

Theory and experiments on evaporating sessile drops of binary liquid mixtures

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Abstract—Theoretical and experimental studies are performed to determine the size, history and lifetime of evaporating sessile drops of binary liquid mixtures. The minimum and maximum drop lifetimes (equivalent to the maximum and minimum vaporization rates, respectively) occur at the azeotropic compositions of the positive and negative azeotropes, respectively. The mechanisms leading to the extremum drop lifetime are due to the changes in composition of the more volatile component during the vaporization process. This composition change tends to suppress vaporization. A new dimensionless parameter is found to determine the intensity of vaporization of a binary liquid drop. Theoretical predictions of the drop size, history and lifetime agree satisfactorily with experimental results.

1. INTRODUCTION

VAPORIZATION is the process of mass transfer from the liquid phase into the vapor phase. It may exhibit itself in two basic types: boiling and convective type vaporizations. When the surrounding temperature T_∞ is equal to or higher than the boiling temperature T_b , the vaporization rate, to a great extent, is controlled by the heat flux from the surroundings to the drop. This is the boiling type vaporization. On the other hand, if T_∞ is below T_b , the vaporization process is governed by mass diffusion of the vapor into the ambient. The concentration of the vapor at the drop surface is determined by the vapor–liquid phase equilibrium of the vapor–air mixture.

In the case of the binary liquid mixtures, the equilibrium vapor pressure P depends on both the temperature T and composition, \bar{x}_i or \bar{y}_i . \bar{x}_i and \bar{y}_i denote the mole fractions of component i in the liquid and vapor phases, respectively. The binary vapor–liquid equilibrium (VLE) may be expressed two-dimensionally in two different forms T vs (\bar{x}_i, \bar{y}_i) at constant P and P vs (\bar{x}_i, \bar{y}_i) at constant T . In both forms, the phase equilibrium diagram is characterized by two curves, called bubble- (or boiling-) and dew-point curves representing the states of saturated liquid and vapor, respectively. For non-azeotropic binary mixtures such as carbon disulfide–benzene mixtures, the bubble- and dew-point curves form a ‘lens’ shape in both the T – x and P – x diagrams, not shown. The significant feature of Figs. 1(a) and (b) is the occurrence of a state of intermediate composition at which the equilibrium liquid and vapor compositions are

identical. Such a state is called an azeotrope which is characterized by the extremum on both the bubble- and dew-points curves under the isothermal conditions. The mixture with the maximum is called the positive azeotrope, while that with the minimum is referred to as the negative azeotrope [1].

It is known that the addition of a foreign component into a pure liquid results in natural convection with the aid of solutal effects. Two important phenomena were disclosed in the boiling type vaporization in binary liquid mixtures. One was the nucleate pool boiling in a large volume of non-azeotropic mixtures, which disclosed the coincidence of the minimum bubble growth rate and the maximum heat flux at the same composition. This peculiar phenomenon, called ‘boiling paradox’, has attracted considerable interest in the past decade [2]. The other phenomenon was the disclosure of the minimum lifetime (or equivalently the maximum evaporation rate) of an azeotropic binary-liquid drop evaporating on a plate in the nucleate-boiling type evaporation regime with the plate temperature between the boiling point and the DNB (deviation from nucleate boiling) point [3].

Recently, real-time holographic interferometry was employed to determine the concentration effect on the binary drop lifetime in the whole concentration range of 0–100% volume ethyl ether in chloroform [4]. The minimum drop lifetime was discovered at an intermediate composition. An examination revealed that the composition corresponding to the minimum lifetime coincided with the azeotropic composition of the binary mixture which is a positive azeotrope. The

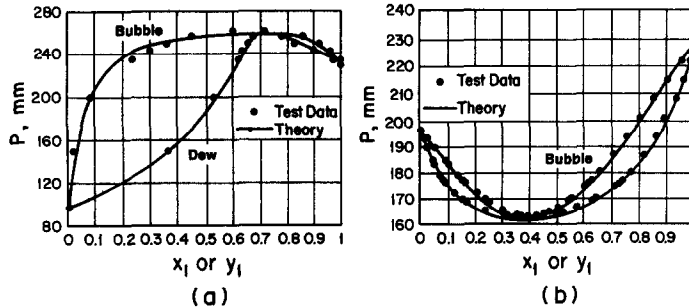


FIG. 1. Binary vapor-liquid phase equilibrium (P - \bar{x}) diagrams at 25°C for: (a) positive azeotrope, acetone-cyclohexane mixtures; (b) negative azeotrope, acetone-chloroform mixtures. data from ref. [1].

to other evaporation regimes and, in particular, are irrelevant to drop combustion. It is important to note that the famous d^2 law for drops in spheroidal combustion is not applicable to the present study. The study is based on the perfect mixing model and can be applied to spray cooling in materials processing and energy shutdown of nuclear reactors.

