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MARANGONI-BÉNARD CONVECTION IN AN EVAPORATING LIQUID THIN LAYER

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ABSTRACT

Marangoni-Bénard convection in evaporating liquid thin layers has been investigated through flow visualization and temperature profile measurement. Twelve liquids, namely ethyl alcohol, methanol, chloroform, acetone, cyclohexane, benzene, methylene chloride, carbon tetrachloride, ethyl acetate, n-pentane, silicone oil (0.65 cSt.), and freon-113, were tested and convection patterns in thin layers of these samples were observed. Comparison among these tested samples shows that some liquids are sensitive to surface contamination from aluminum powder but some are not. The latter is excellent to be used for the investigation of surface-tension driven convection through visualization using the tracer. Two sample liquids, alcohol and freon-113 were particularly selected for systematic study. It was found that the wavelength of Bénard cells would not change with thickness of the layer when it evaporates at room temperature. Special attention was focused on cases in which a liquid layer was cooled from below, and some interesting results were obtained. Convection patterns were recorded during the evaporation process and the patterns at certain time frame were compared. Bénard cells were observed in thin layers with a nonlinear temperature profile and even with a zero or positive temperature gradient. Wavelength of the cells was found to increase as the evaporation progressed.

INTRODUCTION

The first systematic experimental study of the convection in a horizontal liquid thin layer was made by Henri Bénard [1,2]. Although the main portion of his report concerned spermaceti, Bénard also experimented with several different liquids and found that volatile liquids produced permanently unsteady cellular convection in shallow pools. In other words, Bénard actually experimented also with evaporating liquid layers. However, the convection observed in Bénard's experiments was believed to be driven by buoyancy and theoretically explained by Lord Rayleigh [3]. Nowadays it is referred to as Rayleigh-Bénard convection. Block [4] demonstrated the convection cells in a layer thinner than 1 mm with a free surface were produced by variations in surface tension instead of buoyancy. Under motivation of Block's work and others, J. R. A. Pearson [5] proposed a radically different theoretical model from Rayleigh's. In Pearson's model the fluid is assumed to be supporting an adverse linear temperature gradient and all the fluid properties except the surface tension are taken to be independent of temperature. Pearson analytically showed that the Bénard convection can be produced by the variation of surface tension with temperature. This surface-tension-driven convection is now referred to as Marangoni-Bénard convection. D. A. Nield [6] combined these two theories and found that the two agencies causing the Bénard convection (i.e. buoyancy and surface tension forces) reinforce one another and are tightly coupled. Pearson's and Nield's theories predict a critical thickness below which there is stability relative to convection induced by surface tension although Block reported there was no indication that a critical thickness had been reached at least until 50 microns. An open question is: whether or not actually there is a critical thickness? If a critical thickness exists as the theories predicted what is the limiting value and why? Analyses now available can not explain Block's experimental

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results because no one really considered the effects of evaporation on the convection which may be the key point in the experiments conducted by Block. Pearson claimed: " The allied problem of a liquid cooling by evaporation may be treated in a similar fashion.....owing to the loss of fluid from the surface, the effect of evaporation may be reasonably well represented by a given heat loss from the surface,....." Actually, this masked the essential mechanism of the convection fueled by the evaporation. Another question dodged by most investigators is: what would happen when an evaporating liquid layer was cooled from below? Almost all relevant analyses presented until now based on the assumption that the liquid layer is heated from below except Castillo and Velarde [7]. Only a few experiments dealt with Bénard convection in an evaporating liquid layer and Block may have been the only one who conducted an experiment on an evaporating liquid layer cooled from below. Berg [8] described four principal patterns of evaporative convection and pointed out that cells appeared to be the dominant patterns in all liquids for depth of 2 mm or less. He reviewed the Bénard convection dealing with evaporation in detail and analyzed the mechanisms of Bénard convection but never touched upon the mechanism problem of the convection in a liquid layer cooled from below. Zhang and Chai [9] initiated to investigate the evaporative convection in a liquid thin layer cooled from below; some effects of evaporation on the convection were discussed. In the present work, more liquid samples were tested as liquid thin layers (≤ 1 mm). More detailed experimental data for alcohol and Freon-113 were graphed and discussed. The effects of evaporation on the convection, including the structure of patterns and the size (wavelength) change of the convection cells, are experimentally studied.

