INTERFACIAL TURBULENCE IN MINUTE DROPS EVAPORATING ON A FLAT PLATE

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ABSTRACT
      Surface tension/temperature driven interfacial turbulence
      in evaporating minute drops was studied photographyically
      using the laser shadowgraphy. Liquids of relatively low
      boiling point were employed: ethyl ether, acetone, methanol,
      carbon tetrachloride, benzene and heptane. A drop surface
      on a glass plate at room temperature was photographed
      straight down with motion picture. During drop lifetime,
      the interface exhibited a dominant pattern of radial
      stripes over a very brief duration followed by polygonal
      cells and ended in ripples. These interfacial forms are
      related to three stages of drop evaporation. The mechanism
      of interfacial turbulence is proposed.

Introduction

     The occurence of spontaneous, highly localized, interfacial
agitation accompanying mass transfer was first reported by Ward
and Brooks [1] in 1952. This phenomenon known as interfacial
turbulence can cause an order-of-magnitude increase in mass
transfer processes such as gas absorption, liquid-liquid extraction
and evaporation of mixtures on flat or curved interfaces.
Sternling and Scriven [2] treated the phenomenon as an evidence
of hydrodynamic instability, analyzing a simplified, two-dimensional
model (roll cells) by means of the theory of linearized stability
coupled with the hydrodynamic and mass diffusion principles. The

model assumed that under favorable conditions, small, ever-
present fluctuations of concentration or temperature about the
interface may be amplified into fully developed flows spontan-
eously, through the Marangoni effect. Such flow may either be
of a highly irregular pattern corresponding to interfacial
turbulence or of a cellular pattern called cellular convection.
The theory developed the conditions for the initiation of inter-
facial activity and determined the nature of the dominant dis-
turbance, predicting its wave length, amplification factor,
speed of propagation, and temporal period. Using schlieren
photography, Orell and Westwater [3,4] followed the development
of polygonal cells which changed to stripes and later evolved
into ripples. Polygonal cells and stripes are similar to those
observed by Benard for heat transfer. However, the ripples are
unexpected from the Sternling-Scriven theory. Later, Ellis and
Biddulph [5] observed vigorous rippling on the surface of water
during the absorption of acetone or methanol vapor. Using a
capacitance probe, they measured wave amplitudes of up to 0.2 mm,
proportional to the concentration gradient. Instantaneous
surface velocities about 10 cm/sec occurred over distances of 1 cm,
penetrating 1 mm into the liquid.

In some chemical absorption processes (gas absorption
accompanied by a simultaneous chemical reaction), interfacial
turbulence produced by surface tension gradients causes a sub-
stantial increase in the rate of gas absorption [6]. Ruckenstein
and Berbente [7] determined the conditions under which the
Marangoni effect produces interfacial turbulence in the case of
mass transfer accompanied by chemical reaction.

In case mass transfer takes place across rounded interfaces,
disturbances of the same sort occur. The interface is, in many
cases, disturbed by rippling, while the adjoining liquid is
turbulently agitated [for example 8,9]. At times, there are
localized eruptions at the interface. If it is pendent, the
entire drop often pulsates violently and erratically; unattached
drops behave in the same way.

In temperature-driven flows, surface-tension effects can
cause instability in thin liquid layers, for example [10-13].

Reference 13 reveals three basic structural forms of flow in pools
of liquids evaporating at room temperature (the fourth pattern,
vermiculated rolls, appears only in contaminated surface):cellular
network, "ribs" and "streamers" for depths of 2 mm or less, 3 to
5 mm and over 7 or 8 mm, respectively.

Kenning [14] pointed out that random surface motion during
evaporation of thick liquid layers induced by local variations
in temperature would become more pronounded when associated
with changes in composition.