2. THEORY

Transport phenomena related to the vaporization of a binary liquid drop on a substrate are very complex, involving buoyancy- and thermocapillary-induced double diffusion, drop shape and couplings at the phase interfaces. It is difficult to analytically or numerically obtain the general solutions. Therefore, the following assumptions are imposed in order to simplify the formulation.

(1) Because of a minute size with vigorous internal circulation [5], the drop is a lumped system, uniform in both temperature and concentration at any instant. The perfect mixing model is appropriate for the following reasons.

(a) The drop forms a thin liquid layer of lens shape with a maximum height of 0.2–0.3 mm.

(b) Rigorous flow circulation takes place within the liquid phase as revealed by flow visualization [5].

(c) The phenomenon is entirely different from that which occurs in drops under spheroidal combustion during which the drop is spherical and stagnant. It should be noted that for the case with liquid-side heat transport by conduction and gas-side transport by natural convection, the Biot number is estimated to be less than 0.1 for the liquids used in the present study. The flow may not be laminar due to the effects of Marangoni instability [6]. The circulating flow can substantially promote the uniformity in temperature and concentration distribution within the drop.

(2) All physical properties remain unchanged during the evaporation process.

(3) The gas-side transport phenomena are simplified through the use of heat and mass transfer coefficients (h and h_D , respectively) between the drop surface and the ambient. The assumption is valid since

the drop is placed on a plate so that the change of h_D and h with drop size is small.

The mass and heat balance for the drop read, respectively

$$\frac{dm}{dt} = -h_d A_s (C_b - C_\infty) \quad (1)$$

$$m C_p \frac{dT}{dt} = -h A_s (T - T_\infty) + q_s - h_D A_s (C_b - C_\infty) h_{fg} \quad (2)$$

The symbols are defined in the Nomenclature. The change of compositions in the binary liquid drop is derived from the discontinuity of concentrations at the drop surface as

$$\frac{dx_i}{dt} = -\frac{x_i - y_i}{m} h_D A_s (C_b - C_\infty) \quad (3)$$

Here, x_i and y_i denote the mass fractions in the liquid and vapor phases, respectively, under phase equilibrium.

The drop surface area A_s is related to the drop volume V by the shape factor F as

$$A_s = FV \quad (4)$$

For the drops of spherical-segment shape, F can be further generalized as the product of two quantities: one depends on the drop volume as $2(\pi/v)^\alpha$, while the other quantity ψ is insensible to the volume change. It reads

$$F = 2(\pi/v)^\alpha \psi \quad (5)$$

Here, α and ψ are named shape parameters which vary with only the shape that the drop forms on the substrate. The sessile drops undergoing stable interfacial type evaporation take a lens shape which can be idealized as a spherical segment, as depicted in Fig. 2. In general, α and ψ can be determined by two of the four variables R , r , l and β . Since the drop shape depends on the temperature, compositions and drop volume which vary with time, the shape parameters are time dependent. The role of drop shape on the evaporation process is presented later in Section 3.

During the vaporization process with the shape of a spherical segment, three special cases are of interest.

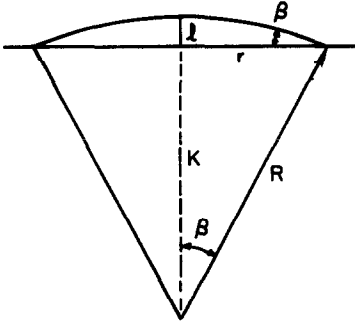


FIG. 2. Geometry of a spherical-segment drop on a flat plate.

(1) Prescribed base radius r : with a given r , the surface area–volume ratio of the spherical segment is obtained and compared with equation (5). It yields

$$\alpha = 1; \quad \psi = \frac{1}{2}r^2 \left[1 + \left(\frac{l}{r} \right)^2 \right]. \quad (6)$$

For $(l/r)^2 \ll 1$, ψ is approximately $r^2/2$ which stays nearly constant. Liquids such as acetone, ethanol, methanol, and ethyl acetate belong in this category [5].

