

AN EVAPORATION EXPERIMENT AND ITS IRREVERSIBLE THERMODYNAMICS*

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Abstract—This paper considers a particular heat and water vapor transfer problem in a nonisothermal steady state system. The results are applicable to a condition that may develop during the normal drying of porous materials. The rate equations are based on the theory of irreversible thermodynamics and the validity of Onsager's reciprocity relation is demonstrated with a macroscopic analysis of this particular model. The results of an experiment are presented which show the effects of temperature, pressure, and salt concentration on the evaporation and heat transfer from a porous plate. These data combined with the thermodynamic flux equations provide a means of calculating the effective concentration of dissolved salts at the air-water interface and the approximate depth of this increased concentration. The analysis also provides a method for determining the rate-limiting process in the experiment.

NOMENCLATURE

- C , concentration of KCl [g mole/l of solution];
 C_p , heat capacity [cal/g degK];
 D , molecular diffusion coefficient [cm^2/s];
 h , absolute enthalpy of water [cal/g mole];
 H , heat of vaporization of water [cal/g mole];
 J , flux [g moles/s cm^2] and [cal/s cm^2];
 l , effective length between the source and the sink [cm];
 L , phenomenological coefficient, units depend on the associated subscript;
 p , vapor pressure of water [cal/ cm^3];
 P , total pressure [cm Hg];
 R , gas constant [cal/g mole degK];
 T , absolute temperature [degK];
 z , depth of salt concentration at the interface [cm];
 Σk , sum of coefficients accounting for molecular heat conduction and thermal radiation, [cal/s cm^2];
 Δ , difference between the source and the sink;

- γ , thermal molecular diffusion coefficient [mole/s cm^2 degK];
 μ , chemical potential [cal/g mole].

Subscripts

- a , water vapor in air;
 e , energy;
 q , heat;
 0 , reference state, pure substance under atmospheric pressure at a temperature T ;
 s , salt in water;
 w , water.

INTRODUCTION

WHENEVER there is a significant rate of evaporation of water from a porous body the process is necessarily nonisothermal. Therefore it is never a simple system involving only a single driving force and flux. For this reason the thermodynamics of irreversible processes appears to offer a useful and systematic structure for studying the general problem [1, 2]. Of particular interest in this analysis is the Onsager reciprocity relation, which is presently based on statistical mechanical arguments. Its scope of validity is being investigated in a wide range of experimental situations. In a review article Miller [3] has compiled numerous experimental data supporting the

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reciprocity relation in isothermal systems. Recent papers dealing with more complex diffusion problems have offered further support [4, 5]. The experimental demonstration of the relation in nonisothermal systems is much less evident, however [3, 6].

In this communication a relatively simple system is taken for study and the driving forces chosen such that the validity of Onsager's relation appears from a simplified analytical argument. This model is of practical interest in many normal drying processes of porous bodies.

THEORY

Consider the following model. Evaporation occurs from a source consisting of a saline water-air interface with a given temperature and vapor pressure which will be referred to as the source. The moisture then moves by molecular diffusion along steady state vapor concentration and thermal gradients through still air or air-filled dry voids. After traveling an effective distance l , the vapor arrives at a sink with a second particular vapor pressure and temperature. The sink may be formed, for instance, by recondensation at a second interface or by a mass flow air current.

Such a system may be treated as discontinuous, i.e. the distance l is analogous to the thickness of a membrane permeable to water vapor and heat separating two phases, the sink and the source. From the theory of nonequilibrium thermodynamics [7] the equations for the water and energy transfer may be written for the steady state as

$$J_w = -L_w \Delta \frac{\mu_w}{T} + L_{we} \Delta \frac{l}{T} \quad (1)$$

and

$$J_e = -L_{ew} \Delta \frac{\mu_w}{T} + L_e \Delta \frac{l}{T} \quad (2)$$

provided the gradients of temperature and vapor pressure are not extreme and the air is stable between the source and the sink.