The evaporation of liquid drops on a heated surface has
important applications to spray cooling, liquid fuel drop
combustion, etc. Research efforts have been concentrated on the
determination of either drop lifetime [for example 15] or heat
transfer rate [for example 16, 17]. Earlier studies indicate
moderate to insignificant contribution of the Marangoni effect
to heat transfer inside a drop during condensation [18] and in
the liquid surrounding a bubble [19] excluding interfacial
instability. Recently, the problem of thermal instability in
drops has begun to attract attention [20]. By employing a linear
stability theory, reference 20 developed the criterion for the
onset of surface-tension induced cellular motion in a hemispherical
drop on a solid surface. However, interfacial turbulence of the
Marangoni type in drop evaporation has not yet been fully explored.
Like in the afore-mentioned mass transfer cases, such localized
stirring at the interface would undoubtly lead to high heat trans-
fer coefficients accompanied by high rates of interfacial phase
change.

The present study is to explore the nature and pattern of
the interfacial activity of minute liquid drops evaporating on
a flat glass plate.

Experimental Apparatus and Procedure

The interfacial activity in the evaporating drops was
observed using a conventional shadowgraphic technique, which
permits us to see and photograph second derivatives of refractive

index (and hence second derivatives of temperature) in a trans-
parent medium. The principles of laser illumination and filter-
ing and the method of shadowgraphy have been described in great
detail in references 21 and 22-23, respectively.

The shadowgraphic arrangement consisted of an optical
system, a test object, and a photographic equipment, as shown
in Fig. 1. The test object was a transparent medium, namely
a minute drop of evaporating liquid, on an optically flat glass
plate, 102 mm x 127 mm in size and 3 mm thick. The optical system
consisted of five components: light source, achromatic objective,
pinhole, a pair of surface mirrors and collimating lens.

A C.W. Radiation Model SP 2, 2.5 mW cylindrical helium-neon
laser was used as the light source. The 20x achromatic micro-
scopic objective was mounted on a frame together with the 10-micron
precision pinhole. The collimating lens was placed at its focal
length from the pinhole. These four components were aligned on a
horizontal bench. Invariably a laser beam contains diffraction
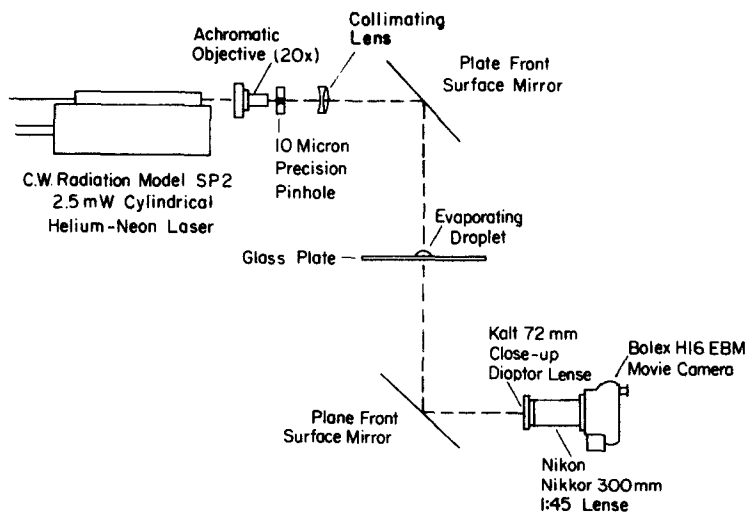noise, flashlamp, or plasma illumination and transverse mode

FIG. 1
A Schematic of the Shadowgraphic System for Photographic
Study of Minute Drops Evaporating on a Solid Surface

structure. Much of this can be removed by a procedure of spatial
filtering-the beam is focused by a high quality lens, the objec-
tive, through the tiny pinhole used to control apertures of laser
beams and as a spatial filter. The parallel beam of light emerging
from the lens was made to pass through the evaporating drop from
top to bottom (thus producing a top view of the interfacial
activity) by an arrangement of two plate front surface mirrors,
102 mm x 127 mm and 6 mm thick, as shown in Fig. 1. The two mirrors
are aluminized on surface nearest objects to be viewed so that
light may be reflected with no refraction. They were mounted in
parallel on a specially-designed frame at 45 degrees. The test
plate was inserted into the vertical light beam between the two
mirrors. The mirror arrangement was used to obtain a horizontal
view of the evaporating drop. The optical path from the lower
mirror was horizontal into a 16-mm Bolex H16 EBM movie camera
having a Nikon Nikkov 300 mm 1:45 lense. A Kalt 72 mm Close-Up
Dioptor lense was mounted on top of the Nikon lense to photograph
the air-liquid interface of an evaporating drop. A drop was carefully

placed on the test plate by means of a 1-μl Monoject micro
syringe with a bent needle. The needle tip was touching the
plate surface before and during the injection to permit the
formation of a calm, unsplashing drop.