(2) Prescribed radius of curvature R : similar to the procedure described in equation (1), one obtains

$$\alpha = \frac{1}{2}; \quad \psi = R^{1/2} \left[1 - \left(\frac{l}{3R} \right)^2 \right]^{-1/2} \quad (7)$$

For $(l/3R) \ll 1$, ψ can be approximated as $R^{1/2}$ and remains almost constant. Cyclohexane, methylene chloride, ethyl ether and benzene are of this type [5].

(3) Prescribed contact angle β : in this case, it yields

$$\alpha = \frac{1}{3}; \quad \psi = \left[\frac{1 - \cos \beta}{1 - \frac{1}{3}(1 - \cos \beta)} \right]^{1/3} \quad (8)$$

A liquid of this type is chloroform [5].

In general, α takes a value between $1/3$ and 1 , while the value of ψ does not change significantly with a reduction in drop size. If the drop temperature variation is small, α and ψ for pure liquid drops may be considered constant during vaporization. In the case of binary liquid drops, however, a mixing rule can be imposed to describe the dependence of the shape factors of individual constituents on their compositions. The present study uses

$$F = \prod_{i=1}^2 f_i^{\bar{x}_i} = 2 \left(\frac{\pi}{V} \right)^{\sum_{i=1}^2 \bar{x}_i \alpha_i} \prod_{i=1}^2 \psi_i^{\bar{x}_i}. \quad (9)$$

Here, α_i and ψ_i denote the shape parameters for pure liquid drops at the same temperature and initial volumes, while \bar{x}_i represents the mole fraction of component i in the binary liquid phase. The mixing rule employed in this study is based on the boundary condition that the shape factor of a pure liquid component should be recovered when the mole fraction of that component becomes unity.

The concentration difference ($C_b - C_\infty$) is the driving force for drop vaporization. For simplicity C_∞ may be assumed zero, while C_b depends on the VLE conditions of the binary system. The conventional thermodynamic approach [7] yields

$$C_b = \frac{M_{bv} P_b}{\bar{R} T \phi_b} \exp \left[\int_{P_b}^P \bar{v}_{bl} dp \right]. \quad (10)$$

The exponent is called the Poynting correction. It is nearly unity when the liquid molar volume \bar{v}_{bl} is small and the total system pressure is moderate. Equation (10) then becomes

$$C_b = \frac{M_{bv} P_b}{\bar{R} T \phi_b}. \quad (11)$$

Here P_b is the equilibrium vapor pressure of the binary system at temperature T and liquid phase composition \bar{x}_i . The mean molecular weight of the binary vapor at the phase composition \bar{y}_i is denoted by M_{bv} , while ϕ_b is the correction parameter (similar to the fugacity coefficient) for non-ideal mixing of the binary vapor with the surrounding gases.

To simplify the analysis, it is assumed that heat transfer from the surrounding atmosphere is negligible and that the heat of vaporization comes from the substrate through the drop base. Energy equation (2) can thus be excluded from the consideration. The main objective of the study is to determine the effects of binary composition on the drop lifetime. Now, equations (4) and (9) are substituted into equations (1) and (3), followed by non-dimensionalizing the variable using

$$m^* = \frac{m}{m_0}; \quad F^* = F V_0^{1/3}; \quad v_b^* = \frac{\bar{v}_b}{\bar{v}_{b0}} \\ t^* = \frac{t h_D}{V_0^{1/3}}; \quad P^* = \frac{P_b}{P_{ba}}. \quad (12)$$

After some manipulation, it yields

$$\frac{dm^*}{dt^*} = -m^* G \quad (13)$$

$$\frac{dx_1}{dt^*} = -(x_1 - y_1) G. \quad (14)$$

Here

$$G = Z_a \frac{\bar{v}_b^* P_b^*}{\phi_b} \left(\frac{\bar{v}_{ba}}{\bar{v}_{b0}} \right) \left(\frac{M_{bv}}{M_{bl}} \right) \\ \times \left[2 \left(\frac{\pi}{m^* v_b^*} \right)^{\sum_{i=1}^2 \bar{x}_i \alpha_i} \prod_{i=1}^2 \psi_i^{*\bar{x}_i} \right]. \quad (15)$$