Onsager's reciprocity relation states that

$$L_{ew} = L_{we}. \quad (3)$$

The validity of this relation may be shown in the following manner:

Since $\mu - \mu_0 = RT \ln p/p_0$, the driving force on the water between the source and the sink may be expressed as

$$\Delta \frac{\mu_w}{T} = R \Delta \ln p/p_0 + \Delta \frac{\mu_{w0}}{T}. \quad (4)$$

Restricting the temperature range to $273.16 < T < 373.16$ degK and taking the heat capacity of liquid water as constant so that $h = C_p (T - T_0) + h_0$, the Gibbs-Helmholtz equation [8] may be integrated to give

$$\Delta \frac{\mu_{w0}}{T} = -C_p \Delta \ln T + (h_0 - C_p T_0) \Delta \frac{l}{T}. \quad (5)$$

Then equation (4) may be written as

$$\Delta \frac{\mu_w}{T} = R \Delta \ln p/p_0 - C_p \Delta \ln T + h_0 \Delta \frac{l}{T} - C_p T_0 \Delta \frac{l}{T}. \quad (6)$$

Using this expansion and a transform [6] to eliminate h_0 , equations (1) and (2) become

$$J_w = -L_w \left(R \Delta \ln p/p_0 - C_p \Delta \ln T - C_p T \Delta \frac{l}{T} \right) + L_{wq} \Delta \frac{l}{T} \quad (7)$$

and

$$J_q = -L_{qw} \left(R \Delta \ln p/p_0 - C_p \Delta \ln T - C_p T \Delta \frac{l}{T} \right) + L_q \Delta \frac{l}{T}. \quad (8)$$

As already shown for a system of this type [6]

$$L_{qw} = HL_w = H \frac{pD_a}{R^2 l T}. \quad (9)$$

When the temperature is around 300°K and the differences are not more than a few degrees, the factor $[\Delta \ln T + T \Delta(l/T)]$ may be neglected. Equation (7) may then be written as

$$J_w = -\frac{pD_a}{RT} \Delta \ln p/p_0 + L_{wq} \Delta \frac{l}{T}. \quad (10)$$

An alternative form given by the kinetic theory of gas [9] is

$$J_w = -\frac{D_a}{RT} \frac{\partial p}{\partial x} + \gamma \frac{\partial \ln T}{\partial x}. \quad (11)$$

Under the relatively small thermal differences encountered in this model the factor γ ($\partial \ln T / \partial x$) will make a negligible contribution to the net flux [10]. Equation (11) then may be integrated between the limits of vapor pressures at the source and the sink to give the flux as equal to $D_a \Delta p / RTl$, using the average values of D_a and T as constants. Since $p \Delta \ln p$ is well approximated by Δp , this result may be used to reduce equation (10) to*

$$L_{wq} \Delta \frac{l}{T} = -\frac{p D_a}{RTl} \Delta \ln p_0. \quad (12)$$

Replacing $\Delta \ln p_0$ with its equivalent in a temperature function as given by the Clausius-Clapeyron equation [8], and solving for L_{wq} , shows that

$$L_{wq} = L_{qw} \quad (13)$$

upon comparison with equation (9).

Equation (13) has been experimentally demonstrated in a system similar to the model proposed here [6]. This experimental verification may be cited as supporting the validity of the preceding assumptions and simplifications used in arriving at the expression analytically.

Combining (1) and (6) to eliminate $\Delta \mu_w / T$, and comparing the result with (7) shows that

$$L_{wq} = L_{we} - hL_w \quad (14)$$

and because of the identity $J_e = J_q + hJ_w$, equation (2) may be written as

$$J_q = -L_{ew} \Delta \frac{\mu_w}{T} + L_e \Delta \frac{l}{T} - hJ_w. \quad (15)$$

Using (6) to eliminate $\Delta \mu_w / T$ and (7) to eliminate J_w from this expression shows that

$$L_{qw} = L_{ew} - hL_w \quad (16)$$

after comparison with (8). Onsager's relation, equation (3), then follows directly from (13), (14), and (16).

* The author was unable to locate any published values of γ for the system of water vapor and air, so it is not known whether or not the small errors induced by dropping $\gamma/l \Delta \ln T$ and $-L_w C_p (\Delta \ln T + T \Delta l/T)$ are compensating.

