

to nonsaturation of specific absorption sites, was demonstrated previously for riboflavin (17).

Increasing the accompanying water volume in fasted subjects might be expected to increase the dissolution rate of chlorothiazide and to accelerate the stomach emptying rate (7). However, neither factor should markedly alter the absorption efficiency of chlorothiazide. Faster stomach emptying would increase the rate at which drug passes the active absorption site and, hence, reduce its bioavailability. Faster dissolution should have little effect since chlorothiazide has been shown to be only 25% bioavailable when dosed as an oral solution (6).

It is instructive to compare chlorothiazide plasma levels and urinary excretion as indicators of chlorothiazide bioavailability. The mean urinary recoveries of chlorothiazide from Treatments A, B, and C were 12.3, 14.9, and 24.7% of the dose, respectively, while the respective areas under the plasma curves to 48 hr were 3.0, 3.4, and 6.4 $\mu\text{g hr/ml}$, indicating excellent agreement between plasma and urine data. The overall correlation between the percentage of dose excreted and the areas under the plasma level curves was 0.726 ($p < 0.001$), while the correlation between peak drug levels in plasma and percent excretion was somewhat lower at 0.480.

Similarly, the mean ratios of the percentage of dose excreted between Treatments A and B, A and C, and B and C were 0.87, 0.51, and 0.63; the equivalent ratios of areas under the plasma curves between these treatments were 0.92, 0.47, and 0.57.

To compare the kinetics of chlorothiazide recovery in urine with those of drug loss from plasma, the urinary excretion rates following the three treatments were calculated at the midpoint of each urine collection interval and the mean values were plotted along with the plasma levels in Figs. 1-3. Comparison of the curves indicates close similarity between the urinary excretion rates and plasma chlorothiazide levels throughout the entire sampling period.

Chlorothiazide absorption, which is normally poor, is doubled by the presence of food. This fact, together with the insensitivity of chlorothiazide absorption to varying fluid volumes, supports the view that chlorothiazide absorption is saturable and occurs at a particular site in the GI intestinal tract (5, 6). For optimal absorption, chlorothiazide tablets should be taken with or immediately after meals.

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Tray Drying of Pharmaceutical Wet Granulations

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Abstract □ It is shown that in tray drying wet granulated materials, the expected log-linear plots give slopes that are inversely proportional to the bed depth, rather than to the depth squared. A model is proposed for this giving diffusion coefficients of the order of magnitude expected for liquid-water diffusion. The temperature dependence also suggests that this is the rate-limiting process rather than vapor diffusion.

Keyphrases □ Tray drying—wet granulated materials, mathematical models □ Models, mathematical—tray drying of wet granulated materials □ Granulations—tray drying, mathematical models

The drying of granulations is an important pharmaceutical operation and tray drying is a frequently used method of water removal. The way in which drying takes place from a tray (1-3) can be visualized in several ways. The drying could be a function of the individual granule rather than of the mass of granules, *i.e.*, the bed. In that case, the bed thickness would not be a factor (in fluid bed drying it is not of great importance). However, bed thickness is important, and the drying is a function of the properties of the bed. In this case there are two possibilities: (a) either the movement through the void space be-

tween the granules is important (*i.e.*, the vapor diffusion is rate determining), or (b) the drying is primarily from the bed surface, and liquid movement up through the bed maintains a water concentration profile in the bed.

BACKGROUND

In the following discussion, the nomenclature outlined in Fig. 1 will be used. The bed is l -cm deep, has a porosity of ϵ (*i.e.*, a solids fraction of $(1 - \epsilon)$). It has a cross-sectional area of A cm^2 . The density of an anhydrous granule is ρ gm/cm^3 . The wet granulation before drying contains C^* g of moisture per g of anhydrous solid, and after drying contains c g of moisture per g of anhydrous solid. If c is the equilibrium concentration, then $C^* - c = C_0$ is the initially removable moisture concentration, and $C' - c = C$ is the moisture concentration which is removable at time t . The latter is the quantity which will be dealt with in the following discussion. Granules containing c g of moisture/g of anhydrous weight will be denoted dry. If x is a distance coordinate measured from the top of the bed (Fig. 1), then the diffusional equation governing the situation is Fick's law, *i.e.*, at time t :

$$\partial C/\partial t = D\partial^2 C/\partial x^2 \quad (\text{Eq. 1})$$

where D is the diffusion coefficient of water. Crank (4) solved this equation using the following boundary conditions: $C = 0$ at $x = 0$, for t