The dimensionless shape parameter, ψ^* , for a pure liquid i , can be expressed as:

case 1

$$\psi^* = \frac{1}{2} \left[\frac{\sin \beta_i}{\pi^{1/3} (1 - \cos \beta_i)^{2/3} [1 - \frac{1}{3}(1 - \cos \beta_i)]^{1/3}} \right]^2; \quad (16a)$$

case 2

$$\psi^* = [\pi^{1/3}(1 - \cos \beta_i)^{2/3} [1 - \frac{1}{3}(1 - \cos \beta_i)]^{1/3}]^{-1/2}; \quad (16b)$$

case 3

$$\psi^* = [(1 - \cos \beta_i)[1 - \frac{1}{3}(1 - \cos \beta_i)]^2]^{-1/3}; \quad (16c)$$

where β_i is the initial contact angle of a pure liquid i on the substrate.

The variables T , P_b , x_i , and y_i ($i = 1, 2$) in equations (13)–(15) are the intensive properties in the VLE. According to the Gibbs phase rule, only two of the six variables are independent. If T and P are chosen as independent variables, the other four must be determined from the conditions of phase equilibrium

$$\bar{f}_i^V = \bar{f}_i^L \quad (17)$$

for $i = 1, 2$ and the constraints

$$\sum_{i=1}^2 \bar{x}_i = \sum_{i=1}^2 \bar{y}_i = 1. \quad (18)$$

The partial molar fugacities, \bar{f}_i^V and \bar{f}_i^L for the vapor and liquid phases, respectively, can be written as [7]

$$\bar{f}_i^V = \phi_i \bar{y}_i P_0 \quad (19)$$

$$\bar{f}_i^L = \gamma_i^0 \bar{x}_i f_i^{0L} \exp\left(\frac{P_b \bar{v}_i}{RT}\right) \quad (20)$$

for $i = 1, 2$. Introducing the ratio of mole fractions $k_i = y_i/x_i$, one can write

$$k_i = \frac{\bar{y}_i}{\bar{x}_i} = \frac{\gamma_i^0}{\phi_i P_b} f_i^{0L} \exp\left(\frac{P_b \bar{v}_i}{RT}\right) \quad (21)$$

for $i = 1, 2$.

The method given in ref. [8] can be used to determine ϕ_i , γ_i^0 , f_i^{0L} and v_i . Alternately \bar{v}_b can be evaluated using the same equation for a pure liquid in terms of Kay's rule. However, ϕ_b for a multicomponent system is hard to predict under equilibrium conditions. It is approximately unity for non-azeotropic mixtures, because a dilute mixture in the gaseous phase behaves like an ideal gas under moderate pressure. For azeotropic binary vapors, ϕ_b deviates from unity, reaching the extremum at the azeotropic composition \bar{x}_a . Therefore, one can write

$$\phi_b = 1 + \xi \quad (22)$$

where ξ is a weak function of compositions in the vapor phase. ξ must satisfy the following constraints:

$$\xi \rightarrow \text{extremum, as } \bar{y} \rightarrow \bar{x}_a \quad (23)$$

$$\xi \rightarrow 0, \text{ as } \bar{y} \rightarrow 0 \text{ or } 1. \quad (24)$$

ξ which satisfies constraints (23) and (24) takes the form

$$\xi = \varepsilon_b S_a(\bar{y}). \quad (25)$$

Here

$$S_a = \left(\frac{1 - \bar{y}}{1 - \bar{x}_a}\right) \left(\frac{\bar{y}}{\bar{x}_a}\right)^B \text{ for } \bar{x}_a \geq 0.5 \quad (26)$$

$$S_a = \left(\frac{1 - \bar{y}}{1 - \bar{x}_a}\right)^{1/B} \left(\frac{\bar{y}}{\bar{x}_a}\right) \text{ for } \bar{x}_a \leq 0.5 \quad (27)$$

$$B = \frac{\bar{x}_a}{1 - \bar{x}_a}. \quad (28)$$

The non-ideal mixing factor ε_b varies with composition, temperature and pressure. It is determined by curve-fitting the test data.

3. RESULTS AND DISCUSSION

The mass balance and composition equations, equations (13) and (14), respectively, are subject to constraints (18) and binary vapor–liquid phase equilibrium conditions (21). The governing equations are numerically integrated by the Runge–Kutta–Gill method. Results are presented in Figs. 3(a) and 4(a) for acetone–cyclohexane mixtures at 14°C.

