Free Convection Mass Transfer during Vaporization of Some Pure Liquids into Air

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When mass is exchanged between a solid or a liquid surface and a moving fluid that carries away the transferred material from the surface, the transfer is said to take place by convection. If the fluid motion that accompanies convection is caused by gravity acting on portions of the fluid that have different densities, the process is called free or natural convection. Differences in densities may arise due to differences in temperature and/or concentration.

Calculations of heat and mass transfer by convection are complicated by the large number of variables involved, but the application of dimensional analysis to heat transfer by convection results in the following significant dimensionless groups influencing the process:

$$\mathrm{Nu} = \phi\,(\mathrm{Re}, \mathrm{Pr}, \mathrm{Gr}) \qquad (1)$$

In the case of free convection heat transfer, a further simplification is possible because, the motion being entirely due to density gradients, the convective velocity may be taken as equal to zero and consequently the Reynolds number drops out in the general correlation to provide:

$$\mathrm{Nu} = \phi\,(\mathrm{Gr}, \mathrm{Pr}) \qquad (2)$$

In gases, the Prandtl number varies very little and the only significant group which influences the free convection process is the Grashof modulus. In the case of streamline flow, where the fluid currents are slow enough for the inertia stresses to be negligible compared to viscous stresses, only the product (Gr × Pr) need be considered. When the convection currents become fully turbulent, the power in the product takes on a higher value. It has been found that up to a value of 10^{+8} for the product (Gr × Pr), the process is in the streamline range and above this value turbulent effects are observed in the boundary layer.

The exact nature of the correlation is best determined by a graphic plot of the dimensionless groups evaluated experimentally.

Considerable work has been done on free convection heat transfer in liquids and gases. Saunders (*11*) and Touloukian (*12*) have exhaustively dealt with vertical and horizontal surfaces, plane and cylindrical. Saunders has tabulated the numerical constants to be used in equations for calculating free convection heat transfer under streamline and turbulent conditions. Kranssold (*8*) has investigated the heat transfer between concentric horizontal and vertical cylinders. Kyte, Nadden, and Piret (*9*) have investigated the natural convection heat transfer under reduced pressure. In the case where the convection currents are going down, the geometry of the cell and of the balance may well affect the results. A quantitative estimation of this effect has not been attempted until now.

The processes of heat and mass transfer are often analogous and may be treated by similar equations. Because of the greater relative development of heat transfer theory, it is frequently useful to set equations for mass transfer processes into forms analogous to those for corresponding heat transfer processes and try to use the correlations of heat transfer variables to predict mass transfer results. But a more direct approach is desirable, aimed at verifying whether this analogy holds under all conditions.

Based on the existing heat transfer theories paralleled in the case of mass transfer by free convection, Wilke (*13*) has derived an equation which is of the same form as the general equation for heat transfer. This equation has been substantially verified for mass transfer for vertical plates. Boelter, Gordon, and Griffin (*2*) have applied the mass transfer analogy to the evaporation of water into air from a free horizontal quiet surface. Their data were correlated according to the equation:

$$\mathrm{Nu}' = 0.643\,(\mathrm{Gr}' \times \mathrm{Sc})^{0.25} \qquad (3)$$

in the range of values for (Gr′Sc) between 10^7 and 2×10^8. If Saunders' equation for the analogous case of heat transfer from a horizontal surface in the streamline range is taken as:

$$\mathrm{Nu} = 0.54\,(\mathrm{Gr} \times \mathrm{Pr})^{0.25} \qquad (4)$$

the ratio

$$\frac{\mathrm{Nu}}{\mathrm{Nu}'} = \frac{0.54}{0.643} = 0.84 \qquad (5)$$

a value which needs to be verified for other cases.

Other than that cited above, there has been practically no work done in the field of free convection mass transfer. Therefore, a comprehensive investigation has been initiated on processes like solid dissolution, electrodeposition, pure liquid vaporization, and sublimation to verify the existing equations or to develop new correlations to define this process more precisely. This article presents results obtained for the vaporization of some pure liquids into humid air.