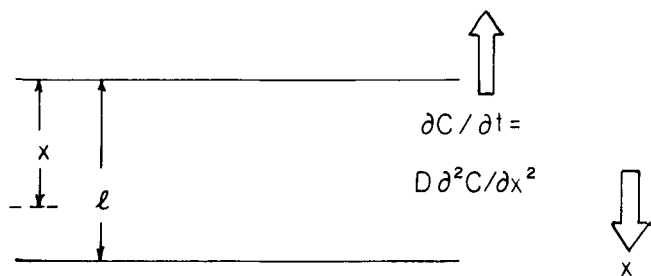


Figure 1—Schematic of drying from a bed.

> 0 (no moisture on the surface); $C = C_0$ for $0 < x < l$ at $t = 0$ (the same amount of moisture exists at each point throughout the interior at time zero); and $\partial C/\partial x$ at $x = 0$ equals zero for $t > 0$. Under these conditions it can be shown that Eq. 1 leads to the equation:

$$\ln(M/M_0) = -\{\pi^2 D/(4l^2)\}t + \ln(8/\pi^2) = -kt - 0.2 \quad (\text{Eq. 2})$$

where M is the amount of removable water or the volume multiplied by $(C' - c)$. The equation is valid after a short lag time, and is based on the stated boundary conditions. For tray drying of granulations, the condition that the surface moisture for $t > 0$ is zero, is questionable. In Eq. 2, k is a drying rate constant given by:

$$k = -\pi^2 D/(4l^2) \quad (\text{Eq. 3})$$

This equation predicts that the drying data (after a short lag time) should be log-linear when the amount of removable moisture is plotted versus time. This drying profile is encountered frequently (e.g., in the so-called falling-rate period). The drying rate constant should have the same temperature dependence as D , the diffusion coefficient for liquid water. Furthermore, k should be inversely proportional to the second power of the bed thickness.

If vapor diffusion through the bed is the rate-limiting step (3-9), it is the movement through the void space of the bed which is rate-limiting. The change from constant to falling rate was postulated (10) to be the change from a process where there is capillary action within the granule to one where there is moisture movement within the void space of the bed. Beyond this some authors (11-13) considered the drying as a receding front down through the bed. This was shown to be feasible (14), and for a fine powder the diffusion coefficient is equal to that of water vapor diffusion.

In this type of drying there will be a dry part containing c g of moisture/g of anhydrous solid (x cm in Fig. 1) and a wet part containing C_0 g of moisture/g of dry solid [the lower $(l - x)$ cm in Fig. 1]. Drying occurs by vapor diffusion over a thickness of x cm through a free cross-section of $A \epsilon$ cm². Fick's law now takes the form:

$$-(1/A\epsilon) dM/dt = \Pi P/x \quad (\text{Eq. 4})$$

for a steady state where Π is a permeability constant, and P is the water vapor pressure. This is assumed equal to zero in the air stream. There is C_0 g of moisture/g of dry solid in the lower $(l - x)$ cm of the bed and it is apparent that P is a function of C_0 .

Figure 1 shows that in drying a section of thickness dx , the amount of water removed is:

$$-dM = C_0 \rho (1 - \epsilon) A dx \quad (\text{Eq. 5})$$

Introducing Eq. 5 into Eq. 4 then gives:

$$[C_0 \rho (1 - \epsilon)/\epsilon] dx/dt = \Pi P/x \quad (\text{Eq. 6})$$

which can be integrated, taking initial conditions into account, to give:

$$x = F \cdot \sqrt{t} \quad (\text{Eq. 7})$$

where:

$$F^2 = 2\Pi P \epsilon / [C_0 (1 - \epsilon) \rho] \quad (\text{Eq. 8})$$

The removable moisture content in the bed at time t (neglecting the amount in the vapor phase) is $M = C_0 (l - x) A (1 - \epsilon) \rho$, and that at time zero it is $M_0 = C_0 l A (1 - \epsilon) \rho$ (Fig. 1), so that:

$$M/M_0 = 1 - (x/l) = 1 - (F/l) \cdot \sqrt{t} \quad (\text{Eq. 9})$$

Equation 9 predicts a drying rate which is a square root of time dependence and where the rate constant is K , given by:

$$K = F/l \quad (\text{Eq. 10})$$

i.e., inversely proportional to bed depth.