Several points are worth noting here. For instance equation (14) shows that the transfer of energy as residual enthalpy of the mass does contribute to the interaction term for water flow in (1) and so a method used previously by this author [11] to test Onsager's relation is not rigorous.

Use of equations (9), (13) and (10) gives a useful result for investigating the mass flow in the model under consideration,

$$J_w = -\frac{p D_a P_0}{RTlP} \Delta \ln p/p_0 - \frac{p D_a H P_0}{lR^2 T^3 P} \Delta T, \quad (17)$$

where the total pressure ratio is added to account for conditions other than those at 76 cm Hg. If the flow is through a porous structure, a factor must also be included to account for its geometry. A simple relation for the flux of heat also follows from these results,

$$J_q = HJ_w + \Sigma k \Delta T. \quad (18)$$

The reader is cautioned that these equations are not complete for transfer across a section of porous material, damp enough for liquid phase surface flow to develop. In such a case more detailed analytical procedures would be required [12, 13, 14].

EXPERIMENT

An experiment was carried out in which the steady-state evaporation rate of water into air was observed as a function of air pressure, temperature and the concentration of KCl in the evaporating solution. The apparatus employed has been previously described in detail [6]. Briefly, it consisted of a vapor gap 1.5 cm across and 19.8 cm in diameter bounded by 2 porous ceramic plates and a lucite ring. The plates were saturated with water or saline solutions and thermally controlled so that a temperature gradient could be developed across the air space between them. They were oriented so that convection currents were negligible in the air gap under steady state conditions. The water would then evaporate from the warm plate, diffuse across the air space and condense on the cooler plate. This flux was measured as were the temperatures of the air-water interfaces.

RESULTS

A portion of the water flux observations are listed in Tables 1 and 2. While it was possible to make direct observations of the heat flux across the air gap in this apparatus [6], more accurate results were obtained by using the observed vapor flux in equation (18) and taking Σk as 0.89×10^{-4} cal/s cm² degK. These results are also shown in Tables 1 and 2.

Random variations in the water flux measurements were generally less than $\pm 0.15 \times 10^{-8}$ g moles/s cm² except at the high thermal gradients around 40°C with the 1.0 M KCl solution. In this case the system seemed to be less stable. Errors in the heat flux are proportional to those of the water flux due to their method of measurement. The fourth significant figure in the reported temperatures must be considered an estimate. The random variations in the steady-state temperatures were never greater than 0.05°C. At the higher thermal gradients, however, the temperatures recorded could be different from the true air-water interface temperature by 0.1°C. The energy input to the reservoir supplying warm water was varied from approximately 1 to 4 W to achieve the thermal gradients reported. About 1/3 of this energy was transported across the air space.

Comparison of the water flux data and equation (17) is of particular interest. Equation (17) shows that the rate of transfer across the air gap may be shown as the sum of two terms. The

first term on the right is significant only when the air at the interfaces is not saturated with water vapor. This could occur if: air currents were to sweep the vapor away faster than it could be supplied across the interface by the solution; or the vapor were to diffuse away into the air faster than the solution could supply it; or some impurity in the air-water interface were to restrict the vaporization of water or reduces its equilibrium relative humidity. The second term on the right accounts for the change in vapor pressures at the interfaces due to the thermal difference. This term should be sufficient to predict the water flux across the air space for a pure system provided the rate of evaporation is not limiting; i.e. the interface can maintain an 100 per cent relative humidity.

The flux predicted by this term,*

$$J_w = \frac{p D_a H P_0}{R^2 T^3 P} \Delta T \quad (19)$$

are presented in the last column of Table 1. The small difference between these values and those experimentally observed may be attributed to uncertainties of the true temperature of the evaporating and condensing sites. These results

* Values of D_a were taken from Dorsey [15] and mean values of T , p , and H were used in this calculation. Values of H were taken as those of pure water since the equilibrium humidity value for given amount of KCl in water is independent of temperature [16].