Photographic Observations

Various liquids of low boiling point were studied including
ethyl ether, acetone, methanol, carbon tetrachloride, benzene,
heptane and water. During the course of evaporation, the inter-
face exhibits a variety of geometrical patterns, depending upon
mainly the initial size and to a much lesser degree on the physical
properties of the liquid. In general, three basic patterns are
observed: radial stripes, polygonal cells, and ripples.

Radial stripes - This pattern is first to appear on the
interface as soon as the drop is released from a needle tip.
All stripes radiate from the surface outward to the drop peri-
phery, as shown in Fig. 2(a). It exists very briefly and the
pattern undergoes rapid changes into a cellular structure. The
stripes represent "cold lines" where cool fluid would plunge in
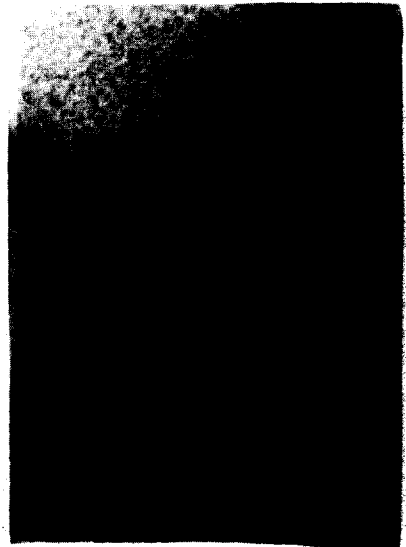sheets from the surface into the interior. Morphogically, they
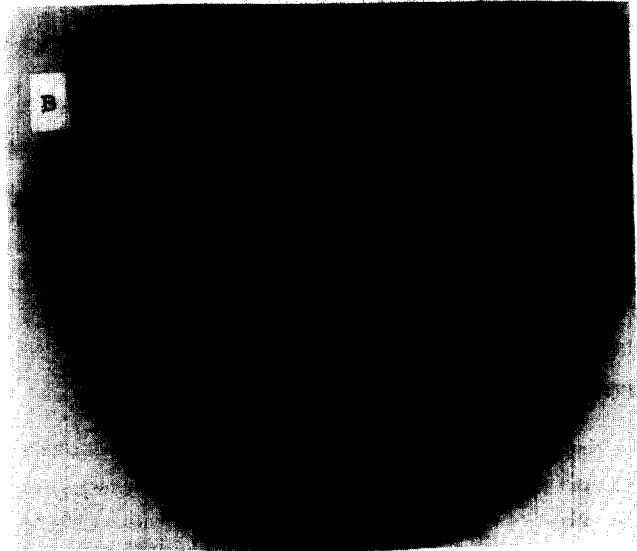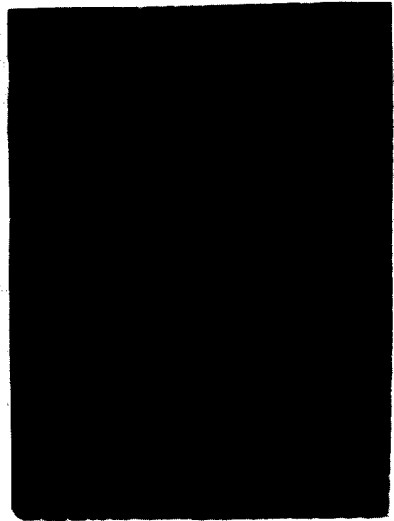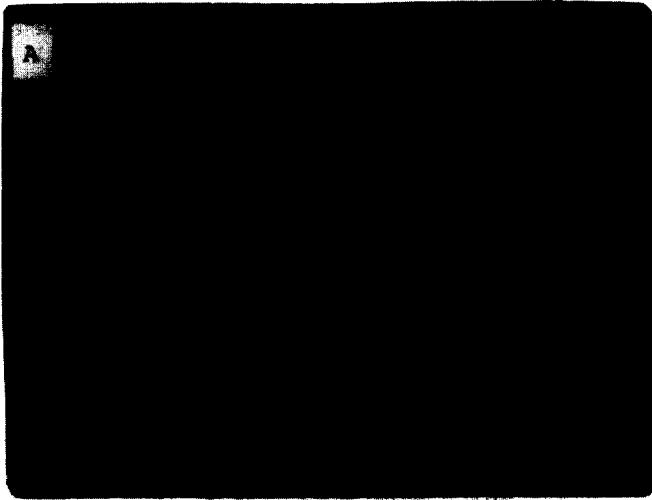are ridges.