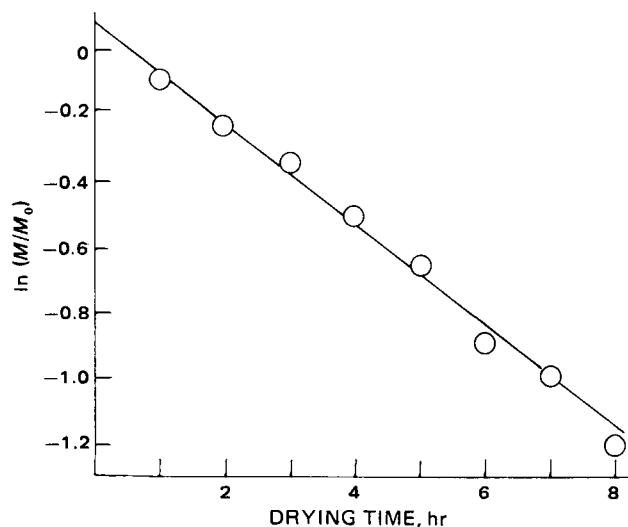


Figure 2—Typical drying profile in tray drying. $T = 50^\circ$, $l = 1.91$ cm ($3/4$ in.).

The above hypotheses (Eq. 2 and Eq. 9) are *a priori* possible for tray drying. The model leading to Eq. 9 would seem more logical in the drying of wet granulations. Several reports (15-18) have reported this type of drying. Some investigators (17, 18) have shown that the equilibrium granule is primarily a fairly coarse and porous body. In tray drying the bed is loosely spread and fairly shallow (19-21), and movement in the highly porous space is probably not rate limiting. Nevertheless, the present report will show that some inconsistencies exist relating to Eq. 9, and the purpose of the present report is to probe which of the two situations (if any) actually exists. If neither applies an alternate model consistent with the data presented will be suggested.

EXPERIMENTAL

Granulations were made in the following manner: 5.75 kg of lactose USP and 0.8 kg of cornstarch USP were mixed in a sigma-type blender¹, 2 liters of cornstarch paste containing 10% cornstarch were added, and the mass kneaded for 15 min. It was passed through a No. 4 hand screen and spread onto a drying tray, which was placed in a tray drying oven². The tray was removed at given intervals and weighed *in toto* on a balance³. The temperature was recorded on a digital readout thermometer⁴. Weights were usually monitored every hour for the complete drying period. Experiments were carried out at five bed depths and four different temperatures, as shown in Table I.

DISCUSSION

It should first be stated that the drying curves in this study were all semilogarithmic in time (Fig. 2). Hence, Eq. 9 is not obeyed in the strictest sense. Equation 2, however, is not strictly obeyed either. The drying curves have intercepts close to zero⁵ (or slightly above), certainly not close to -0.2. The least-squares fit values for the slopes and intercepts are shown in Table I. Adherence to the semilogarithmic relation $\ln(M/M_0) = -k^*t$ seems excellent under all conditions.

The conventional diffusion model leading to Eq. 2 requires that the rate constants, k , be inversely proportional to the second power of the bed depth. A plot of $\ln k$ versus $\ln l$ is shown in Fig. 3. This plot has a slope of ~ -1.0 and hence k appears to be inversely proportional to l and not inversely proportional to l^2 . The model leading to Eq. 2 fails on two points. Equation 9 would appear to fail based on the time dependence, but the bed thickness effect, on the surface, would be of the correct dependence. The following presents a probe of whether Eq. 9 could indeed be the correct model. This is in spite of the time dependence failure. This may

¹ Z-bar blender, J. H. Day Co., Cincinnati, Ohio.

² Stokes model 38A dryer. Tray is 71.1 cm \times 88.9 cm (28 in. \times 35 in.) Oven is 91.4 cm (36 in.) wide and 144.8 cm (57 in.) high with pilot tube. It is 82.55 cm (32.5 in.) deep and has a 16-tray capacity. The air velocity is 8.93 m³/min (280 ft³/min).

³ Mettler, P.S. 30.

⁴ Mensor Co. platinum probe thermometer with digital readout.

⁵ It should be pointed out that the models presented do not account for thermal effects. The time required for initial thermal equilibrium, experimentally, could account for the zero or slightly positive intercepts.