EXPERIMENTAL APPARATUS AND PROCEDURE

The apparatus used in the present study is the same as in reference [9]. It consisted of a test section with a cooling base, a refrigerated circulator, a thermocouple set, a computer data acquisition system, a CCD camera mounted on a microscope with zoom lens and connected to a video recorder, a color video printer and a color monitor. The test section is made by a flat polished copper base plate and four glass side plates. The four glass plates of 2 mm thick and 6 mm wide are glued on the copper plate of 1.5 mm thick to form a square trough of 50.8 x 50.8 mm. The side walls of 6 mm high are surrounded with expanded polystyrene of 5 mm thick for thermal insulation. The test section is sat on the aluminum top plate of the cooling base which is cooled through a refrigerated circulator. The circulator has a working temperature range of -23 to 150 °C, regulated by PID controller, with an accuracy of ± 0.02 °C. The convection patterns in the test layer can be recorded with the video recorder through the CCD camera mounted on the zoom microscope and monitored with the color monitor in the meantime. Any interesting picture shown on the monitor, either in real time or when playing back a recorded tape of an experiment, can be caught by the video printer and printed out within 30 sec. Eight thermocouples are connected to the computer data acquisition system to record the temperature profiles of the test liquid layer and of the air close to the liquid surface. The thermocouples were divided into two groups, four in each. Each group was mounted on a 3-Axis stage with differential micrometers and so can be moved in 3-D with an accuracy of 0.0005 mm. The thermocouples are made with fine CHROMEGA and ALOMEGA wires 0.0005 in.(0.0127 mm) in diameter and deployed one over another. The distances between the thermocouple tips can be accurately measured through the microscope. The lowest thermocouple of Group I (numbered from lowest one as #1, #2, #3, and #4) can be adjusted to touch to the bottom of the test section through the 3-Axis stage and can therefore measure the layer temperatures at the bottom, and at the positions of 0.278 mm, 0.496 mm, and 0.754 mm from the bottom. The lowest one in Group II (numbered from the lowest one as #5, #6, #7, and #8) was placed at 1.000 mm from the bottom and so can measure the temperature at the surface of the test liquid layer when the initial thickness of the layer is set 1.000 mm and the temperatures of the air at the positions of 0.258 mm, 0.555 mm, and 0.892 mm from the liquid surface can be measured by #6 to #8. All of the thermocouple tips were adjusted in a perfect alignment perpendicularly through the micrometers on the stages.

Although the thermocouple wires were already calibrated by the manufacturer, the divergence of the temperature measurements between the thermocouples was still more than $0.5\text{ }^{\circ}\text{C}$ due to the material inhomogeneities. Therefore a rigorous sifting was necessary. In order to obtain meaningful temperature profiles within very tiny distances, a measurement divergence among the eight thermocouples should not exceed $0.05\text{ }^{\circ}\text{C}$ in the temperature range of 0 to $30\text{ }^{\circ}\text{C}$. Only one out of approximately 20 thermocouples was found to be qualified for use in the thermocouple set. The thermocouples in both Group I and II were also specially calibrated before and after every test was made to ensure the data obtained to be reliable. Test liquids mixed with aluminum powder of $1 - 3\text{ }\mu$ diameter are injected into the test section speedily and smoothly through a syringe and the convection patterns and the temperature profiles in the test liquid layer and in its vicinal air were recorded simultaneously.

RESULTS AND DISCUSSION

Twelve liquids, namely ethyl alcohol, methanol, chloroform, acetone, cyclohexane, benzene, methylene chloride, carbon tetrachloride, ethyl acetate, n-pentane, silicone oil (0.65 cSt.), and Freon-113, were tested and convection patterns in the thin layers were observed. All of the samples were first tested at a thickness of 1 mm or less to observe surface-tension driven convection and then at thicknesses of 1.5 mm and 2 mm , respectively, to observe the buoyancy effects on the convection. Quite different convection patterns were found in the sample layers with a same thickness, for example 1 mm . Figure 1 shows the typical patterns in an 1 mm layer of silicone oil (0.65 cSt.), alcohol, freon-113, and n-pentane, respectively, which evaporate at room temperature ($24\text{ }^{\circ}\text{C}$). The wavelength λ , which equals to H/δ , can be determined from the pictures shown in Fig.1, where H is the mean value of the distance between the centers of two neighboring cells, and d , the thickness of the layer. It is obvious that silicone oil and alcohol have more regular Bénard cells and the wavelengths are 2 and 3, respectively. Freon-113 has a kind of combination of cells and rolls, while more violent and complex motion appears in the n-pentane layer in which small cells are covered by large cells or rolls. It can be seen that cells in the freon-113 layer have a wavelength of about 6. Methanol layer has the same convection pattern as freon-113 has (not shown). However, some samples, such as acetone, benzene, and cyclohexane exhibited convection cells in an 1 mm layer only for very short period of time just after the liquid is deployed and then quieted down, while others, like carbon tetrachloride, ethyl acetate, chloroform, and methylene chloride never exhibited convection at this thickness. The reason may be that the surface of these liquids is very sensitive to the "contamination" caused by the aluminum powder. The liquids which are not sensitive to surface contamination from aluminum powder, such as alcohol, freon-113, n-pentane, silicone oil, and methanol, are excellent for the investigation of surface-tension driven convection in an evaporating thin layer through flow visualization using aluminum powder as the tracer. The wavelength of the cells depends on the liquid properties and the evaporation conditions but not the thickness of the layer. For example, alcohol layer evaporating at room temperature has the wavelength of about 3 for different layer thickness, while for freon-113 the value of λ is about 6. It is obvious that there is no critical thickness be reached until 0.1 mm . However, two evidences demonstrate the existence of a critical thickness: first, in the last stage of evaporation, no cells appeared in the rim of the layer which is drying out; second, the cells disappeared at about 0.1 mm thick when a thicker freon-113 layer evaporated and the thickness decreased with the evaporation.

