The Accommodation Coefficient and the Evaporation Coefficient of Water

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INTRODUCTION

Previous experiments* have already indicated that the maximum rate of evaporation of water into a vacuum is not so great as would be expected theoretically. The ratio of the experimental to the theoretical rate is defined as the evaporation coefficient \( f \) and has been found to have a value of about 0.04 for pure water at temperatures about 0°C. This result would indicate that, of the vapour molecules striking the liquid surface, about 96% must return to the vapour without entering the liquid. It is therefore of interest to enquire whether these vapour molecules attain temperature equilibrium with the surface or rebound at once before this equilibrium can be established. In the present paper experiments are described in which vapour molecules are incident on a liquid surface which is at a temperature lower than that of the vapour itself and the energy transferred to the surface by the vapour molecules is measured. If \( \alpha \), the accommodation coefficient, is defined as usual as the ratio of the energy actually transferred to the maximum possible transfer, it is found that for water at 10°C—

\[
\alpha = 1.0 \\
\frac{f}{0.036}
\]

so that, while only a very small fraction of the vapour molecules enter the liquid, all of them reach temperature equilibrium with the surface before re-evaporating into the vapour.

* Method

If a drop of water is allowed to form on a glass tip in a vessel maintained at a pressure \( p \) which is lower than the saturated vapour pressure corresponding to the temperature of the drop, steady evaporation takes place from the surface of the latter throughout the period of its formation.

* Alty, 'Phil. Mag.,' vol. 15, p. 82 (1933).
This evaporation cools the surface. When the drop is fully formed it falls from the tip and may be collected and the drop weight determined. The surface tension can be deduced therefrom and hence the surface temperature of the evaporating drop and the saturated vapour at this temperature may be obtained. This data makes possible the direct calculation of $f$ as follows.

If the temperature is sufficiently low and therefore the saturated vapour pressure not too great, the rate at which molecules leave the liquid will not be affected appreciably by the presence of the surrounding vapour. Further, the rate of transfer of molecules from a liquid to its saturated vapour must be equal to the rate from the vapour to the liquid so that

the number of molecules leaving the liquid at temperature $T$ and entering a vacuum per sec

$= \text{the number leaving the liquid and entering the saturated vapour at temperature } T \text{ per sec}$

$= \text{the number leaving the saturated vapour and entering the liquid}$

$= \text{the number striking the liquid surface } \times f$

$= \frac{1}{4}Ncf \text{ per sq cm, if } N \text{ is the number of vapour molecules per cc and } c \text{ is their average velocity.}$

Thus if $A$ is the area of the drop surface at time $t$, the total mass evaporating into a vacuum during the formation of the drop will be

$$m = \frac{1}{4}Ncf \int_0^t Adt \text{ gm}$$

$$0.2439 \frac{P_s}{\sqrt{T_s}} f \int_0^t Adt \text{ gm}$$

if the saturated vapour pressure $(P_s)$ at the temperature $(T_s)$ of the drop surface is expressed in millimetres of mercury; $\int_0^t Adt$ is given in square centimetres per second, and $t_0$ is the total time of formation of the drop in seconds.

When the evaporation takes place into a space at a pressure $p$ instead of into a vacuum, some molecules return to the liquid and the net amount of evaporation will be

$$\bar{m} = 0.2439 \frac{P_s - p}{\sqrt{T_s}} f \int_0^t Adt.$$  \hspace{1cm} (1)

The mass $\bar{m}$ may be equated to the mass actually evaporated from the drop. This may be determined experimentally as the difference between
the drop weight and the mass of water supplied to the drop during its formation. When the experimental values of the different quantities are inserted in equation (1), the value of the evaporation coefficient $f_1$ may be derived at once.

Suppose now that the water supplied to the drop enters it at a known temperature $T_b$. Then it appears that the drop may acquire heat in three ways, viz.:

(1) from the warm water entering the drop;
(2) by conduction of heat along the glass tip;
(3) from the gas surrounding the drop (since this gas will be at a temperature higher than that of the evaporating surface).