FIG. 2

Typical Interfacial Flow
Patterns in Minute Drops

Polygonal Cells - This pattern usually does not encompass the whole surface, leaving some regions bare and inactive as brigth areas in Fig. 2 (b). The dark oval background represents the laser light. The cells froming the network are mostly irregular polygons with cell partitions well defined. Warm fluid from the interior rises to the surface at each cell center and returns to the interior along the cell partitions as cooled liquid. Each is a Benard cell constituting a small torus independent of the other cells. As time passes, the drop thickness reduces, particularly more rapidly along the drop periphery within which the liquid forms a ring of very thin film. Very closely spaced fine concentric ripples are observed in the very thin liquid film. The width of the ring grows with its innner periphery moving radially inward faster than the retreating of the outer periphery as the drop continues to evaporate. Meanwhile, the thickness of the drop proper within the ring grows thinner and thinner accompanied by a violent variation of the cellular patterns in size and shape.

Ripples - In the final stage of drop evaporation, unconfined ripples grow rapidly inward taking over the area occupied by cellular patterns, like a flood. Often the process proceeds so fast, leaving the traces of large cellular patterns superimposed on the background of unconfined ripples as shown in Fig. 2(c). Depending on the initial shape of the drop, sometimes, a perfect concentric form of ripples is observed, Fig. 2(d). Its periphery rapidly retreats inward until the completion of liquid evaporation. Ridges appear as darkened lines corresponding to "cold lines" at lower temperatures, while troughs emerge as bright lines represent· ing "hot lines" at higher temperature.

The above-mentioned anomalous behavior of water disclosed in the case of pool evaporation [13] was also observed in the present study. As shown in Fig. 2(e), the motion picture recorded no trace of interfacial activity in the evaporating drop of various minute size at room temperature. The fluid is characterized by higher thermal conductivity and viscosity which, during the pre-convection period are capable of damping and destroying small thermal and mechanical disturbances such that no fluid motion results. From the viewpoint of thermocapillary effect, water has substantially higher surface tension and relatively low surface

tension-temperature gradient. This combined feature discourages
the occurence of interfacial activity. On the other hand, subst-
antially low value of the coefficient of thermal expansion and
high kinematic viscosity are not favorable in causing convective
flow by the buoyancy driving mechanism in the case of large drops.

Discussion of Results

    Very vigorous and irregular instabilities occur during the
evaporation of a drop on a solid surface. The random motion of
warm liquid toward the interface to supply the heat for evaporation
results in turbulence thus reinforcing the rate of evaporation.
Unconfined ripples on the interface are observed in the final

stage of drop lifetime. A mechanism is proposed to explain
the cause of such an interfacial turbulence.