Table 1. Water and heat flux when no salt is present

Temp. of cool interface (degC)	Temp. of warm interface (degC)	Grad. temp. (degC/cm)	Total air pressure (cm Hg)	Heat flux (Cal/s cm ²)	Observed water flux (g mole/s cm ²)	Calculated water flux (g mole/s cm ²)
14.35	18.15	2.53	64	7.1×10^{-4}	3.4×10^{-8}	3.57×10^{-8}
26.40	28.20	1.20	64	4.9	3.2	3.16
26.81	28.44	1.09	64	4.7	3.15	2.90
28.50	31.28	1.85	64	8.0	5.3	5.55
36.43	37.73	0.87	64	5.1	3.8	3.75
38.53	41.13	1.73	64	11.2	8.6	8.54
15.20	20.20	3.33	44	12.0	7.2	7.38
30.13	32.81	1.77	44	11.3	8.4	8.55
39.28	41.18	1.27	44	11.3	9.25	9.30
27.60	29.88	1.52	85	5.6	3.4	3.26
13.31	18.25	3.29	114	7.0	2.5	2.57
27.27	29.75	1.63	114	4.9	2.6	2.56

Table 2. Water and heat flux with KCl concentration as a variable

Temp. of cool interface (degC)	Temp. of warm interface (degC)	Grad. temp. (degC/cm)	Heat flux (Cal/s cm ²)	Water flux (g mole/s cm ²)	Reduction in water flux due to the KCl solution (%)
1.0 M KCl Solution					
12.33	13.95	1.08	1.7×10^{-4}	0.2×10^{-8}	84
13.66	16.84	2.12	4.2	1.3	54
15.13	19.62	2.99	6.4	2.25	48
26.0	27.20	0.80	1.1	0.05	98
27.83	30.72	1.93	5.2	2.5	55
28.82	32.35	2.35	7.4	4.1	45
35.80	36.95	0.77	1.6	0.5	84
36.38	38.26	1.25	4.2	2.4	56
37.83	40.68	1.90	7.3	4.6	50
39.51	42.68	2.11	8.5	5.5	53
0.7 M KCl Solution					
11.35	13.47	1.41	2.8×10^{-4}	0.9×10^{-8}	44
12.28	15.35	2.05	4.4	1.6	35
26.86	28.90	1.36	3.5	1.65	55
26.96	29.18	1.48	3.5	1.43	65
28.20	31.10	1.93	6.1	3.4	41
28.20	31.20	2.00	6.2	3.4	42
37.83	40.10	1.33	4.8	2.95	54
38.10	40.12	1.35	4.6	2.7	58
39.45	42.47	2.01	8.89	6.0	43
0.4 M KCl Solution					
13.64	17.13	2.33	5.6×10^{-8}	2.3×10^{-8}	26
14.18	17.88	2.47	6.2	2.75	19
26.30	27.59	0.86	2.0	0.8	63
28.08	30.40	1.55	5.7	3.5	22
28.73	31.81	2.05	7.8	4.8	25
36.39	37.59	0.80	2.6	1.5	56
38.18	40.20	1.35	6.8	4.8	26
39.60	42.11	1.67	9.1	6.7	22

suggest that the air-water interfaces are able to maintain essentially equilibrium humidity values, so the rate-limiting process is the diffusion of water vapor through air. This conclusion has also been suggested by Wiegand* based on the evaporation of free water. At higher average temperature, or for greater thermal gradients, or under lesser air pressures this may or may not be the case.

* Dr. C. L. Wiegand, Soil and Water Conservation Research Division, Agriculture Research Service, USDA, Weslaco, Texas; a private communication.

Equation (19) does not predict the vapor flux when KCl is present in the evaporating solution. However, it is useful to demonstrate the degree of reduced transfer caused by the salt as shown in the last column of Table 2. The percentage decrease in evaporation increases as the thermal gradient drops and as the salt concentration rises. The net heat flux is also reduced by the KCl solutions as demonstrated by comparing the data in Tables 1 and 2.

