An attempt at enhancing direct contact evaporation utilizing the surface dilational viscoelasticity

YASUHIKO H. MORI and SHIGEKI END0

Department of Mechanical Engineering, Keio University, 3-14-1 Hiyoshi. Kohoku-ku. Yokohama 223, Japan

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Abstract-This work aims to show a potential utility of the 'surface film' in enhancing 'interfacial heat transfer'. The particular problem considered is to enhance the heat transfer to-and the evaporation ofsingle lenses of a volatile liquid floating on the surface of a stagnant pool of water. By causing, with the aid of an external force, alternate stretch and compression of a surface film spread on the water surface around each lens. we have succeeded in making the lens undergo periodical expansion/shrinkage, resulting in a significant shortening of the evaporation time of the lens.

1. INTRODUCTION

IT IS GENERALLY believed that a clean surface of a pure Newtonian liquid in contact only with its own vapor and/or an inert gas has no rheological property. That is, the surface is assumed to show no resistance against any externally-forced dilational and/or geometrical change in the very surface. Such a nature of a clean surface is reasonably ascribed to the molecular relaxation process within the surface being sufficiently fast compared to the macroscopic motions forced to the surface under any experimental condition [l]. If the surface is 'contaminated' by adsorbing molecules of another compound, there appear rheological properties in the sense that the surface now shows a viscoelastic response when an external force is applied to change its area and/or shape. Such rheological properties come from the fact that the adsorption/ desorption as well as the diffusion of adsorbate molecules are relatively slow and hence the surface tension is no longer uniform or isotropic at any instant [1].

In various heat and mass transfer operations there exist fluid interfaces (or surfaces) which probably have such rheological properties as mentioned above because of coexistence of two (or more) fluid substances or of surfactant contaminants naturally adsorbed at the interfaces. Usually rheological properties are a negative factor in heat and mass transfer operations, because they readily retard the motion of each fluid interface, resulting in a significant decrease of the rate of heat or mass transfer across the interface. However, the rheological properties may turn out to be a positive factor, when we intend to use an external force to move the interface and thereby enhance heat or mass transfer. The external force can be efficiently transmitted by the interface itself, if it has appreciable rheological properties, giving an agitation concentrated on a region adjacent to the interface. To our

knowledge no attempt has been reported so far on such an active augmentation of heat or mass transfer as the one suggested above.

The work reported in this paper aims to demonstrate the utility of the surface rheology in enhancing heat transfer in a particular system : a single lens of a volatile liquid floating on the surface of a stagnant pool of a less volatile liquid evaporates due to the heat supply from the bulk of the pool. This kind of direct contact evaporation has potential applicabilities to, e.g. water desalination units, geothermal-heat recovery systems and industrial-waste-heat recovery systems, and has been studied by some research groups [2-91. The factor controlling the rate of heat transfer to—and evaporation of—each floating lens of a volatile liquid is a relatively weak convection induced, by a local cooling caused by the evaporation itself, in the hotter substrate liquid [8,9]. In the present work we have tried to force each lens to undergo periodical expansion/shrinkage, by utilizing the surface rheology-particularly the dilational viscoelasticity-of the substrate-liquid surface in contact with the common vapors of the two fluids, which possibly results in an increased rate of heat transfer and evaporation. The details of the expected mechanism of the motion of a lens and the substrateliquid surface are given in Section 2. Later sections are devoted to the description of the experimental work and to the results obtained.

2. PRINCIPLE-SURFACE RHEOLOGY IN FILM/LENS SYSTEM

Suppose a single liquid lens of substance 1 floating in equilibrium on the surface of liquid substrate of substance 2 (Fig. 1). Substance 2 may be water or some aqueous compound, while substance 1 may be a

NOMENCLATURE

volatile, oily compound such as a lower hydrocarbon. The lens and the substrate are both in contact with the mixture of saturated common vapors of the two substances. Note that we generally abbreviate hereafter 'substance 1 in the liquid state' to 'liquid 1' and 'substance 2 in the liquid state' to 'liquid 2'.