It is seen in Fig. 3(a) that the time history of evap-

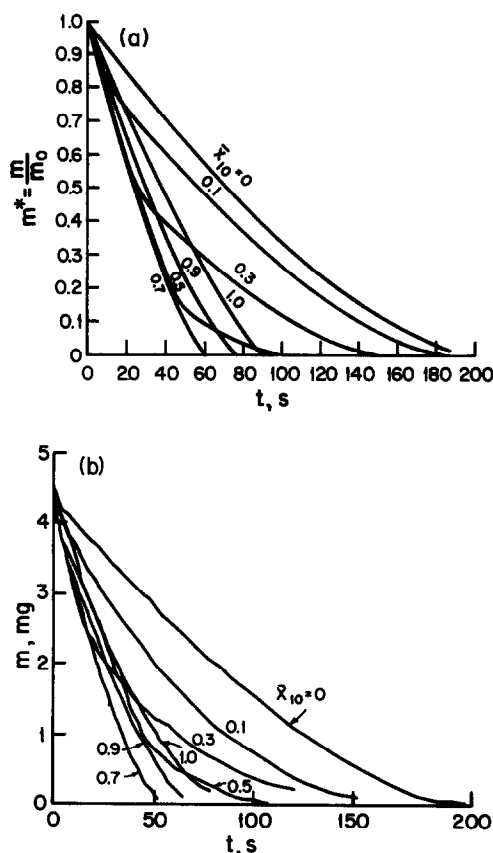


FIG. 3. Time history of drop mass of acetone–cyclohexane mixtures with various initial compositions evaporating in air at 14°C: (a) theory; (b) experiments x_{10} in volume fraction.

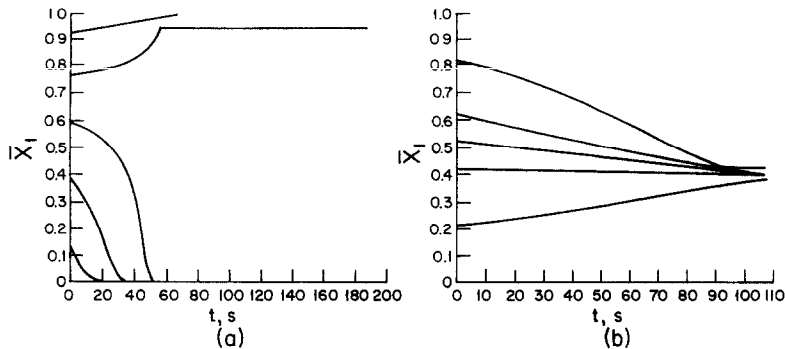


FIG. 4. Theoretical time history of liquid compositions in binary liquid drops evaporating in air: (a) positive azeotrope, acetone-cyclohexane mixtures at $T_\infty = 14^\circ\text{C}$; (b) negative azeotrope, acetone-chloroform mixtures at $T_\infty = 18^\circ\text{C}$.

oration rates varies with the initial composition. The fastest evaporation rate occurs at the initial acetone composition of 0.7 by volume, which is about the azeotropic composition. Figure 4 depicts that the drop composition varies drastically with time. Figure 4(a) is for the acetone-cyclohexane mixtures, a positive azeotrope, at 14°C . If the initial acetone composition \bar{x}_{10} is less than the azeotrope \bar{x}_a , of 0.72 then the acetone composition \bar{x}_1 diminishes toward zero with time as vaporization progresses. On the other hand, if $\bar{x}_{10} > \bar{x}_a$, then \bar{x}_1 increases toward unity with time. The phenomena can be explained using the P - x diagram in Fig. 1(a). For $\bar{x}_{10} < \bar{x}_a$, the acetone composition in the vapor phase \bar{y}_1 is greater than that in the liquid phase \bar{x}_1 at a given pressure. As vaporization proceeds, more liquid acetone evaporates, resulting in a continuous reduction in \bar{x}_1 . Conversely, for \bar{x}_{10} exceeding \bar{x}_a , \bar{y}_1 is lower than \bar{x}_1 and both increase as vaporization progresses. Figure 4(b) is the \bar{x}_1 - t diagram for the acetone-chloroform mixture, a negative azeotrope, at 18°C . It is observed that \bar{x}_1 congregates to the azeotropic composition \bar{x}_a , irrespective of \bar{x}_{10} being greater than or less than \bar{x}_a . This phenomenon is seen in Fig. 1(b): as vaporization proceeds, \bar{x}_1 moves along the dew line toward \bar{x}_a with any values of \bar{x}_{10} . For both the positive and negative azeotropes, a change in \bar{x}_1 causes a lowering of the equilibrium vapor pressure, resulting in a reduction of the vaporization rate. It is thus concluded that the composition change in a binary liquid drop during the vaporization process causes the suppression of vaporization.