Let these three heat quantities be $W_1$, $W_2$, $W_3$ cals. Then the total amount of heat supplied to any drop must be equal to the total amount leaving the tip during the life time of the drop, i.e., to the sum of the latent heat of vaporization required by the evaporating liquid and the amount of heat carried away by the falling drop itself. Hence if $m$ gm evaporates from the drop and $M$ gm is the total mass of water entering it, we must have

$$W_1 + W_2 + W_3 = mL + (M - m) T_a$$

if $L_1$ is the latent heat of vaporization at $T^o$ C and $T_a$ C is the mean temperature of the falling drop.

But $W_1 = MT_b$, and $W_2$ may be calculated at once from the dimensions of the tip so that

$$W_3 = mL_{T_a} + (M - m) T_a - MT_b - W_2.$$

But $W_3 = $ heat supplied by the vapour striking the surface = mass of gas striking the surface $\times$ specific heat at constant pressure $\times$ $(T_g - T_s) \times \alpha$ if $T_g$ is the temperature of the vapour at a distance of one mean free path from the drop surface.

Therefore

$$W_3 = 0.2439 \frac{P}{\sqrt{T_g}} \int_{t_0}^{t} Adt \cdot C_p \cdot (T_g - T_s) \cdot \alpha,$$

or, equating the two values of $W_3$, we have

$$\alpha = \frac{mL_{T_a} + (M - m) T_a - MT_b - W_2}{0.2439 \frac{P}{\sqrt{T_g}} \int_{t_0}^{t} Adt \cdot C_p (T_g - T_s)}.$$

This is the expression used in the calculation of $\alpha$. 
EXPERIMENTAL

In the earlier work, water was used at temperatures about 0°C and at these temperatures the amount of evaporation from a drop is not very large (about 4 mg). It is clear that, if this evaporation could be increased, the accuracy of the experiment would also be increased considerably. This increase in evaporation is particularly necessary for the determination of \( \alpha \), as equation (3) reduces to the ratio of two rather small quantities if \( \bar{m} \) is small. If, however, \( \bar{m} \) is large, it is possible to make a more reliable estimate of \( \alpha \).

![Diagram of experimental setup](image)

Fig. 1

For these reasons the earlier apparatus was modified in several particulars and the essential features of the new apparatus are shown in fig. 1. Pure water is stored in the container C and passes through the fine capillary F to the tip T on which the drop is formed. The glass vessel G is connected at X to a high speed oil pump which keeps the pressure in G sufficiently low. The pressure in this vessel is measured by means of a travelling microscope and the manometer M. The general method of conducting an experiment was described in the earlier paper (loc. cit.) and will not be repeated.

The simplest method of increasing the amount of evaporation is to raise the temperature of the water entering the drop and thus supply...
more heat to the latter. For this purpose the capillary F was surrounded by an electrically controlled thermostat W, whose temperature could be varied up to 80° C. This increase in temperature introduced a new experimental difficulty. The drops were previously collected in small glass vessels containing a layer of light oil to prevent further evaporation after the drop entered the oil. When the warmer water was supplied to the drops, the temperature of the oil slowly increased during the course of an experiment until it was considerably above the temperature whose saturated vapour pressure was equal to the pressure in G. When this happened, the oil-water mixture boiled explosively and ruined the experiment. The glass vessels previously used were therefore replaced by the brass containers B. These fitted into brass cylinders H, soldered into a brass plate as shown in fig. 1, and good thermal contact was ensured by placing a little oil in H before introducing the containers B. The outer cylinders were immersed in a thermostat whose temperature was adjusted to be about 1° C higher than the temperature whose saturated vapour pressure was equal to the actual pressure in G. This temperature had to be fixed rather carefully. If it was too high, the oil exploded out of the container; if it was too low, water vapour condensed on to the cylinder and so made it impossible to determine the drop weight. On the other hand, once the correct temperature for any particular experiment was attained, the experiment could proceed without further trouble and with no sign of condensation. In order to avoid any errors from this source the brass containers were highly polished so that the first signs of condensation could be easily detected. With this arrangement it was possible to increase the number of drops collected in an experiment to about 50. In the earlier work no more than 10 could be collected safely; in most of the present experiments the number was 30 to 50.