EXPERIMENTAL

Figure 1 diagrammatically represents the general setup of the equipment. The evaporating vessel consists of a shallow cylindrical glass vessel, 4 inches in diameter and enclosed in a close-fitting wooden jacket, to prevent setting up of convection currents by the sides of vessel. About a 2-foot length of 30-gage Nichrome wire is coiled and remains submerged in the liquid. The leads are taken out through two glass tubes of very small diameter, supported in a stand. Low tension voltage is applied from the secondary of a variable transformer. A sensitive thermometer is suspended from the stand to record the temperature. The evaporating vessel is placed in one pan of a sensitive balance and counterpoised. The whole assembly, excluding the variable transformer, is enclosed in a large glass chamber to eliminate stray external convection currents. In every case sufficient liquid is introduced to fill the vessel to the brim and a variable voltage is applied to maintain the temperature at the desired level; depending upon the temperature of the liquid, from 1 to 5 grams are removed from the counterpoising pan and the time taken for the pointer of the balance to swing back to the original position is recorded. The experiments are repeated until concurrent values are obtained. It has been possible to

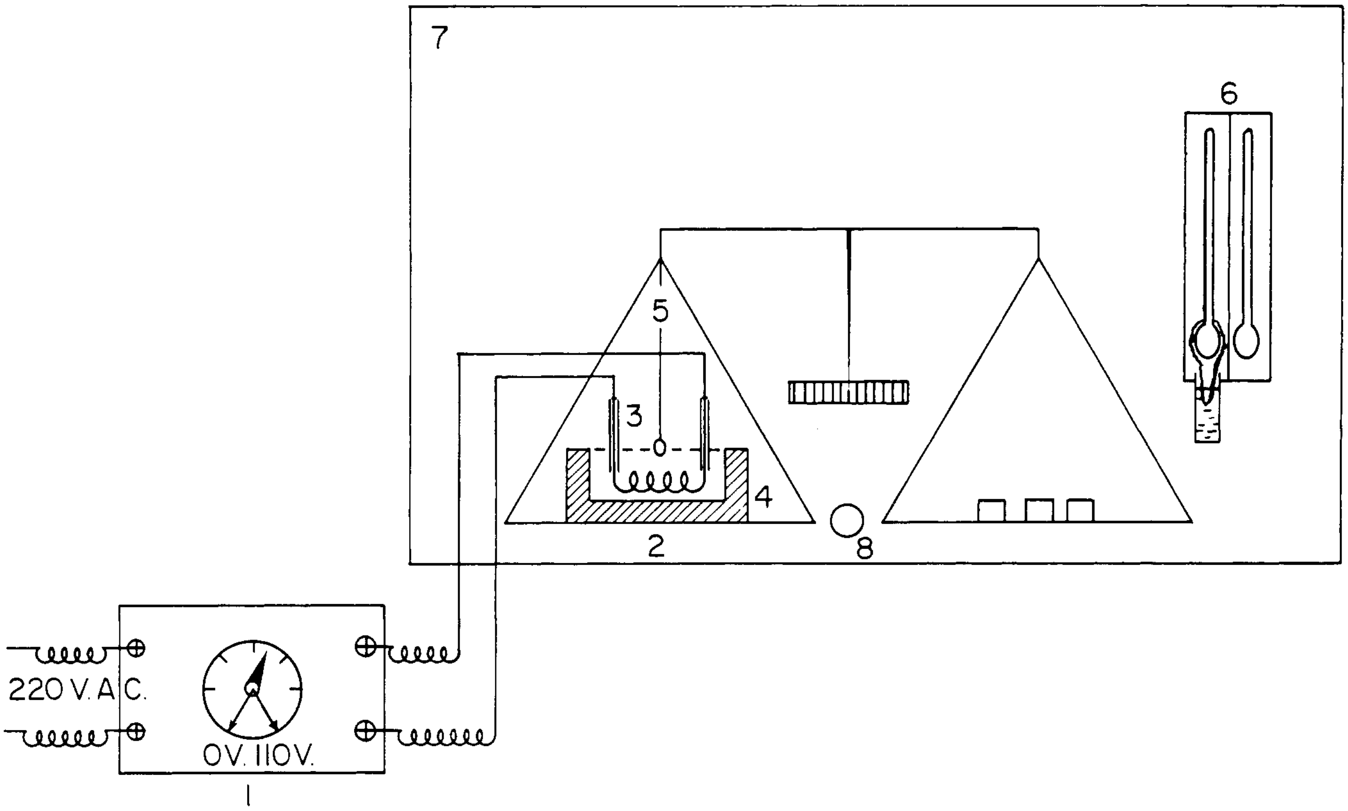

Figure 1. Experimental setup for measurement of vaporization rate of some liquids
1. Variable transformer
2. Sensitive balance
3. Pair of glass tubes to lead in Nichrome coil
4. Thermal insulation for cylindrical evaporating unit
5. Sensitive thermometer
6. Dry bulb-wet bulb assembly
7. Large glass chamber
8. Control knob

control the temperature within ±0.1°C. A wet- and dry-bulb thermometer assembly is also introduced in the chamber and readings are taken from time to time.

CALCULATIONS

In order to evaluate the Nusselt, Grashof, and Schmidt numbers, the following calculations are made.

Mass Transfer Coefficient. As no liquid film is present, the gas film alone controls the process of vaporization. The evaporation rate is obtained from experimental data. From the vapor pressure data (*5*) fugacity potential is calculated. Except in the case of water, where the humidity of the bulk air is to be determined from the dry- and wet-bulb readings, to evaluate the fugacity potential, in all other cases, the vapor tension