If the initial spreading coefficient of liquid 1 on liquid 2 is positive, while the final spreading coefficient is negative, as accepted for many combinations of liquids. a trace amount of substance 1 must be spread out to a thin, probably monomolecular adsorbed film on the surface of liquid 2 around the lens [lo]. Further, it is very likely in any experiment that surfactant contaminants are also adsorbed on the surface, resulting in the formation of a so-called 'mixed film'. unless a special precaution is taken to rigorously purify the system. Figure 1 gives an illustrative schematic of a lens and such a 'surface film' as mentioned above, which are confined to a specified area one end of which is limited by a movable floating barrier. Each balland-stick standing at the substrate-liquid surface represents each adsorbate molecule which may either be a surfactant molecule having both hydrophobic and hydrophilic radicals or a molecule of substance 1 having no hydrophilic radical. Because of the surface film

FIG. 1. **A** lens of liquid 1 floating on the surface of liquid 2 and an adsorbed film around the lens.

consisting of such adsorbate molecules, the surface tension of the substrate liquid σ_2 is lowered from the intrinsic surface tension of pure liquid 2, σ_{20} , to a certain value, σ_{2e} .

Now suppose that we increase the total area A_{total} occupied by the lens and the film by, e.g. pulling the barrier to the left. This results in a decrease in the number density of adsorbate molecules in the surface film and thereby an increase in σ_2 from the former equilibrium value σ_{2e} toward σ_{20} . Thus the lens is pulled outward more strongly and hence expands to occupy a larger area. In this course some molecules of substance 1 are possibly drawn out of the lens to the film. At the same time some in the vapor phase and some dissolved in the substrate liquid may be adsorbed to replenish the surface film. If either or both of these rate processes are present, the increase of σ_2 should depend not only on the extent of a change in A_{total} but also on the rate of the change, dA_{total}/dt . In other words, the surface film should show an apparent viscoelasticity, which is characterized by the surface dilational modulus *E* and the surface dilational viscosity κ defined, respectively, as

$$
E = d\sigma_2/d \ln A_{\text{film}}
$$

$$
\sigma_2 - \sigma_{2e} = \kappa (dA_{\text{film}}/dt)/A_{\text{film}}
$$

where A_{film} is the area of the surface film. The area of the lens, $A_{\text{lens}} \equiv A_{\text{total}} - A_{\text{film}}$, should change depending on the viscoelasticity of the film. In order to facilitate the understanding of the expected dynamic behavior of the film/lens system, we show in Fig. 2 some mechanical models possibly simulating the behavior. Models (a) and (b) correspond to the expansion behavior in the absence and in the presence of the

FIG. 2. Mechanical spring-dashpot models representing the rheological behavior of a surface film/lens system subjected to expansion $((a)$ and $(b))$ or compression $((c)$ and $(d))$: (a) with no adsorption during expansion ; (b) with adsorption during expansion ; (c) with neither desorption nor film collapse during compression; and (d) with desorption and/or film collapse during compression.

adsorption of molecules of substance 1 from either the vapor phase or the substrate liquid phase, respectively.

Next suppose that we contract the total area A_{total} . If neither a desorption of the adsorbate molecules from the film nor a collapse of the film (i.e. monolayerto-bilayer transition **[l** 11) occurs while the film/lens system is compressed, the film may continue to press the lens as a rigid film would do. Model (c) in Fig. 2 represents this case. If a desorption of molecules of substance 1 occurs resulting in an increase in the number density of the residual surfactant molecules in the film, and/or a local collapse of the film occurs, σ_2 will change. Since both the desorption and the film collapse are rate processes, there will appear a viscoelasticity of the film, if both or either of them happen. This situation may be simulated by model (d) in Fig. 2. Both in expansion and in compression, σ_2 will relax to its equilibrium value σ_{2e} [12], if we interrupt the change in A_{total} , resulting in the relaxation of the lens toward its original equilibrium shape.

The above consideration suggests that if we repeatedly alternate an increase with a decrease in the area *A total* covered by a surface film and a lens, the lens will exhibit a periodical expansion/shrinkage responding to the change in A_{total} . This reciprocal motion of the lens and the film will induce a convection in the substrate liquid beneath and around the lens, and, if the system is not isothermal, will enhance the heat transfer between the substrate liquid and the lens.