Figures 5(a) and (b) plot the drop lifetime vs composition for acetone-cyclohexane and acetone-chloroform mixtures, respectively. In each figure, the solid and broken lines represent different values of Z_a which depends on the ambient conditions (T_∞ , P_∞) and the azeotropic properties (such as P_{ba}). It is observed that the larger Z_a is, the shorter is the drop lifetime, implying a stronger vaporization. Therefore, Z_a reflects the strength of vaporization. The most noteworthy is the coincidence of the extreme

(maximum or minimum) lifetime with the extreme pressure in Fig. 1, at the same azeotrope. A minimum amount of energy is required for vaporization at the azeotropic composition, in the case of a positive azeotrope which is characterized by the minimum boiling point (on the T vs x_1 or y_1 plot, not shown). Therefore, the drop lifetime is a minimum at the azeotrope but the combined thermal and solutal Marangoni effects on internal circulation within the drop reach a maximum. At the azeotrope where the equilibrium liquid and vapor compositions are identical, the vaporization speed should be a maximum because the process requires no mass transfer which is usually an order of magnitude slower than heat transfer. The situation is opposite in the case of a negative azeotrope. The heat for evaporation is a maximum at the azeotrope which is characterized by the maximum boiling point. Accordingly, the drop would take a maximum time to complete its evaporation process, while the combined thermal and solutal Marangoni effects on natural circulation inside the drop achieve a minimum.

Numerical computations reveal that drop shape has a strong influence on the vaporization process. For a sessile drop, its shape is characterized by the two

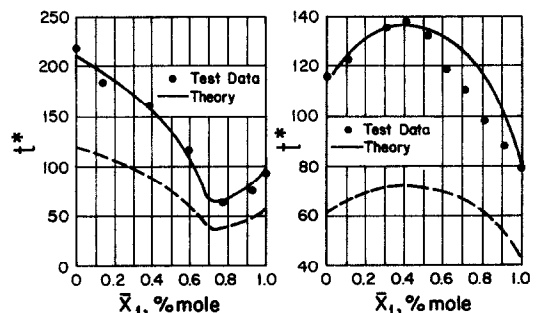


FIG. 5. Lifetime of binary liquid drops evaporating in air: (a) positive azeotrope, acetone-cyclohexane mixtures with $Z_a = 0.713 \times 10^{-3}$ (solid line) and 1.24×10^{-3} (broken line); (b) negative azeotrope, acetone-chloroform mixtures with $Z_a = 0.433 \times 10^{-3}$ (solid line) and 0.821×10^{-3} (broken line).

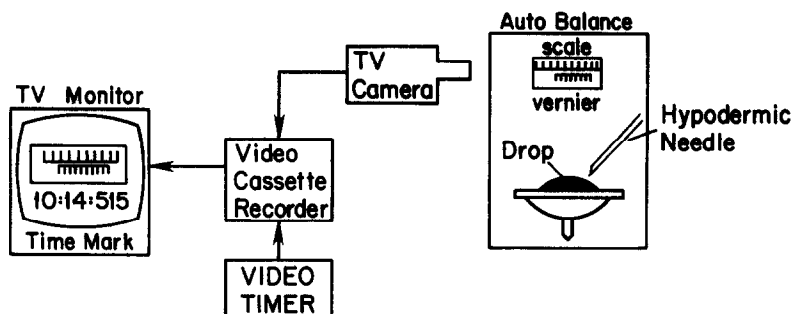


FIG. 6. A schematic of the experimental apparatus for drop evaporation.