of the liquid at once provides the fugacity potential. The average film temperature is determined as the arithmetic mean of the interfacial and bulk temperature and all physical properties are evaluated at this temperature. Therefore, a knowledge of the vaporization rate, surface area, and fugacity potential allows the mass transfer coefficient to be evaluated.

Density. The average density of the film as a function of temperature and composition is easily determined from a knowledge of the molecular weights of the three components —air, water, and liquid.

Viscosity. The viscosities of the vapors are taken from the literature (*6*, *10*). In some cases, the viscosity data are not available as a function of temperature. They are determined from the equation proposed by Wilke (*15*), modifying that of Hirschfelder, Bird, and Spotz (*4*), and a set of nomographic charts and collision integrals. The gas mixture viscosity is determined by Wilke's method (*3*).

Diffusion Coefficient. In all cases except that of water, the effective diffusion coefficient is calculated from Wilke's equation (*14*), the variation with temperature being determined from the equation indicated in International Critical Tables (*7*). In some cases, the binary diffusion coefficients are not available. They have been determined from Arnold's equation (*1*). Except for minor information, the above estimated physical properties are sufficient to evaluate the dimensionless groups. As the concentration of the vapor increases in the film, the Schmidt number varies considerably.

DISCUSSION

Figure 2 diagrammatically represents the fugacity temperature and density profiles in the boundary layer, set up during the free convection process. In order to simplify the mechanism of mass transfer, an equivalent film thickness is assumed whose resistance to diffusion is equivalent to that through the boundary layer and wherein the gradients are linear.

It is further assumed that the boundary layer in the present case has a finite thickness, in which the entire partial pressure and temperature gradients are accommodated, so that outside the boundary layer, in the bulk, the temperature and concentration of the diffusing material are uniform. By using a large chamber to house the experimental assembly and allowing sufficient time before taking readings, the above assumption is to some extent justified.