3. **EXPERIMENTAL**

We began our experimental work by testing some different techniques for expanding and compressing film/lens systems using a hydrocarbon and water as substances 1 and 2, respectively [13]. Various techniques for expanding or compressing the surface film are found in the literature on surface chemistry. Two difficulties were noted in applying these techniques to the present study.

(1) The rates of expansion and compression required to obtain a significant heat transfer enhancement must be much higher than those used by surface chemists in studying the properties of surface films. Any experimental operation giving such high rates of expansion and compression can readily induce undesired disturbances, in both the surface and the bulk of the substrate liquid, that may yield secondary effects on the heat transfer.

(2) The test system should be completely enclosed in a temperature-controlled, deaerated chamber to observe the evaporation of lenses in a well-defined condition [6-91.

Considering these requirements we decided to employ the experimental scheme which is illustrated in Fig. 3 and described below.

In short, the experimental setup constructed was a combination of an apparatus similar to those used in previous lens-evaporation experiments in our laboratory [7-91 and a device which enabled a reciprocal longitudinal motion of a pipe crossing the surface of the substrate liquid which was water. The main portion of the apparatus (shown in Fig. 3(a)) consisted of vertically-oriented, concentric cylindrical columns made of transparent polymethyl methacrylate (PMMA). The inner column served as the test chamber, while the outer column served as a constanttemperature bath. In the test chamber a circular pipe of 190 mm i.d. and 120 mm height was welded to the bottom plate to form a container of water which was to serve as the substrate liquid. The surface of the water pool in the container could readily be renewed if necessary by feeding additional water from an external reservoir into the water pool to flood it. The residual space in the test chamber had been filled with air when n-decane or n-octane was used to form a lens on the water pool surface. When n-pentane was used instead of n-decane or n-octane, the air had been evacuated from the chamber and n-pentane vapor was supplied from an external boiler to fill the space in the chamber. In any case the total pressure in the chamber was nearly equal to atmospheric pressure (101.3 kPa).

The device to move a pipe, which was partially immersed in the water pool, may need some explanation. It was a circular Teflon pipe of 118 mm i.d. or a PMMA pipe of 152 mm i.d. It was rigidly hung at the lower end of a Duracon (polyacetal) rod which was inserted into the test chamber along its central axis from its top. The rod could be moved axially in both directions with the aid of a rack-and-pinion mechanism coupled with a variable-speed motor, while the assembly about the rod prevented any leak of vapor or air from or into the chamber. Further, the rod could be continuously reciprocated with a

FIG. 3. The main portion of the apparatus (a) and the arrangement of optical components for photographic recording (b). The boiler and the condenser illustrated in (a) were used only in the experiments using n-pentane as the lens-forming liquid.

constant stroke by the addition of a special switch assembly to the rack-and-pinion mechanism.

Each experimental run was started by so lowering the Duracon rod that the pipe was immersed in the water pool to an appropriate depth. The liquid to form a lens on the water surface was fed to a Pyrex nozzle submerged in the water pool. A drop released from a nozzle was buoyed up to the water surface, where it turned into a thin biconvex lens. Then the pipe was set in descending or ascending motion with a prescribed speed. If a part of the film that had been on the water surface clung to the pipe wall (just like in the case of Langmuir-Blodgett film formation), the

FIG. 4. Hypothetical illustration of the behavior of a surface film driven by a Teflon or a PMMA pipe in a vertical motion. Concerning the dependency of the behavior on the pipe material (Teflon or PMMA). see Section 4.

film should certainly be expanded pulling the lens radially (see Fig. 4). The film deposited on the pipe wall during the above process may be released again onto the water surface, when reversing the direction of the pipe motion, now resulting in a compression of the film itself and the lens.

It may be necessary to note that the top cover plate of the pipe, which connected the pipe to the Duracon rod, had vent holes so that the vapor or air in the pipe was slightly compressed or expanded while the pipe was moving. This cover plate also had a mirror on its lower face. which enabled us to observe the plan view of a lens on the water surface. The plan view of the lens and a scale indicating the instantaneous elevation of the moving rod were simultaneously recorded by a 35 mm motor-driven camera (Fig.3(b)).