parameters α and ψ . In the case of a binary liquid mixture, the parameters are determined by those of the constituent pure liquids under the same conditions by means of the mixing rule. The shape type is characterized by α , while ψ depends on both the initial size and the contact angle with the substrate. Both the drop shape and contact angle can be determined by a simple optical technique, called the reflection-interference method [9]. The values of α and ψ for three different types of drop vaporization, on a clean glass plate into the atmosphere, are found to be: acetone ($\alpha = 0.85$, $\psi = 1.7$), cyclohexane (0.65, 1.8) and chloroform (0.45, 3.5), by observation. The binary vapor of an azeotropic mixture, such as acetone-chloroform or acetone-cyclohexane, mingled in a non-condensable gas (such as air in the atmosphere) behaves as a non-ideal mixture. The non-ideal mixing factors for the two typical binary mixtures (one positive the other negative azeotrope) are obtained to be $\varepsilon_b = -0.25$, for acetone-cyclohexane (with $x_a = 0.72$) and $\varepsilon_b = 0.06$, for acetone-chloroform (with $x_a = 0.39$), by fitting the experimental data.

4. EXPERIMENTAL APPARATUS AND PROCEDURE

The experimental apparatus is schematically illustrated in Fig. 6. It consisted of a precision auto balance, a TV camera, a video cassette recorder, a video timer and a TV monitor. A glass plate was placed on the tray of the auto balance. The balance scale was automatically read by the TV camera and recorded in the video cassette, from the instant a liquid drop was placed on the plate until the completion of drop evaporation. An enlarged scale was installed on the auto balance for easier readings. The experimental procedure is as follows.

(1) The auto balance was balanced after the glass plate was placed on the receiving tray.

(2) A TV camera recorded the scale readings. Simultaneously, the video timer set a 1/100 s time scaling on the TV screen.

(3) A minute drop was carefully placed on the glass plate by means of a hypodermic needle and the video recorder began to monitor the change of the scale.

(4) Video recordings were played back on the TV monitor and the time mark for every 0.1 mg change in drop mass was read.

The step response of the auto balance was investigated to determine the recovery characteristics of the balance scale from a shock resulting from the placement of a liquid drop. In order to determine the mechanical instability at small times following a step disturbance, a water drop with very slow evaporation was used to monitor the variation of scale readings. The scale read a slightly higher value (an overshoot of 0.2 mg for a 9.4 mg water drop) over approximately a few seconds and then became stabilized (at a reading of 9.4 mg), see Fig. 7. Hence, the step-response time (for an overshoot to completely decay) is about 4–8 s for drops of 5–10 mg. The existence of the step-response time was demonstrated in all tests in the form of a rapid fall of drop mass, as seen in Fig. 3(b).

5. EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

Azeotropic mixtures of acetone (more volatile) and cyclohexane were tested by varying the composition. Figure 3(b) shows a typical result, in which \bar{x}_{10} represents the initial volumetric concentration of cyclohexane and m is the initial mass of a drop. It is

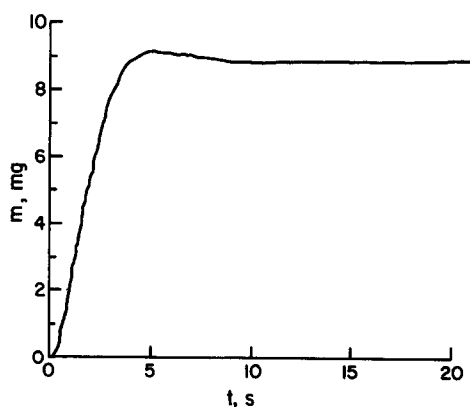


FIG. 7. Transient response of auto balance to loading of a water drop.

observed that in a certain range of small concentrations of binary mixtures with cyclohexane (less volatile component), shorter drop lifetimes occur than with pure acetone. In other words, the evaporation rate of acetone drops is enhanced in the presence of a small concentration of cyclohexane, for example the drops with $\bar{x}_1 = 0.5$ and 0.7 , evaporate faster than the pure acetone drop during the initial and intermediate stages of the evaporation process.

The solid circles in Fig. 5 depict the drop lifetime t^* vs the initial acetone composition \bar{x}_{10} . In Fig. 5(a) for acetone–cyclohexane mixtures, the minimal lifetime occurs near the composition of 70% acetone. In the case of acetone–chloroform mixtures in Fig. 5(b), the maximum drop lifetime occurs at the acetone concentration of about 50%, when \bar{x}_{10} exceeds 50%, the drop lifetime reduces almost linearly with \bar{x}_{10} .