Table I. Vaporization of Water into Quiet Humid Air

Interface Temp., t °C.	Liquid Vaporized, Grams	Time of Vaporization, Min.-Sec.	Vaporization Rate		Fugacity of Water at t°, Atm.	Fugacity of Water in Air, Atm.	Fugacity Potential, Atm.	Mass Transfer Coefficient, Lb.Mole/ Hr.Sq.Ft. Atm.	Film Compn., Mole Fraction		Av. Density of Film, Lb./ Cu.Ft.	Viscosity of Film at Av. Temp., C.p.	
			Lb./hr.sq.ft	Lb.mole/ hr.sq.ft.					Water vap.	Air		Air	Water vapor
25	1	82-10	0.0185	0.00103	0.03125	0.01772	0.0135	0.0756	0.033	0.967	0.0731	0.018	0.0098
30	0.5	15-15	0.0497	0.00276	0.04187	0.01772	0.0242	0.1144	0.038	0.962	0.0723	0.0182	0.01
35	0.5	8-56	0.0849	0.00471	0.0555	0.01772	0.0378	0.1248	0.044	0.956	0.0716	0.0184	0.0101
40	0.5	5-20	0.1421	0.0079	0.07280	0.01772	0.0551	0.1433	0.052	0.948	0.0708	0.0185	0.0102
45	1	6-35	0.2303	0.0128	0.09454	0.01772	0.0767	0.1665	0.062	0.938	0.0699	0.0185	0.0103
50	1	4-33	0.3333	0.0185	0.1217	0.01772	0.1040	0.1780	0.074	0.926	0.0690	0.01865	0.0104
55	2	5-33	0.5513	0.306	0.1553	0.02014	0.1352	0.2260	0.09	0.91	0.0681	0.01875	0.0105
60	2	4-12	0.7219	0.0401	0.1965	0.02014	0.1764	0.228	0.108	0.892	0.0671	0.0188	0.0106
65	2	3-4	0.9888	0.0550	0.2467	0.02014	0.2266	0.2424	0.128	0.872	0.0660	0.0189	0.0107
70	5	4-45	1.596	0.0887	0.3075	0.02014	0.3084	0.3084	0.15	0.85	0.0649	0.0191	0.0108

Table II. Vaporization of Absolute Ethyl Alcohol into Quiet Humid Air

Interface Temp., t °C.	Liquid Vaporized, Grams	Time of Vaporization, Min.-Sec.	Vaporization Rate		Fugacity of Ethyl-Alcohol at t °C., Atm.	Mass Transfer Coefficient, Lb.Mole/ Hr.Sq.Ft. Atm.	Compn. of Film at Av. Film Temperature, Mole Fraction			Av. Density of Film Lb./ Cu.Ft.	Av. Viscosity of Film Lb./ Ft.Sec. $\times 10^4$
			Lb./hr.sq.ft.	Lb.mole/ hr.sq.ft.			Alcohol	Air	Water		
25	1	30-20	0.0600	0.00130	0.07764	0.01399	0.0374	0.9456	0.017	0.07515	0.1177
30	1	21-34	0.0843	0.00183	0.1033	0.01479	0.049	0.9342	0.0168	0.07504	0.1183
35	1	15-10	0.1240	0.00269	0.1364	0.01593	0.0639	0.9196	0.0166	0.07506	0.1187
40	1	8-8	0.1864	0.00405	0.1780	0.02277	0.0817	0.9020	0.0163	0.0752	0.1179
45	1	5-50	0.2599	0.00565	0.2290	0.0267	0.1028	0.8813	0.0159	0.07549	0.1168
50	1	4-10	0.3639	0.00791	0.2924	0.02706	0.1276	0.857	0.0154	0.07592	0.1163
55	2	6-17	0.4839	0.01052	0.3693	0.02848	0.156	0.829	0.015	0.07652	0.1152
60	2	4-56	0.6006	0.01305	0.4640	0.02879	0.1883	0.7973	0.0144	0.07726	0.1140
65	2	4-1	0.7518	0.01634	0.5906	0.02892	0.228	0.758	0.014	0.07826	0.1124

