabilize.

Acknowledgement This work was performed in the Lawrence Berkeley Laboratory under the auspices of the U.S. Atomic Energy Commission.

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CONVECTION NATURELLE Á UNE SURFACE GAZ-LIQUIDE Á PARTIR DE PROFILS DÉPENDANT DU TEMPS

Résumé— Cette étude expérimentale concerne l'initiation de la convection naturelle aux interfaces gaz–liquide. On refroidit par la base des fluides couvrant un large domaine de nombre de Prandtl et placés dans un champ de température monodimensionnel et vertical. Les résultats montrent que les conditions d'apparition de la convection à l'interface gaz–liquide et à l'interface solide–liquide sont les mêmes si l'on élimine les effets d'ondes de surface, de ménisque et de gradient de température à la surface. Dans ces conditions spéciales la déstabilisation par tension interfaciale est absente, même si le nombre de Marangoni est trois fois plus grand que la valeur critique estimée.

FREIE KONVEKTION VON EINER GAS—FLÜSSIGKEITS—GRENZFLÄCHE. BEI ZEITABHÄNGIGEN PROFILEN

Zusammenfassung—Diese experimentelle Arbeit befasst sich mit dem Einsetzen von freier Konvektion an Gas—Flüssigkeitsgrenzflächen. Fluide in einem weiten Prandtl-Zahlbereich wurden von oben in einem senkrechten eindimensionalen Temperaturfeld gekühlt. Die Ergebnisse zeigen, dass die Bedingung beim Einsetzen von beobachtbarer Konvektion an einer Gas—Flüssigkeitsoberfläche dieselben sind wie an einer Fest—Flüssigkeitsgrenzfläche, wenn Effekte durch Oberflächenwellen, Miniskus und Oberflächentemperaturgradienten ausgeschaltet werden. Unter diesen Bedingungen gibt es keine Destabilisierung der Oberflächenspannung auch wenn die Marangoni—Zahl um drei Grössenordnungen über ihrem vorausgesagten kritischen Wert liegt.

ЕСТЕСТВЕННАЯ КОНВЕКЦИЯ ПРИ ЗАВИСИМЫХ ОТ ВРЕМЕНИ ПРОФИЛЯХ НА ПОВЕРХНОСТИ РАЗДЕЛА ГАЗ-ЖИДКОСТЬ

Аннотация—Проведено экспериментальное исследование возникновения естественной конвекции на поверхностях раздела газ-жидкость. Охлаждение жидкостей в большом диапазоне чисел Прандтля осуществлялись сверху в вертикальном одномерном температурном поле. Результаты исследования показали, что конвекция на поверхности раздела газ-жидкость и твердое тело-жидкость возникает при одинаковых условиях, если исключить из эксперимента поверхностные волны, мениск и температурный градиент на поверхности. При таких условиях наблюдалось отсутствие нарушения устойчивого поверхностного натяжения, не смотря на то, что число Марангони было на три порядка выше расчетного критического значения.