By means of these modifications it was found that the amount of evaporation could be increased satisfactorily. Now, when a considerable mass of vapour leaves the drop per second, there must be rather violent convection currents in the container G and it was considered advisable to measure the pressure in G as near to the drop surface as possible. In the earlier work, the pressure was determined by a manometer attached to the stopper S but, with the larger rates of evaporation used in the present experiments, this type of measurement might lead to some error in \( p \). Consequently the manometer was connected to the tube D whose open end was within 1 mm of the drop surface. The actual pressure in G was varied from experiment to experiment by means of the mercury bubblers N (glycerine was used for the low pressure work).
The driving pressure in C was measured by means of a cathetometer reading to 0.001 cm, and was corrected for the slight change in pressure during an experiment caused by the removal of some water from C. Condensation of warm water on the glass of the apparatus was prevented by winding the whole with resistance wire and keeping it at a higher temperature than that of the water being used.

RESULTS

Preliminary Experiments—Before the determination of \( f \) and \( \alpha \) can be carried out a number of preliminary experiments must be completed.

A—Firstly the radius of the tip must be accurately known. This was measured directly and also deduced from the weight of very slowly formed drops at a known temperature at which the surface tension was already known. This method gives the effective tip radius and agreed well with that directly determined. The average of a number of measurements was

\[
r = 0.17485 \text{ cm}.
\]

From the weight of any given drop and the radius of the tip the surface tension can be determined and thence the surface temperature. For the particular tip used, the relation between drop weight and surface temperature could be represented by the equation

\[
T_s = -20.52 + 9.8064 m - 0.1677 m^2. \tag{4}
\]

B—The mass of liquid flowing through the capillary F at a temperature of 25.00° C was measured as a function of the driving pressure \( P \) (cm of Hg at 0° C) and could be represented by the equation

\[
M = (9.450 P + 16.725) \text{ mg/min}. \tag{5}
\]

The mass flowing at any other thermostat temperature may be derived from this by applying a correction for the change of viscosity.

C—Finally it is known that the weight of a falling drop only gives the true surface tension of a liquid when the drop is formed extremely slowly. When, as in these experiments, the drop is formed more rapidly, its mass is greater than normal and, before inserting the drop weight in equation (4), it must be corrected for this hydrodynamical effect. This is done by measuring the drop weight as a function of the rate of flow. If \( w \) is the drop weight when the rate of flow is \( M \) gm/min, and \( w_0 \) the weight for a very small rate of flow, then the percentage increase in weight is

\[
100 \frac{(w - w_0)}{w} = \Delta \text{ (say)},
\]

and this quantity is shown in fig. 2 as a function of \( M \). This
curve was obtained with water at 20.9°C and, of course, will be valid only at this temperature. In view of the fact that the thermostat temperature used in the experiments ranged from 20°C to 70°C it was no longer possible to ignore the temperature variation of Δ as was previously done (loc. cit.). Instead Δ was measured at a number of temperatures and the result is shown in fig. 3 in which the ratio (∆/20.9) is plotted against the temperature. From these two curves the measured drop weight may be corrected for the rate of flow before being inserted in equation (4) to give the surface temperature.

\[ \Delta_{20.9} \]

\[ \begin{array}{cc}
0.50 & \\
1.00 & \\
1.50 & \\
\end{array} \]

Rate of flow, gm/min.

D—The area of the evaporating surface is required. This was determined, as in the former work, by obtaining a number of photographs of the drop during its formation and deducing the area and \( \int_{0}^{t_{0}} Adt \) therefrom.

Calculation of \( f \) and \( \alpha \)—The calculation of \( f \) was performed in exactly the same manner as in the earlier work and the results obtained are included in Table I which also gives all the data required in the calculation. In spite of the fact that the drop itself was smaller than in the earlier experiments it will be seen that the amount of evaporation per drop was much greater than formerly, being as great as 40 mg in one experiment.
The pressure in the experimental vessel is very considerably lower than the saturated vapour pressure corresponding to the temperature of the surface as is shown in the column \( (P_s - p) \). This increase in \( \bar{m} \) should increase the accuracy of the determination of the experimental rate of evaporation while that in \( (P_s - p) \) should likewise increase the accuracy of the theoretical estimate.