Table III. Vaporization of Benzene into Quiet Humid Air

Interface Temp., t °C.	Liquid Vaporized, Grams	Time of Vaporization, Min.-Sec.	Vaporization Rate		Fugacity of Benzene at t °C., Atm.	Mass Transfer Coefficient, Lb.Mole/ Hr.Sq.Ft. Atm.	Compn. of Film at Av. Film Temperature, Mole Fraction			Av. Density of Film Lb./ Cu.Ft.	Av. Viscosity of Film Lb./ Ft.Sec. $\times 10^4$
			Lb./hr.sq.ft.	Lb.mole/ hr.sq.ft.			Benzene	Air	Water		
30	1	12-5	0.1238	0.001587	0.1550	0.01025	0.072	0.908	0.02	0.0819	0.1168
35	1	9-23	0.1616	0.002072	0.1938	0.01070	0.0882	0.892	0.0198	0.08311	0.1162
40	1	7-2	0.2156	0.002765	0.2390	0.01155	0.1068	0.874	0.0194	0.0848	0.1149
45	1	5-40	0.2676	0.003431	0.2930	0.0117	0.1278	0.853	0.0189	0.0867	0.1142
50	1	4-30	0.3370	0.00432	0.3546	0.01216	0.1506	0.831	0.0184	0.0887	0.1131
55	2	6-55	0.4374	0.005608	0.4264	0.01315	0.1758	0.806	0.0179	0.0910	0.1120
60	2	6-20	0.5685	0.007291	0.5091	0.01430	0.203	0.780	0.0171	0.0935	0.1107
65	2	4-12	0.7220	0.009258	0.6050	0.0153	0.2325	0.751	0.017	0.9060	0.1096
70	2	2-40	1.083	0.01390	0.7160	0.0194	0.2632	0.719	0.0169	0.0989	0.1075

Table IV. Vaporization of Carbon Tetrachloride into Quiet Humid Air

Interface Temp., t °C.	Liquid Vaporized, Grams	Time of Vaporization, Min.-Sec.	Vaporization Rate		Fugacity of Carbon Tetrachloride at t °C., Atm.	Mass Transfer Coefficient, Lb.Mole/ Hr.Sq.Ft. Atm.	Compn. of Film at Av. Film Temperature, Mole Fraction			Av. Density of Film Lb./ Cu.Ft.	Av. Viscosity of Film, Lb./ Ft.Sec. $\times 10^4$
			Lb./hr.sq.ft.	Lb.mole/ hr.sq.ft.			CCl_4	Air	Water		
25	1	6-25	0.2363	0.001537	0.1507	0.0102	0.0701	0.9133	0.0165	0.0959	0.1162
30	1	5-15	0.2889	0.001875	0.1881	0.0107	0.0856	0.8976	0.0162	0.1007	0.1156
35	1	3-54	0.3889	0.00253	0.2318	0.0111	0.1039	0.8802	0.0159	0.1049	0.1150
40	1	3-8	0.484	0.003147	0.2839	0.0111	0.1243	0.86	0.0155	0.1104	0.1144
45	1	2-43	0.5582	0.00363	0.3453	0.0121	0.1472	0.8877	0.0151	0.1165	0.1138
50	2	3-45	0.8089	0.00526	0.4173	0.0126	0.1724	0.813	0.0147	0.1233	0.1132
55	2	2-50	1.07	0.00696	0.499	0.01395	0.1996	0.7863	0.01417	0.1306	0.1128
60	2	2-2	1.491	0.00970	0.5932	0.0164	0.2288	0.7575	0.0137	0.1383	0.1124
65	2	1-30	2.022	0.01314	0.6985	0.0188	0.258	0.7278	0.0132	0.1459	0.1122

In the case of ethyl alcohol vaporization, sampling of the air farthest from the evaporating surface indicated only traces of the diffusing component for a considerable length of time. Therefore, assuming the fugacity potential of the diffusing component to be equivalent to the vapor pressure at the interface will not involve much error in computing the mass transfer coefficient in the case of organic liquids.