The fluids used to form liquid lenses were n-decane. n-octane and n-pentane, having 99, 91 and 99% wt certified purities, respectively, which were distilled once in an all-glass distillation apparatus in our laboratory. The water was taken from a commercial apparatus which comprised a reverse-osmosis device and an ion-exchange device as well as an all-glass distillation unit. The procedure of the cleaning of the test chamber and other components to be in contact with the test fluids was similar to that described in ref. 171. More details of the experimental work are given in ref. [13].

4. **RESULTS AND DISCUSSION**

Figure 5 exemplifies typical responses of A_{lens} , the plan-view area of each lens, to such vertical motions of Teflon and PMMA pipes as represented by x plotted against time t . Here x denotes the increase in the immersion depth of either pipe in the water after the instant $t = 0$, which was determined as the sum of the distance that the pipe had traveled downward displacing the water and the rise of the surface of the water. An increase and a decrease in x represent a descending and an ascending motion of the pipe. respectively. It is clearly shown in Fig. 5 that a lens is expanded by a descending motion of the Teflon pipe and an ascending motion of the PMMA pipe. We confirmed that this is just the same in cases of n-octane

FIG. 5. Expansion and shrinkage of n-decane lenses. The Teflon pipe (a) and the PMMA pipe (b) started to descend $(\frac{dx}{dt} > 0)$ at $t = 0$ and then ascended $(\frac{dx}{dt} < 0)$ in the case of the graphs on the left-hand side. The pipes first ascended in the case of the graphs on the right-hand side. Each symbol represents a class of data obtained with the same lens in each run. $T_{\infty} = 50.0$ °C, $V_0 = 140$ mm³.

and n-pentane lenses as well. This fact can well be explained by assuming such behaviors of the surface film as those illustrated in Fig. 4, though they probably oversimplify the real processes to some extent. The difference in the film behavior depending on the pipe material is reasonably ascribable to a difference in its hydrophobicity which should cause a difference in the orientation that the adsorbate molecules prefer to take on the pipe wall. In the following we show only the results obtained with the Teflon pipe.

Figure 6 shows the relation between the change in the lens area A_{lens} and the change in the total area A_{total} assumed to be covered by the film and the lens, while the Teflon pipe was driven downward, immediately a lens was formed, and then turned back as in the case of experimental runs shown on the graph on the lefthand side of Fig. 5(a). In Fig. 6 ΔA_{lens} is the change in A_{lens} from its initial value. A_{total} and related quantities are defined as follows *:*

$$
A_{\text{total}} = \pi D_{\text{p}}^2 / 4 + \Delta A_{\text{total}}
$$

\n
$$
\Delta A_{\text{total}} = \pi D_{\text{p}} x
$$

\n
$$
A_{\text{total}} = |dA_{\text{total}}/dt| = |d\Delta A_{\text{total}}/dt|
$$

FIG. 6. Relation between the change in lens area and that in the total area when the Teflon pipe was once lowered and then raised. Each symbol represents a class of data obtained with the same lens in each run: (a) n-decane, $T_x = 50.0^{\circ}$ C, $V_0 = 140$ mm³; (b) n-pentane, $T_x = 37.5$ °C, $V_0 = 60$ mm³.

where D_p designates the inside diameter of the Teflon pipe, 118 mm. It is recognized in Fig. 6 that each lens expands at a rate of 10-30% of that of the total area. In the course of compression, however. the shrinkage of the lens below its initial area is not remarkable. It seems that if the compression is continued, the lens takes asymptotically a constant area. In general, the dependency of the results on \dot{A}_{total} is not significant. This fact suggests that the factor controlling the lens behavior is the dilational elasticity *E* rather than the dilational viscosity κ . A detailed inspection of the results indicates that each n-decane lens shows larger areas in the compression process than in the preceding expansion process, and thus describes an anticlockwise loop on the $\Delta A_{\text{tens}}-\Delta A_{\text{total}}$ plane as directed by arrows in Fig. 6(a). This tendency is presumably ascribed to such a delay of the response of the lens shape to the change in σ_2 as caused by a hydrodynamic resistance inside the lens and that in the boundary

FIG. 7. Definitions of ΔA_{total} and a_{total} when the Teflon pipe was in a reciprocating motion.

layer beneath the lens. In the case of n-pentane lenses, however, *Alens* is always smaller in the compression process than in the preceding expansion process, resulting in a clockwise loop on the $\Delta A_{\text{lens}}-\Delta A_{\text{total}}$ plane as illustrated in Fig. 6(b). This seems to represent an appreciable loss of lens volume due to evaporation during the expansion and compression processes.