For the calculation of \( \alpha \) equation (3) must be rearranged somewhat. We have

\[
\alpha = \frac{mL_{s} + (M - \bar{m}) T_a - MT_b - W_2}{0.2439 \frac{p}{\sqrt{T_g}} C_p (T_g - T_s) \int_{0}^{t} \text{Adt}},
\]

or

\[
\alpha (T_g - T_s) = \frac{[mL_{s} + (M - \bar{m}) T_a - MT_b - W_2] \sqrt{T_g}}{0.2439 \frac{p}{\sqrt{T_g}} C_p \int_{0}^{t} \text{Adt}},
\]

and

\[
\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right) = \frac{[mL_{s} + (M - \bar{m}) T_a - MT_b - W_2] \sqrt{T_g}}{0.2439 \frac{p}{\sqrt{T_g}} C_p (T_b - T_s) \int_{0}^{t} \text{Adt}},
\]

and \( C_p = 0.465 \) for water vapour.

Now \( T_g \), the temperature of the gas striking the drop surface, is unknown. It must, however, be between \( T_s \), the lowest temperature in the system, and \( T_b \), the highest temperature in the system, and since in the experiments \( (T_b - T_g) \) is never very large, we may replace \( \sqrt{T_g} \) on
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<th>Expt No</th>
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<th>(P_s - p)</th>
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<th>T_b °C</th>
<th>T_s °C</th>
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<th>t_o secs</th>
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the right-hand side by $\sqrt{T_s}$. The error will then not be large and the resulting value of $\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)$ will be too small.

Similarly, the exact value of $T_a$, the average temperature of the falling drop, is not known. If we replace $T_a$ by $T_s$, i.e., if we assume that all the mass of the falling drop is at the temperature of its surface, we shall once more obtain a value of $\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)$ which is too small.

With these simplifications equation (6) may be replaced by the inequality

$$\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right) > \frac{8 \cdot 816 \sqrt{T_s}}{p (T_b - T_s) \int_0^{t_0} \text{Adt}} [m (L_{T_s} - T_s) - M (T_b - T_s) - W_2].$$

Now the conduction term $W_2$ will depend on the temperature difference between the tip and the thermostat. It would have a maximum value if the tip were at the temperature $T_s$, when the actual value of $W_2$ for the tip used in the experiments would be

$$W_2 = \frac{4 \cdot 26 \times 10^{-5} (T_b - T_s) t_0}{d},$$

where

- $A$ is the area of cross-section of the glass capillary in sq cm,
- $K$ is the specific thermal conductivity of the glass, and
- $d$ is the length in cm of the glass capillary projecting from the thermostat.

Inserting the values of the constants we find

$$W_2 = 4 \cdot 26 \times 10^{-5} (T_b - T_s) t_0 \text{ cals}$$

if $t_0$ is the time of formation of the drop in seconds.

In just the same way as for the previous approximations, we shall obtain a minimum value for $\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)$ by giving $W_2$ its maximum value. Hence the value of $\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)$ may be written

$$\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right) > \frac{8 \cdot 816 \sqrt{T_s}}{p (T_b - T_s) \int_0^{t_0} \text{Adt}} [m (L_{T_s} - T_s) - (T_b - T_s) (M + 4 \cdot 26 \times 10^{-5} t_0)]$$

where

$$[\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)]_{\text{min}}$$

In this expression every term on the right-hand side is measurable and so the minimum value of $\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)$ may be determined. This value may be written $[\alpha \left( \frac{T_g - T_s}{T_b - T_s} \right)]_{\text{min}}$. 
The value of \( T_g \), the temperature of the gas striking the drop surface, will vary from one experiment to another. When the rate of evaporation \( (\dot{m}/t) \) is small, or the pressure \( (p) \) in the experimental vessel is high, \( T_g \) will not differ very markedly from \( T_s \). If, however, \( \dot{m}/t \) becomes large, convection currents will become more and more pronounced in the vessel and warm vapour may be swept towards the drop surface. Similarly, if \( p \) becomes small the mean free path in the gas increases and it becomes less and less difficult for the warmer gas to approach the surface. For these reasons it may be expected that \( T_g \) and therefore \[ \alpha \left( \frac{T_g - T_s}{T_b - T_s} \right) \] will increase with the quantity \( (\dot{m}/tp) \), and the two quantities are plotted against each other in fig. 4. The curve clearly shows the well-marked relation between them.