In all cases the interfacial temperatures are higher than that of bulk air. In the case of water, the combined influence of both the temperature and density gradients on the free convection process is to intensify the convection

Av. Visc. of Film Lb./Ft.Sec. ×10⁴	Diffusion Coefficient, Sq.Feet/Hr.	$\left[\frac{\rho_\infty}{\rho s} - 1\right]$	Nu′	Gr′ ×10⁻⁶	Sc	Gr′× Sc′ ×10⁻⁶
0.1208	1.008	0.005	9.55	0.219	0.59	0.129
0.1221	1.022	0.029	14.26	1.213	0.595	0.72
0.1232	1.039	0.055	15.36	2.22	0.599	1.33
0.1236	1.048	0.081	17.5	3.17	0.60	1.90
0.1238	1.068	0.124	19.86	4.72	0.60	2.83
0.1246	1.083	0.137	20.79	5.02	0.601	3.01
0.1250	1.098	0.17	25.74	6.01	0.602	3.62
0.1257	1.114	0.208	25.22	7.06	0.606	4.28
0.1260	1.129	0.252	25.92	8.25	0.609	5.03
0.1263	1.145	0.304	31.57	9.57	0.612	5.86

Effective Diffusion Coefficient, D, Sq.Ft./Hr.	$\left[\frac{\rho_\infty}{\rho s} - 1\right]$	Nusselt No.	Grashof No., ×10⁻⁶	Schmidt No.	Gr × Sc, ×10⁻⁶
0.5206	0.0410	3.38	1.994	1.084	2.161
0.5290	0.0375	3.49	1.8	1.073	1.932
0.5391	0.0358	3.68	1.707	1.056	1.80
0.5467	0.0366	5.08	1.776	1.032	1.833
0.5562	0.0399	6.4	1.988	1.002	2.028
0.5650	0.0452	5.6	2.297	0.9763	2.242
0.5737	0.0521	5.55	2.742	0.9448	2.59
0.5815	0.0603	6.47	3.3	0.9114	3.014
0.5924	0.0717	7.10	4.147	0.8728	3.619

Effective Diffusion Coeff., D, Sq.Ft./Hr.	$\left[\frac{\rho_\infty}{\rho s} - 1\right]$	Nusselt No.	Grashof No., ×10⁻⁷	Schmidt No.	Gr × Sc, ×10⁻⁷
0.3760	0.1704	9.904	0.999	1.366	1.365
0.3876	0.1875	9.902	1.14	1.299	1.486
0.3887	0.2073	10.52	1.35	1.254	1.69
0.3951	0.228	10.22	1.57	1.200	1.88
0.4017	0.2492	10.16	1.83	1.142	2.09
0.4082	0.2693	10.42	2.07	1.085	2.25
0.4146	0.2896	10.66	2.47	1.028	2.534
0.4225	0.3111	10.85	2.85	0.9723	2.77
0.4291	0.3284	12.21	3.32	0.912	3.024

Effective Diffusion Coeff., D, Sq.Ft./Hr.	$\left[\frac{\rho_\infty}{\rho s} - 1\right]$	Nusselt No.	Grashof No., ×10⁻⁸	Schmidt No.	Gr × Sc, ×10⁻⁸
0.305	0.3623	39.99	2.65	1.43	3.79
0.308	0.397	39.1	3.18	1.35	4.29
0.315	0.431	41.4	3.85	1.25	4.81
0.321	0.4647	39.6	4.61	1.17	5.44
0.326	0.4917	36.0	5.55	1.08	6.00
0.331	0.5239	41.2	6.76	1.02	6.76
0.338	0.5496	42.5	7.89	0.92	7.26
0.342	0.5728	46.5	9.26	0.85	7.86
0.356	0.5926	48.0	10.81	0.78	8.42