Table I—Least-Squares Parameters for the Drying Data (Eq. 2)

Temperature	Thickness		Intercept	Negative of Slope, k hr	Correlation Coefficient, r^2	1000/ T , °K ⁻¹	ln k	ln k (adjusted to 12.5°) ^a
	l , cm	ln l						
60°	2.54	0.93	0.1073	0.1595	0.997	3.002	-1.836	-3.828
60°	2.54	0.93	0.1110	0.1472	0.995	3.002	-1.916	-3.908
55°	2.54	0.93	0.0480	0.119	0.999	3.047	-2.129	-3.941
50°	5.08	1.63	0.0086	0.0481	0.999	3.104	-3.034	-4.618
50°	2.54	0.93	0.034	0.1045	0.999	3.095	-2.259	-3.879
50°	2.54	0.93	0.040	0.0992	0.999	3.095	-2.311	-3.931
50°	1.91	0.65	0.0877	0.1544	0.997	3.104	-1.868	-3.452
40°	1.27	0.24	0.059	0.116	0.998	3.193	-2.154	-3.096
40°	1.91	0.65	0.0311	0.1028	0.999	3.193	-2.275	-3.382
40°	1.91	0.65	0.110	0.1093	0.984	3.193	-2.214	-3.503
40°	2.54	0.93	0.019	0.0697	0.999	3.193	-2.663	-3.442

^a It is shown at a later point that the activation energy for k is 8,000 cal/mole. The adjustment to 12.5° ($1/T = 0.0035$) has hence been made by the formula: $\ln k(12.5^\circ) = \ln k - 4000(0.0035 - 1/T) = \ln k - 14 + 4(1000/T)$ where $\ln k$ is the value listed in the next to the last column. The average value of $\ln k$ at a 2.54-cm depth (six values) is -3.822 with a standard error of the mean of 0.078 and the average value of $\ln k$ at a bed depth of 1.91 cm (three values) is -3.446 with a standard error of the mean of 0.035. The means and 95% confidence limits are shown in Fig. 3. The least squares fit of $\ln k(25^\circ)$ regressed on $\ln l$ for all the values (11 points) is: $\ln k(25^\circ) = -1.14 \ln l - 2.754$ (correlation coefficient: -0.938).

not be serious since it has been shown (23) that in limited ranges, square root relations may have the appearance of semilogarithmic plots.

To evaluate this, theoretical values of M/M_0 have been calculated for various drying times, t , according to Eq. 9 (Table II). The total drying time, θ , for situations given by Eq. 9 are the points in time where M reaches zero at $x = l$, i.e., where $(F/l) \cdot \sqrt{\theta} = 1$. If, for example, this were 9 hr, then $(F/l) = 1/3$. Values for θ are shown in the headings of Table II for four chosen values of (F/l) . Once (F/l) is fixed one can calculate M for various times; four time values ($t = 1, 2, 3,$ and 4 hr) are chosen here and are shown in Table II. The value of $\ln(M/M_0)$ calculated for each value of t is shown in the table. For each value of (F/l) there are hence four values of t and $\ln(M/M_0)$. The least-squares fit of:

$$\ln(M/M_0) = -k''t + b \quad (\text{Eq. 11})$$

has been calculated and the least-squares fit parameters for slope ($-k''$) and intercept (b) shown (Table II). The correlation coefficients are good, showing that the square root data are well approximated by a log-linear equation (Eq. 2) for the four chosen values of (F/l) . The intercepts are close to zero. An explanation for the approximation is given in Appendix I.

It is seen from the last two lines in Table II that there is a connection between the calculated values of k'' and the chosen values of (F/l) . This is presented graphically in Fig. 4, and good linearity seems to exist demonstrated by the fact that the least-squares fit:

$$\ln k'' = 2.24 \ln(F/l) + 1.055 \quad (\text{Eq. 12})$$

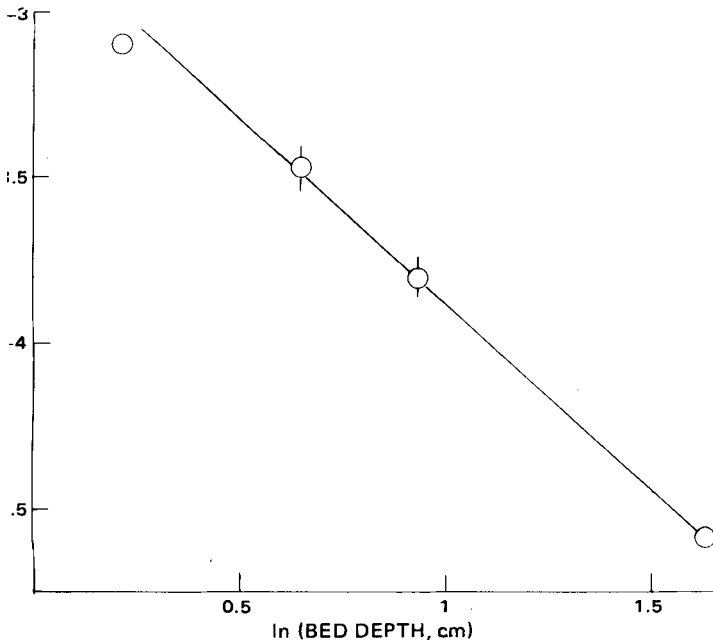


Figure 3—Drying rate constants (adjusted to standard temperature) as a function of bed depth. Detailed data are shown in Table I. Least-squares fit is $\ln k = -1.14, \ln l - 2.754$.

has a correlation coefficient of 0.997. Hence k'' is approximately inversely proportional to the bed depth squared. Equations 2 and 9 are therefore experimentally indistinguishable. When the amount of removable moisture is plotted logarithmically versus time, approximately straight lines will occur in both cases. According to either model, however, the drying rate constants should be inversely proportional to the bed depth squared (or raised to a power higher than 2). Over a longer period of drying time the approximation leading to Eq. 11 is less precise and the treatment, so far, would favor some model based on the general principles leading to Eq. 2.

The original boundary conditions leading to Eq. 5 are that $C = 0$ at the surface, that C equals a non-zero value at the interior, and that dC/dx is equal to zero at $t > 0$ right at the surface of the bed. Other reasonable concentration profiles can be visualized in the interior as shown in Fig. 5. In this figure it is assumed that the moisture content increases steadily (although not linearly) down through the bed; it attains its largest value at the bottom ($x = l$) and is smallest at the surface ($x = 0$). The profile will change with time; both $C(l)$ and $C(0)$ will decrease. If it is assumed that $C(l)$ is proportionally larger than $C(0)$ at all times, so that:

$$C(l) = \gamma C(0) \quad (\text{Eq. 13})$$

at all times (where γ is a time-independent constant), then it is possible to find a trigonometric function such that Eqs. 1 and 13 are both satisfied and that the drying rate constant k will be inversely proportional to the bed depth, l . The function:

$$C(x,t) = (\beta \sin \lambda x + \cos \lambda x) \cdot \exp(-\lambda^2 D t) \quad (\text{Eq. 14})$$

is a solution to Eq. 1 (β and λ are constants discussed later). It will obey Eq. 13 provided that:

$$\gamma = \beta \sin \lambda l + \cos \lambda l \quad (\text{Eq. 15})$$

which is simply a numerical requirement implying time independence.

For $k (= \lambda^2 D)$ to be proportional to l^{-1} , it is necessary that λ^6 be proportional to $l^{-1/2}$, or:

$$\lambda = \alpha \cdot l^{-1/2} \quad (\text{Eq. 16})$$

For the function to be monotonically increasing in the interval (i.e., having no maximum, which would occur if the trigonometric argument

Table II—Data Adhering to Eq. 9 Treated According to Eq. 11

Parameter	ln(M/M ₀) at Time t ^a			
	6	9	12	15
1	-0.52	-0.41	-0.34	-0.30
2	-0.86	-0.64	-0.52	-0.45
3	-1.23	-0.86	-0.69	-0.59
4	-1.70	-1.10	-0.86	-0.73
(F/l)	0.408	0.333	0.289	0.258
Correlation coefficient, ln(M/M ₀) versus t	-0.997	-0.9999	-0.9999	-0.9999
k''	-0.391	-0.229	-0.173	-0.143
Intercept	-0.1	-0.18	-0.17	-0.16
ln(F/l)	-0.90	-1.11	-1.24	-1.35
ln k''	-0.94	-1.47	-1.75	-1.94

^a Total drying time = θ .