More interesting results were obtained when an evaporating liquid thin layer was cooled from below. As described in reference [9], Bénard cells can be observed clearly in a liquid thin layer with a nonlinear temperature profile and even with zero or positive temperature gradient. More detail observation has found that the wavelength of the cells in the evaporating layers cooled from below changed with time during the cooling process in which the temperature profile varied. Generally, the wavelength would become larger and larger as the evaporation progresses. Figure 2 shows the temperature histories and convection patterns in an

evaporating freon-113 layer at different moments when its bottom is cooled to 11.14 °C. Moments A, B, and C correspond to 5, 15, and 45 seconds after the liquid deployment is finished. It can be seen that moment A is a converse point of the temperature profile in the layer from a negative to a positive temperature gradient and the temperature gradient at this moment is almost zero. However, active Bénard cells with wavelength $\lambda = 1$ (the layer thickness $\delta = 0.994$ mm) can be observed clearly, as shown in Fig. 2 (b). Even more brisk Bénard cells can be observed at moments B and C with a wavelength of 3.5 ($\delta = 0.983$ mm) and 10 ($\delta = 0.950$ mm), respectively, as shown in Fig. 2 (c) and (d). It can be seen that the cell construction tends to become more complex as the evaporation progresses. Apparently, the strong nonlinear profile of temperature in the layer could play a key role in these phenomena. An increase of the wavelength with evaporation was also found in a thin layer of alcohol even with a zero temperature gradient (when the layer bottom was cooled to 16.6 °C) and with a positive temperature gradient (when the layer bottom was cooled to 12.3 °C).

In reference [9], a modified form of Marangoni number, $Ma = -\sigma m_h \delta^2 / k \mu \alpha$ was introduced to take into consideration of the evaporation effects, where σ is the surface tension gradient with respect to temperature, m , evaporation rate, h , latent heat of the liquid, d , the layer depth; k , μ , α are thermal conductivity, viscosity, and thermal diffusivity, respectively. However, in view of the above discussed experimental results, δ , the layer depth, may need to be modified to account for the effect of the nonlinear temperature gradient in the layer. It is conceivable that the depth of the temperature gradient conversion point in the layer could be used in place of δ in defining a more meaningful Marangoni number.

CONCLUSIONS

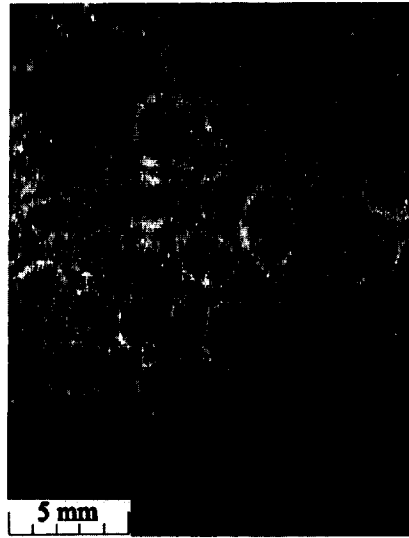
Studies of evaporating thin liquid layers clearly demonstrated that Bénard cells and similar thermocapillary flows can be triggered and sustained even if the layers were cooled from below. It is found that the wavelength of the cells depends on the liquid properties and the evaporation conditions but not the thickness of the layer when it evaporates at room temperature, while the wavelength would become larger and larger with the evaporation when the layer is cooled from below. Evaporation and nonlinear temperature profile may play an important role in the capillary convection. The traditional static instability theory and modeling of Marangoni-Bénard convection can no longer describe the complex phenomena involving a dynamic, evaporating free surface. Further study and more experimental data are needed to adequately account for both evaporation and temperature gradient conversion in an evaporating liquid layer.