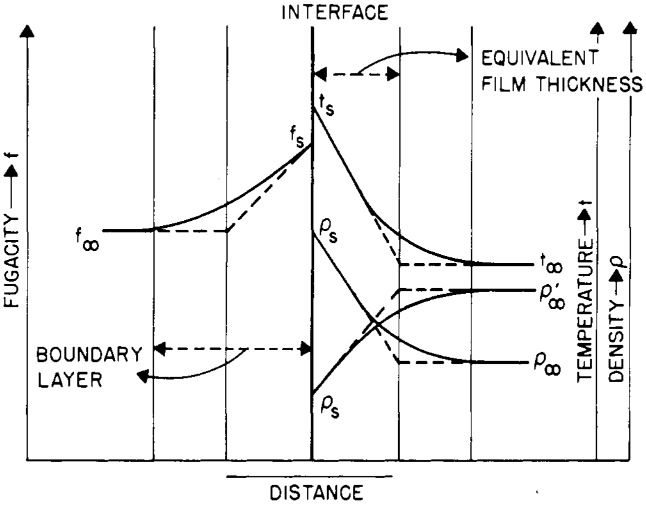

Figure 2. Fugacity, temperature, and density profiles during isothermal vaporization of some pure liquids into quiet humid air

currents. In all other cases where the average molecular weight of the film is higher than that of bulk air, the two gradients act against each other, thereby effectively reducing the free convection currents. In fact, in some cases the process is almost entirely governed by molecular diffusion and the effect of free convection currents is almost negligible. It is, therefore, natural to anticipate two sets of correlations, one for liquids whose molecular weights are lower and the other for those whose molecular weights are higher than that of bulk air. Figure 3 substantially verifies the above statement.

For water the general correlation is:

$$Nu' = 0.62\ (Gr' \times Sc)^{0.25} \qquad (6)$$

In the case of the other liquids,

$$Nu' = 0.17\ (Gr' \times Sc)^{0.25} \qquad (7)$$

Taking water vaporization as an analogous case to heat transfer, under identical (Gr′ × Sc) values:

$$\frac{Nu}{Nu'} = \frac{0.54}{0.62} = 0.872$$

a value which is nearly the same as that in Equation 5. For identical processes again,

$$\frac{Nu}{Nu'} = \frac{(Pr)^n}{(Sc)} \qquad (8)$$

a ratio whose value may be determined from a knowledge of the Schmidt and Prandtl numbers for the various systems.

Equation 7 for organic liquids is analogous to that

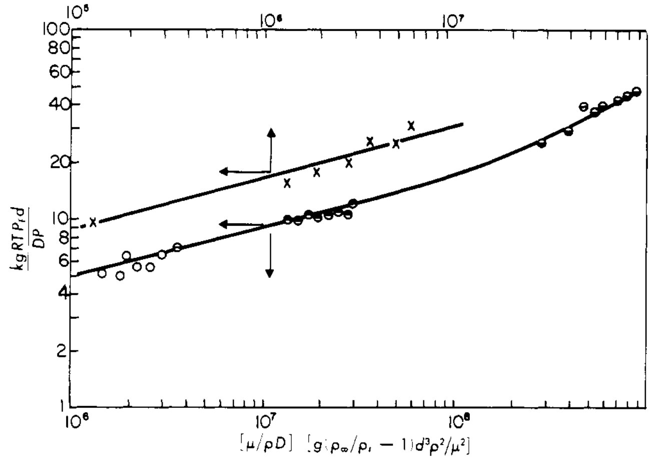

Figure 3. Correlation of free convection mass transfer data during isothermal vaparization of some common liquids in quiet humid air