The effect of the expansion/shrinkage of each lens on its evaporation rate was investigated by reciprocating the Teflon pipe at a constant speed (0.6 \sim 20 mm s^{-1}) and a stroke (up to 40 mm), and thereby giving to A_{total} (or ΔA_{total}) such a saw-toothed oscillation as illustrated in Fig. 7. The variation of ndecane lens diameter $D = [(4/\pi)A_{\text{lens}}]^{1/2}$ with time in each evaporation process is demonstrated in Figs. 8(a) and (b) : the results obtained with the Teflon pipe held stationary are given in the former, and those obtained with the Teflon pipe in motion at a speed of 1.2 mm s^{-1} in the latter. It is evident that the time required for complete evaporation of each lens, t_v , was reduced by half as the result of the motion of the pipe.

In the case of the evaporation of n-pentane lenses which was much faster, irrespective of the Teflon-pipe motion, than that of n-decane lenses, we found that a much higher speed of pipe motion was necessary to promote the evaporation further. Figure 9 shows the results obtained in a series of experiments in which some different strokes of the pipe were selected while the speed was set at a 20 mm s^{-1} , the maximum speed available with our experimental apparatus. It is recognized that the evaporation time t_v drastically shortens as the stroke is increased, resulting in an increase in the amplitude a_{total} and a decrease in the frequency f of the oscillation of ΔA_{total} . Such an enhancement of evaporation should of course be ascribed in part to the oscillatory motion itself of each lens as well as of the surface film, as we expected. At the same time, the enhancement may well have a contribution from an increase in the average lens area. We noted that each lens is expanded to a larger extent than it is compressed by the motion of the Teflon pipe (Fig. 6), resulting in the lens area, averaged over each period of expansion/shrinkage, exceeding the area of a stationary lens with the same volume. We observed

FIG. 8. Variations of diameters of evaporating n-decane lenses in the absence (a) and presence (b) of reciprocating motion of the Teflon pipe. In (b), D_{max} and D_{min} indicate the maximum and the minimum of the oscillating diameter *D* of each lens: $T_{\infty} = 50.0$ °C, $V_0 = 140$ mm³. (a) Teflon pipe held stationary. (b) Teflon pipe in reciprocating motion, $\dot{A}_{\text{total}} = 480 \text{ mm}^2 \text{ s}^{-1}, a_{\text{total}} = 1200 \text{ mm}^2, f = 0.1 \text{ Hz}.$

FIG. 9. Dependency of evaporation time t_v of n-pentane lens on the amplitude of oscillation of the total film/lens area, a_{total} . The data points on the ordinate, where $a_{total} = 0$, represent the measurements of t_v in the absence of any motion of the Teflon pipe: $T_{\infty} = 37.5$ °C, $V_0 = 60$ mm³, and $A_{\text{total}} = 7800$ mm² s⁻¹ (except for the data points on the ordinate).

that at the largest stroke of the pipe, 40 mm, D_{max} was three to four times as large as D_{\min} , and hence we can assume that the average lens area at each stage of evaporation was significantly increased over the area that a lens with the same volume would take in the absence of the pipe motion. At present we cannot evaluate the effect of the oscillatory motion and that

of an increased area of each lens on its evaporation separately.

One may suspect that the enhanced evaporation encountered in our experiments had also a contribution from a convection in the bulk and some vibration at the surface of the substrate water both caused by the direct momentum transfer from the pipe wall irrespective of the presence of the surface film. In order to evaluate these possible effects which can confuse the interpretation of the experimental results, we repeated the evaporation experiments with a ring of 108 mm in diameter, made of a Teflon tubing of 2 mm o.d., floating on the water surface. Each lens was formed on the water surface confined by the ring, and it evaporated, while being insulated from the surface film which was spread outside the ring and hence subjected to the expansion and compression responding to the pipe motion. We observed, under this condition, little expansion/shrinkage of evaporating lenses and detected no reduction in the evaporation time t_v . Thus, we can conclude that the enhancement of evaporation as recognized in Fig. 9 should be ascribed to the viscoelastic behavior of the surface film connecting each lens to the moving pipe wall.