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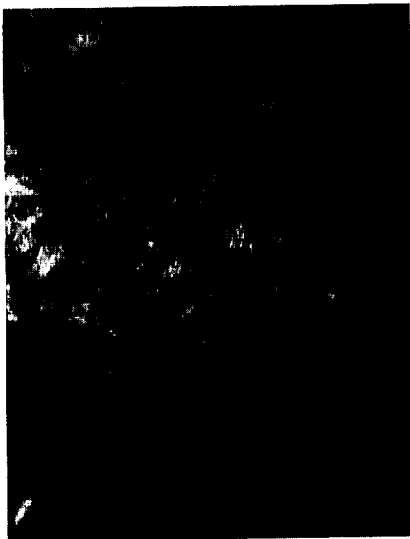
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(a) Silicone oil (0.65 cSt.)



(b) Alcohol

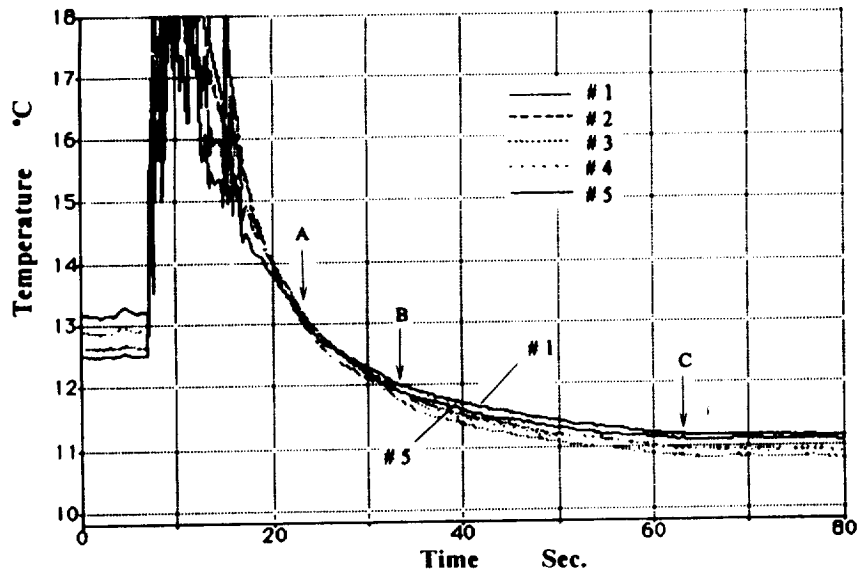


(c) Freon-113

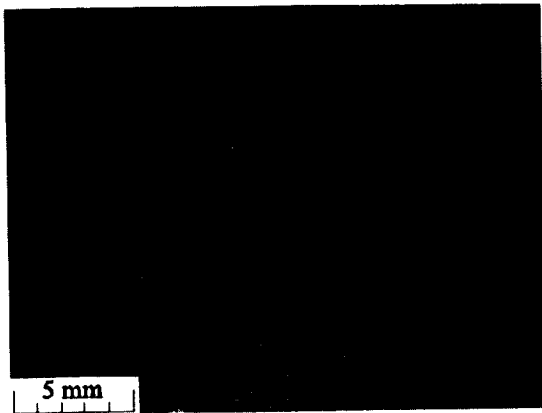


(d) n-pentane

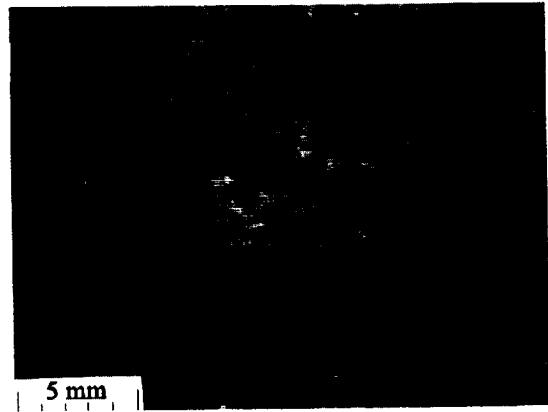
Fig. 1 Convection patterns in an 1 mm layer evaporating at room temperature



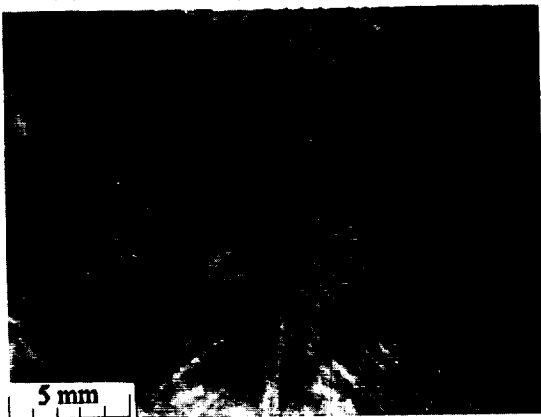
(a) Temperature histories



(b) Convection pattern at moment A



(c) Convection pattern at moment B



(d) Convection pattern at moment C

Fig. 2 Temperature histories and convection patterns in a thin layer of freon-113 cooled from below