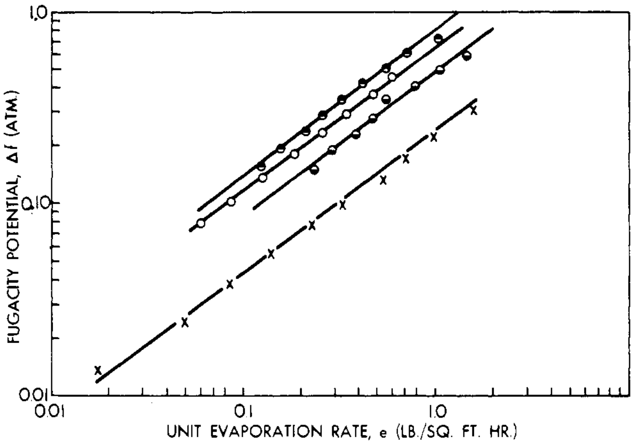

Figure 4. Dependence of vaporization rate on fugacity potential

defining the process of heat transfer from a horizontal plate facing downward (7).

The unit vaporization rate is plotted against fugacity potential for each liquid in Figure 4 and the data can be well correlated by the following equation:

$$e = C \cdot \Delta f^{1.32} \tag{9}$$

where the value of C varies from liquid to liquid.

Liquid	C
Water	6.58
Carbon tetrachloride	2.96
Ethyl alcohol	1.80
Benzene	1.33

It is anticipated that continuation of this part of the investigation under sub- and superatmospheric pressure will provide valuable information, from which more general relations can be derived.

NOMENCLATURE

Cp = heat capacity at constant pressure, B.t.u. per °F.
D = diffusion coefficient, sq. feet per hour
d = characteristic dimension, feet
e = unit vaporization rate, $\frac{\text{lb.}}{\text{hr. sq. ft.}}$
f = fugacity, atm.
g = acceleration due to gravity, ft./sec.^{2}
h = heat transfer coefficient, B.t.u./hr. sq. ft. °F.
k = thermal conductivity, B.t.u./ft. °F./ft.

kg = mass transfer coefficient, lb. moles/hr., sq. ft. atm.
P = total pressure, atm.
pf = film pressure factor, atm.
R = gas constant, atm. cu. ft./(lb. mole.) OR °
t = temperature, °F.
u = convective velocity, feet per second
μ = viscosity of air film, lb./ft. hr.
ρ = density of air film, $\frac{\text{lb.}}{\text{cu. ft.}}$
θ = reciprocal average temperature, $\frac{1}{°\text{F.}}$
Δf = fugacity potential of vapor, atm.
Δm = temperature difference between interface and bulk air, °F.
ϕ = unique function defined by Equations 1 and 2

Dimensionless Groups

$$\frac{Cp \cdot \mu}{k} = \text{Prandtl number}$$

$$\frac{\mu}{\rho D} = \text{Schmidt number}$$

$$\frac{g \cdot \theta \cdot \Delta m \cdot d^3 \cdot \rho^2}{\mu^2} = \text{Grashof number}$$

$$\frac{g \cdot d^3 \cdot \rho^2 \cdot \frac{\rho_\infty}{\rho s} - 1}{\mu^2} = \text{Grashof number for mass transfer}$$

$$\frac{hd}{k} = \text{Nusselt number}$$

$$\frac{kg \cdot R \cdot T \cdot pf \cdot d}{D \cdot P} = \text{Nusselt number for mass transfer}$$

$$\frac{du\rho}{\mu} = \text{Reynolds number}$$

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Upper Explosive Limits of Cumene

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In common with most other hydrocarbons, cumene forms explosive mixtures with air. These explosive mixtures are limited or rendered nonexplosive in three ways: by diluting with air until the mixture is too lean to explode (lower limit), by diluting with fuel until the mixture is too rich to explode (upper limit), and by diluting with an inert substance until there is insufficient oxygen to explode.

Previously, only a small amount of information was available concerning the explosive limits of cumene (*1–3*), and this information pertained only to the limit as a function of the oxygen-nitrogen-cumene ratio at atmospheric pressure. However, it is impossible to predict the effect of

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