Even though equilibrium values of p/p_0 are known for various concentrations of KCl

solutions [16] equation (17) cannot yet be used to predict J_w because the KCl concentration at steady state will be higher at the interface than in the bulk solution. This is due in part to the "Gibb's surface excess" phenomenon [8] and in part to the "Soret effect" [9], but it is primarily due to the concentration of salt in the interface as the water evaporates. At this time the best use of equation (17) is made by measuring the net flux of vapor when the cooler plate contains pure water and calculating the value of p/p_0 for the interface containing the salt. These results may be related to an effective concentration [16] of KCl as shown in Fig. 1. Here experimental errors are in the neighborhood of 0.5 mole/l. The concentration of salt at the interface initially rises with the rate of water flux and decreases with a rising average temperature. The temperature dependence of the concentrations is obviously related to the temperature coefficient for the diffusion of KCl in water [16].

Upon the completion of more accurate experimental measurements it will be interesting to analyse the simultaneous movements of heat, water, and salt through the porous plate from the bulk solution to the drying interface with the

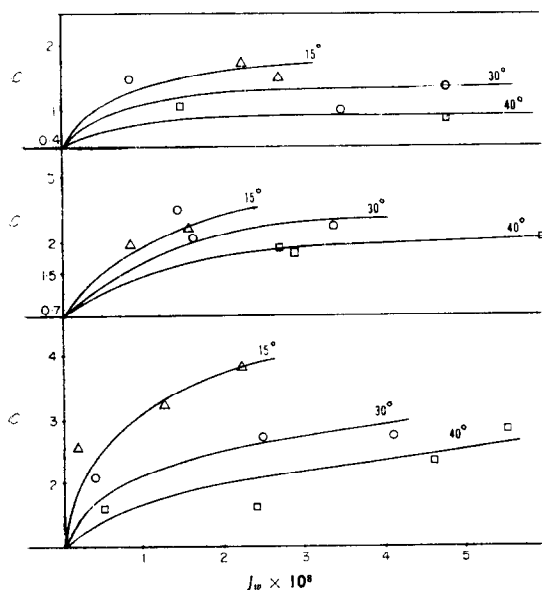


FIG. 1. The apparent concentration (g mole/l) of KCl at the evaporating interface as a function of the rate of evaporation. The curve parameters represent the average temperature in the vapor gap.

methods of nonequilibrium thermodynamics. At the present time it is perhaps worthwhile to estimate the depth of the salt concentration region by neglecting the thermal gradient and supposing that at a steady state the concentration gradient is linear. Then

$$1.8 J_w C = D_s \frac{\Delta C}{z} \times 10^{-3} \quad (20)$$

where D_s may be taken as 1.86×10^5 cm²/s [16]. The data in Fig. 1 then suggest that z was approximately 0.5 cm at 30°C and $J_w = 2.5 \times 10^{-8}$.

Several observations of interest can be made concerning the transfer of heat across the air space. This flux was not an independent function of thermal gradient, but was significantly affected by the pressure, average temperature, and KCl concentration of the evaporating solution. A certain percentage of the net heat which moved across the air-water interface was involved in the evaporation of water as may be seen from the two terms on the right of equation (18). This ratio (the heat used in evaporation per net heat transfer) was not a constant. As shown by the experimental curves in Fig. 2, the drying efficiency of the heat flux was a function of both the average temperature and the concentration of KCl in the evaporating solution. The curves affected by the KCl solution have positive slopes,

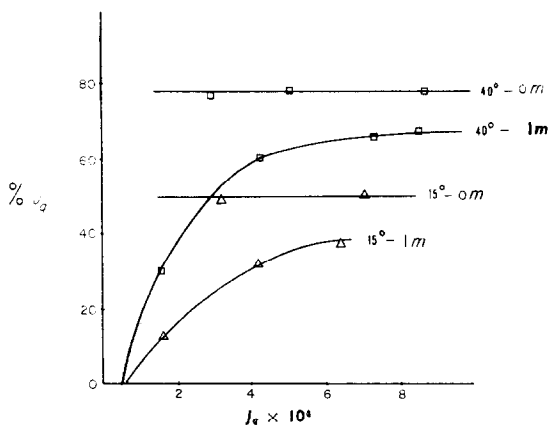


FIG. 2. The percent of heat flux crossing the interface which is used in evaporation as a function of the net heat flux. The curve parameters are the average temperature of the vapor gap, °C, and the concentration of KCl, g mole/l, in the bulk solution.

since the salt and thermal differences were developing opposing forces on the water vapor diffusion. This ratio would also decrease as the pressure rises and the thermal radiation properties of the air-water interface approach those of an ideal black body.