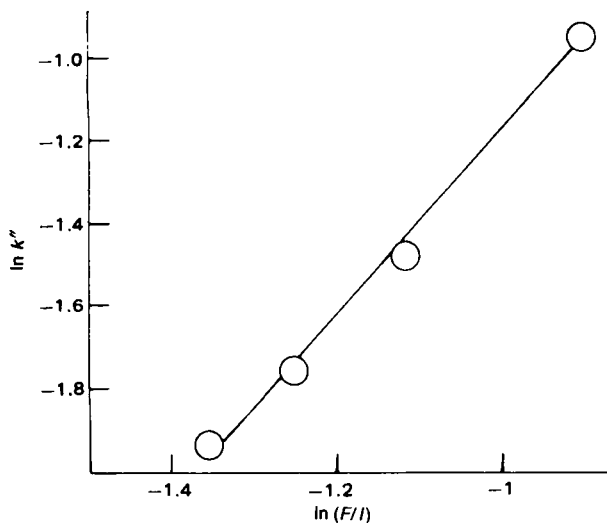


Figure 4—Data from Table II (9 hr drying time); $\ln M/M_0$ plotted as a function of time according to Eq. 9 for data adhering to Eq. 17.

could have the value $\pi/2$, Appendix II), it is necessary that:

$$\lambda l < \pi/4 \quad (\text{Eq. 17})$$

Inserting this in Eq. 16 gives:

$$\alpha l^{1/2} < \pi/4 \quad \text{so} \quad \alpha \leq \pi/(4\sqrt{l}) \quad (\text{Eq. 18})$$

The amount of moisture in the bed at time t is now given by:

$$M = A(1 - \epsilon)\rho e^{-\lambda^2 D t} \int_0^l (\beta \sin \lambda x + \cos \lambda x) dx = A(1 - \epsilon)\rho e^{-\lambda^2 D t} (1/\lambda)(\sin \lambda l - \beta \cos \lambda l + \beta) \quad (\text{Eq. 19})$$

and at time zero it is:

$$M_0 = A(1 - \epsilon)\rho(1/\lambda)(\sin \lambda l - \beta \cos \lambda l + \beta) \quad (\text{Eq. 20})$$

Combining Eqs. 19 and 20:

$$\ln(M/M_0) = -k^* t \quad (\text{Eq. 21})$$

where:

$$k^* = (\alpha^2 D/l) \quad (\text{Eq. 22})$$

This has the correct dependence on l (as opposed to Eqs. 3 and 12). The treatment developed produced three models, two of them conventional (denoted A and B below) and one proposed (denoted C below), the latter based on a trigonometric concentration profile with boundary conditions different from A. The drying curves predicted by the three models are:

model A; $\ln(M/M_0) = -kt - 0.2$ (see Eq. 2)

where $k = -\pi^2 D/(4l^2)$ (see Eq. 3)

model B; $\ln(M/M_0) = -k''t + b$ (see Eq. 11)

where k'' is approximately proportional to $l^{-2.5}$ (see Eq. 12)

model C; $\ln(M/M_0) = -k^*t$ (see Eq. 21)

where $k^* = (\alpha^2 D/l)$ (see Eq. 22)

Models A and B fail on two counts: (a) the experimental drying curves are log-linear with zero or slightly positive intercepts (Table I), and (b) the drying rate constants are experimentally proportional to l^{-1} , not l^{-2} . This prevents calculation of D , the diffusion coefficient, by these models. Model C, on the other hand, predicts zero intercepts and proportionality of drying rate constants to l^{-1} when the fraction of removable moisture is plotted log-linearly in time. This is in accord with experimental results, and permits order of magnitude calculation of D . A test for the reasonability of the model is therefore such a calculation, which should result in values of D in the range expected for a diffusion coefficient, and having a predictable temperature dependence.