    Since the surface tension is temperature dependent, pre-
ferential phase change will give rise to surface tension difference
between adjacent slots (lines) or spots (points) in the liquid
interface. Slots of preferential evaporation will have a lower
surface tension than the neighboring interface liquid and will
tend to spread. Sternling and Scriven [2] described these slots
or spots as "small, ever-present disturbance" of concentration
or temperature about the interface in their hydrodynamic instability
theory of interfacial activity. The driving force is the
temperature difference between the plate surface and the ambient.
Low driving forces produce small surface tension differences.
Consequently interface slots will spread slowly causing slight
rippling which decays eventually. As the driving force and the
rate of preferential evaporation increase, the surface tension
gradient increases resulting in greater rippling. At some
critical driving force, the rate of evaporation is strong enough
to instantaneously produce slots of very steep surface tension
gradient. The resulting spreading is so rapid that the momentum
of the spreading liquid is sufficient to break the low surface
tension slot, thus exposing subjacent liquid drawn from below the
interface. A pair of ridges (or an annulus in the case of spot)
of low surface tension are then formed and expand outward, leaving
behind a region of high surface tension which tends to reverse
the spreading motion. This causes liquid to rush from the bulk

and from the spreading film toward the high surface tension region, resulting in a build up of liquid into another ridge (or a large ripple in the case of spot). Finally, both "hot lines" (warm troughs) and "cold lines" (cold ridges) both of which are called "streamers" are formed in the interface. The sequence of events is illustrated in Fig. 3.

The-first-power-of-the-diameter law is valid for the evaporation of a single drop on a heated plate [24]: $D_o^2 - D^2 = kt$, where D is the base diameter of the drop at the time t; D_o, its initial value; and k, the evaporation constant. One can classify the time

PREFERENTIAL
EVAPORATON

SURFACE FORCE

SLOT OF LOW
SURFACE TENSION

SLOT SPREADS,   EXPOSING
FORMING RIDGES   BULK LIQUID

CENTRAL
MOTION REVERSES,
FORMING RIDGE

COLD RIDGES   WARM TROUGHS
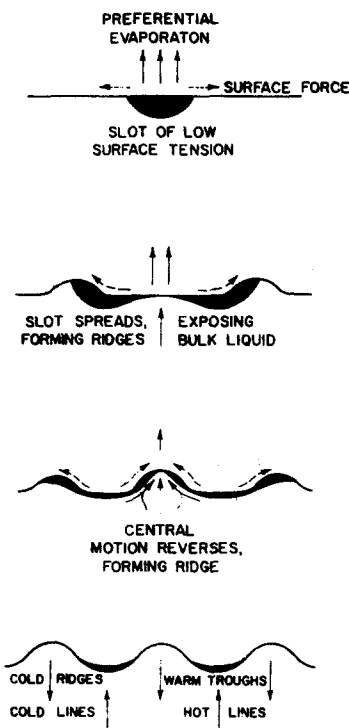
COLD LINES   HOT LINES

FIG. 3
A Model for the Mechanism of
Interfacial Turbulence in a
Minute Drop Evaporating on a
Solid Surface

history of drop evaporation into three stages according to the rate of surface regression $dD/dt=k/[2(D_o^2-kt)^{1/2}]$ which varies from $k/(2D_o)$ to infinity: In the initial stage of evaporation where surface regression is nearly constant at $k/(2D_o)$, the interface is dominated by radial stripes. The intermediate stage that follows begins when the drop surface withdraws more rapidly with time, during which polygonal cells appear in the interface. Finally, ripples encompass the interface in the asymptotic stage of evaporation where the surface regression rate is extremely high.

In the case of condensation, heat transfer causes an increase in surface tension. Hence a localized spot of preferential condensation has a higher surface tension than its surrounding interface liquid. This spot has no tendency to spread. With no motion being induced, there is no ripple formation and therefore the interface remains calm.

Concluding Remarks

Three fundamental structural forms of interfacial flow, radial stripes, polygonal cells and ripples, are disclosed and related to three stages of drop evaporation. The mechanism of ripple formation is proposed as caused by the liquid-air interface surface tension changes in a negative direction. The effect of fluid properties on interfacial turbulence is discussed. The study provides some insight into interfacial activities in drop evaporation on a plate. However, effort must be continued, in particular, to investigate the wave characteristics of interfacial turbulence such as the wavelength and speed of propagation.

Acknowledgement

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