CONCLUSIONS

For a particular model with nonisothermal diffusion, the Onsager reciprocity relation between energy and water vapor phenomenological coefficients may be demonstrated by using a particular choice of driving forces and a macroscopic analysis. This analysis leads to useful equations in studying the evaporation of water and saline solutions from a porous surface. The study shows that under the conditions of this experiment:

- (a) The rate of vapor diffusion into air was the rate-limiting process in drying, rather than the ability of the air-water interface to supply its equilibrium value of vapor pressure.
- (b) The effective concentration of KCl at the evaporating interface approached four times its concentration in the bulk solution at 15°C.
- (c) The effective depth of the increased concentration of KCl was in the neighborhood of 0.5 cm.
- (d) The efficiency of heat consumed in evaporation was not directly proportional to the heat flux across the evaporating interface, but decreased significantly as the average temperature decreased and as the concentration of KCl in the bulk solution increased.

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Résumé—Cet article considère un problème particulier de transport de chaleur et de vapeur d'eau dans un système non-isotherme en régime permanent. Les résultats sont applicables à un état qui peut se développer pendant le séchage normal de matériaux poreux. Les équations de vitesse sont basées sur la théorie de la thermodynamique irréversible et la validité de la relation de réciprocity d'Onsager est démontrée à l'aide d'une analyse macroscopique de ce modèle particulier. On a présenté les résultats d'une expérience qui montrent les effets de la température, de la pression et de la concentration en sels sur l'évaporation et le transport de chaleur à partir d'une plaque poreuse. Ces données combinées avec les équations thermodynamiques de flux fournissent un moyen de calculer la concentration effective en

sels dissous à l'interface air-eau et la profondeur approchée de cette augmentation de concentration. L'analyse fournit aussi une méthode de détermination du processus de limitation de vitesse dans l'expérience.

Zusammenfassung—In der Arbeit werden besondere Probleme des Wärme- und Wasserdampftransports in einem nichtisothermen, stationären System betrachtet. Die Ergebnisse lassen sich auf Verhältnisse anwenden, die während der normalen Trocknung poröser Materialien auftreten. Die Gleichungen beruhen auf der Theorie der irreversiblen Thermodynamik; die Gültigkeit von Onsagers Reziprozitätsbeziehung wird durch eine makroskopische Analyse dieses speziellen Problems nachgewiesen. Die Versuchsergebnisse zeigen den Einfluss der Temperatur, des Druckes und der Salzkonzentration auf die Verdampfung und den Wärmeübergang von einer porösen Platte. Diese Daten, zusammen mit den Gleichungen für den thermodynamischen Fluss ermöglichen die Berechnung der wirksamen Konzentration des nahe der Luft-Wasser Trennfläche gelösten Salzes und die ungefähre Tiefe dieser erhöhten Konzentration. Die Analyse liefert auch eine Methode zur Bestimmung des geschwindigkeitbegrenzenden Prozesses im Versuch.

Аннотация—В статье рассматривается частная задача переноса тепла и водяного пара в неизотермической стационарной системе. Результаты эксперимента можно применить к условиям обычной сушки пористых материалов. Уравнения скорости переноса выводятся на основе теории термодинамики необратимых процессов, а справедливость соотношений взаимности Онзагера подтверждается макроскопическим анализом этой частной модели. Приведены результаты эксперимента, которые показывают влияние температуры, давления и концентрации соли на испарение и теплообмен пористой пластины. Эти данные вместе с уравнениями термодинамического потока представляют основу для расчета эффективной концентрации растворившихся солей на поверхности раздела фаз «воздух-вода» и приблизительную глубину этой увеличившейся концентрации. Этот анализ также включает метод определения интенсивности процесса переноса в эксперименте.