Theoretical prediction for the time history of the drop size for acetone–cyclohexane mixtures at 14°C , Fig. 3(a), agrees well with the experimental results in Fig. 3(b). For the positive azeotrope in Fig. 5(a), theory agrees very well with experiments. In Fig. 5(b) for the negative azeotrope, theory and experiments are in good agreement for \bar{x}_{10} up to 50% beyond which theory overpredicts the drop lifetime. The disagreement was mostly due to changes in both the ambient temperatures and humidities between the days of the experiments. It is therefore concluded that the validity of the theoretical model is borne out by experiments.

It is important to note that the theory can also be applied to non-azeotropic binary mixtures although no result is presented here.

6. CONCLUSIONS

A theoretical model is developed to determine the size, history and lifetime of evaporating sessile drops of binary liquid mixtures. Results agree well with experiments. Important disclosures are given below.

(1) The maximum and minimum drop lifetimes exist at the azeotropic composition of the positive and negative azeotropes, respectively.

(2) A change in \bar{x}_1 causes a reduction in the equilibrium vapor pressure, thus suppressing the vaporization process.

(3) The parameter Z_a determines the strength of vaporization in a binary liquid drop. The larger Z_a , the higher the vaporization rate.

The limitation of the model is that the heat of drop evaporation must be supplied by the substrate with negligible contribution from gas-side heat transfer. Otherwise, unsteady heat transfer would affect drop evaporation. This is especially important in free spherical drops [10].

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THEORIE ET EXPERIENCES SUR DES GOUTTES SESSILES DE MELANGES BINAIRES LIQUIDES EN EVAPORATION

Résumé—On étudie théoriquement et expérimentalement l'histoire et la durée de vie de gouttes sessiles de mélanges binaires liquides qui s'évaporent. Les durées de vie minimales et maximales (respectivement aux taux de vaporisation respectivement les plus forts et les plus faibles) sont atteintes pour les compositions azéotropiques, respectivement les azéotropes positifs et négatifs. Les mécanismes qui conduisent à une durée de vie extrême sont dus aux changements de composition du composant le plus volatile pendant le mécanisme de vaporisation. Ce changement de composition tend à supprimer la vaporisation. Un nouveau paramètre sans dimension est trouvé pour déterminer l'intensité de vaporisation d'une goutte liquide binaire. Les prédictions théoriques de l'évolution de la taille de la goutte et de la durée de vie s'accorde avec les résultats expérimentaux.

THEORIE UND EXPERIMENTE ZUR VERDAMPFUNG VON HAFTENDEN TROPFEN BINÄRER FLÜSSIGKEITSGEMISCHE

Zusammenfassung—Größe, zeitliche Entwicklung und Lebensdauer haftender verdampfender Tropfen binärer Flüssigkeitgemische werden theoretisch und experimentell untersucht. Die minimale und die maximale Lebensdauer der Tropfen (gleichbedeutend mit maximaler bzw. minimaler Verdampfungsrate) tritt bei den azeotropen Zusammensetzungen der positiven bzw. negativen Azeotrope auf. Die Mechanismen, die zu einer extremen Lebensdauer der Tropfen führen, beruhen auf der Konzentrationsänderung der leichter flüchtigen Komponente während des Verdampfungsvorgangs. Ein neuer dimensionsloser Parameter zur Bestimmung der Verdampfungsintensität eines binären Flüssigkeitstropfens wird ermittelt. Theoretische Berechnungen der Größenentwicklung und der Lebensdauer der Tropfen stimmen befriedigend mit experimentellen Ergebnissen überein.

ТЕОРИЯ И ЭКСПЕРИМЕНТЫ ПО ИСПАРЕНИЮ НЕПОДВИЖНЫХ КАПЕЛЬ БИНАРНЫХ ЖИДКИХ СМЕСЕЙ

Аннотация—Теоретически и экспериментально определяются размеры, изменение во времени и продолжительность жизни испаряющихся неподвижных капель бинарных жидких смесей. Минимальное и максимальное снижения времени жизни (эквивалентные соответственно максимальной и минимальной скорости испарения) происходят в азеотропных смесях положительных и отрицательных компонентов соответственно. Механизмы, обуславливающие экстремальное снижение времени жизни, вызваны изменениями содержания наиболее летучего компонента в процессе испарения. Изменение состава приводит к подавлению испарения. Предложен новый безразмерный параметр для определения интенсивности испарения капли бинарной жидкости. Теоретические расчеты удовлетворительно согласуются с экспериментальными данными.