5. **CONCLUDING REMARKS**

The work reported in this paper has demonstrated a potential utility of the surface dilational viscoelasticity in enhancing a kind of 'interfacial heat transfer'. It has been found that the direct contact evaporation of discrete organic-liquid lenses floating on the water surface can be enhanced extensively by applying an external force in such a way that it is transmitted to each lens through an adsorbed surface film spread at the water surface around the lens, causing a periodical expansion/shrinkage of the lens. The vertical-pipe device that we used in the present work is no more than an invention for laboratory experiments, and probably has little applicability to practical evaporators. Other devices to drive surface films are probably conceivable, which are appropriate for particular applications.

The surface viscoelasticity, which is the key to the enhancement technique dealt with in this work, is presumably highly dependent on the degree of contamination by surfactant impurities in the experimental system. The results obtained in this work are considered to represent the surface viscoelasticity in a fairly clean experimental system. If the system is contaminated to a larger extent as expected in practical applications, the surface viscoelasticity will become more prominent, which is a favorable condition in applying the present enhancement technique.

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ESSAI D'ACCROISSEMENT DE L'EVAPORATION PAR CONTACT DIRECT A PARTIR DE LA VISCOELASTICITE

Résumé-On tente de montrer l'utilité potentielle du "film de surface" dans l'accroissement du "transfert de chaleur interfacial". Le problème particulier considéré est d'augmenter le transfert de chaleur-et l'évaporation-de lentilles isolées d'un liquide volatil flottant sur la surface d'un bain stagnant d'eau. En provoquant à l'aide d'une force externe des étirements et des compressions alternées d'un film de surface étalé sur la surface de l'eau autour de chaque lentille, on réussit un étalement et rétrécissement alternés qui provoque une réduction du temps d'évaporation des lentilles.

EIN VERSUCH ZUR VERBESSERUNG DER DIREKTKONTAKTVERDAMPFUNG UNTER AUSNUTZUNG DER DEHNUNGS-VISKOELASTIZITAT DER OBERFLACHE

Zusammenfassung-Diese Arbeit versucht, eine mögliche Anwendung des "Oberflächen-Films" zur Verbesserung des Warmeiibergangs an Beriihrungsllachen aufzuzeigen. Das speziell betrachtete Problem ist der Wärmeübergang an-bzw. die Verdampfung von-einzelnen Tropfen einer leichtflüchtigen Flüssigkeit, die auf einer ruhenden Wasseroberflache schwimmen. Durch abwechselndes Auseinanderziehen und Verdichten eines Oberflächen-Films, der um jeden Tropfen herum aufgebracht wird, mittels einer äußeren Kraft, gelang es, die Tropfen zu periodischem Ausdehnen und Schrumpfen zu veranlassen. Dieses führte zu einer deutlichen Verkiirzung der Verdampfungszeit der Tropfen.

ПОПЫТКА ИНТЕНСИФИКАЦИИ ПРЯМОГО КОНТАКТНОГО ИСПАРЕНИЯ ПУТЕМ ДЕФОРМИРОВАНИЯ УПРУГОВЯЗКОЙ ПОВЕРХНОСТИ

Аннотация- Работа предпринята с целью показать потенциальную возможность использования "поверхностной пленки" для интенсификации "межфазного теплообмена". В частности, рассматривалась возможность интенсификации теплопереноса к отдельным линзам летучей жидкости, плавающим на неподвижной поверхности воды, и их испарение. Попеременно растягивая и **CXGiMall (C llOMOlUbH3 IIpHJIOYeHHOii U3BHe CHJIbI) UOBepXHOCTHyiO IIJIeHKy Ha IIOBepXHOCTH B0LU.i** вокруг каждой линзы, оказалось возможным периодически растягивать и сжимать линзы, что значительно сокращало время их испарения.