Now it is not possible under any circumstances for \( T_g \) to exceed \( T_b \), since \( T_b \) is the temperature of the brass stopper \( S \) and so is the highest temperature with which the gas in \( G \) is in contact. Hence at all times

\[ T_g \leq T_b \]

and therefore, since \( \alpha \) can never exceed unity,

\[ \alpha \left( \frac{T_g - T_s}{T_b - T_s} \right) \leq 1. \]
But from fig. 4 it appears that for large values of $(\bar{m}/tp)$,

$$\left[ \alpha \left( \frac{T_g - T_b}{T_b - T_s} \right) \right]_{\text{min}} = 1,$$

so that in such experiments it must follow that

$$T_v = T_b$$

and

$$\alpha = 1.$$

Therefore in experiments with a rapid rate of evaporation into a vessel at low pressure, it seems that the temperature of the gas close to the drop is equal to that of the hottest part of the system and that the heat exchange between the gas and the surface is exactly what would be expected from a theoretical point of view if every impinging vapour molecule reaches thermal equilibrium before leaving the surface and returning to the vapour.

Since the accommodation coefficient of water vapour at a liquid water surface is thus found to be unity, fig. 4 shows the variation of $[(T_g - T_s)/(T_b - T_s)]_{\text{min}}$ with $(\bar{m}/tp)$. It appears that for small values of $(\bar{m}/tp)$, the gas temperature $T_g$ does not differ greatly from $T_s$, and only when $(\bar{m}/pt)$ is greater than about $15 \times 10^{-5}$ does $T_g$ become equal to $T_b$. When the drop is surrounded by stagnant vapour, as in experiments at low $(\bar{m}/tp)$, this stagnant layer may affect the rate of evaporation and hence the value of $f$, and, in fact, it may be seen from Table I, or fig. 5 that this is so, $f$ being quite definitely reduced in the experiments for which $(\bar{m}/tp)$ is small. No effect of this nature could, however, be present in the experiments in which $T_g = T_b$, as there is clearly no difficulty in vapour molecules from the body of the gas reaching or leaving the surface. Consequently the best value of $f$ is probably obtained by taking the average of all experiments for values of $(\bar{m}/tp)$ greater than $15 \times 10^{-5}$. In this way the value

$$f = 0.034$$

is obtained or, if the exceptionally low value 2.54 should be rejected, $f = 0.036$.

**DISCUSSION**

The results as a whole show that the rate of evaporation of water is only about 0.036 of that deduced from kinetic theory considerations. Therefore more than 96% of the vapour molecules striking the surface return to the vapour without entering the liquid. They do, however,
remain on the surface long enough to reach temperature equilibrium with it. This behaviour is very unexpected for a liquid, as any molecule condensing on the surface would be expected automatically to become part of the liquid. The result suggests that the surface behaves rather like a semi-solid on which adsorption and re-evaporation of the vapour molecules may take place, the chance of an adsorbed molecule entering the liquid during its lifetime on the surface being small. The idea of the liquid surface obtained in this way would appear to fit in well with the theory of the structure of water recently proposed by Fowler and Bernal.

In conclusion, we wish to thank Mr. H. H. Penley and Mr. J. H. Buck who obtained for us the data from which fig. 3 was drawn.

**Summary**

The evaporation coefficient of water and the accommodation coefficient for water at a liquid water surface have been measured and it has been shown that

\[
\text{the evaporation coefficient} = 0.036, \\
\text{while the accommodation coefficient} = 1.0.
\]

These results indicate that, so far as the interaction with the vapour molecules is concerned, the surface behaves more like a solid than a liquid. Most of the vapour molecules incident on it are unable to penetrate into the liquid, but, on the other hand, they are able to attain temperature equilibrium with the surface before re-evaporating into the vapour phase.

* * J. Chem. Phys.,* vol. 1, No. 8, p. 515 (1933).