It is seen from the inequalities in Eq. 18 that at the limit, α is equal to $\pi/(4\sqrt{l})$, which when inserted in Eq. 22 gives an upper limit for k of

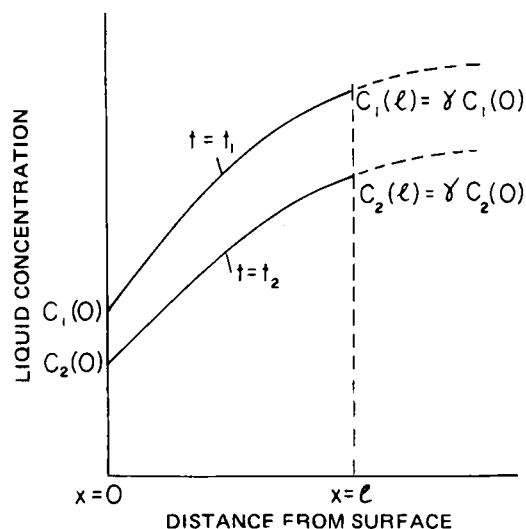


Figure 5—Anticipated moisture concentration profiles in a tray (bed) at two different times, t_1 and t_2 . Curves are according to Eq. 14, and boundary conditions are according to Eq. 13.

$D\pi^2/(16l^2)$. The upper limit for D is then:

$$D = 16kl^2/\pi^2 \quad (\text{Eq. 23})$$

At 50° , for instance (Table I), $l = 2.54$ cm, and $k = 0.1$ hr $^{-1}$, so that:

$$D = (16)(0.1)(2.54)^2/(3600)(3.14)^2 = 28 \times 10^{-5} \text{ cm}^2/\text{sec}, \quad (\text{Eq. 24})$$

which is a reasonable upper limit. Wilburn, *et al.* (23) quote a value of 2×10^{-5} cm 2 /sec at 30° for the liquid diffusion coefficient of water. On the other hand, the D -value in Eq. 24 is not in line with the vapor diffusion coefficient for water [which at 8° is 0.25 cm 2 /sec, (24)].

The value for D determined in Eq. 24 is an upper limit.

A second criterion is the temperature dependence. It is seen from Eq. 22 that k^* should have the same temperature dependence as D :

$$\ln k^* = \ln D + \ln(\alpha^2/l) = -E_d/RT + N \quad (\text{Eq. 25})$$

where N is a constant, and E_d is the activation energy for the diffusion coefficient of water. The data in Table I (2.54-cm bed depth) have been plotted in this fashion in Fig. 6. The least-squares fit line is

$$\ln k^* = -(4015/T) + 1.021 \quad (r^2 = 0.99) \quad (\text{Eq. 26})$$

where r^2 is the correlation coefficient. Although the value for E_d (8030 cal/mol) is somewhat higher than that for the self-diffusion of water ($E_d = 4000$ cal/mole) (24), it is orders of magnitude different from the value of E_d for water vapor.

The model C, where the positional dependence of moisture concentration is a single argument trigonometric term, and where the

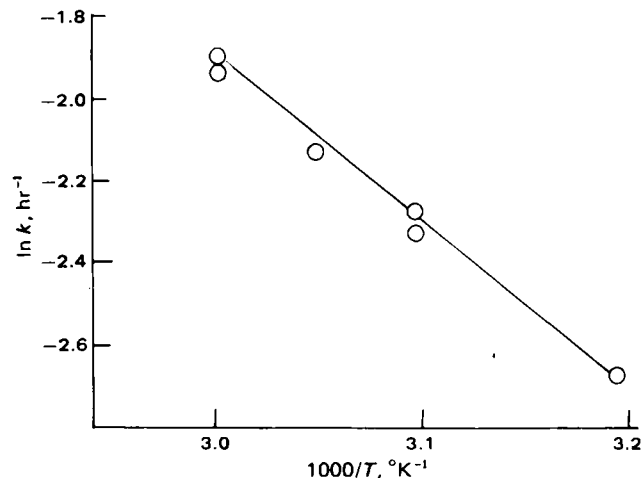


Figure 6—Arrhenius plot of drying rate constants, 2.54-cm bed.

boundary restriction is that the moisture concentration on the bottom of the tray (during steady state) is a constant (*i.e.*, time-dependent) factor times that on the surface, predicts a behavior in agreement with experimental data. The drying data are log-linear with a zero intercept. The drying constant is inversely proportional to bed depth and the drying rate constant is logarithmically linear with inverse temperature. The absolute magnitude of calculated diffusion coefficients and their temperature dependence are in fair agreement with that expected for liquid-water diffusion.

APPENDIX I

If the approximation:

$$y = 1 - (F/l) \cdot \sqrt{t} \simeq Ae^{-kt}, (A \simeq 1) \quad (\text{Eq. A1})$$

holds, then:

$$(1 - y)^2 = (F/l)^2 \cdot t \simeq (1 - Ae^{-kt})^2 \\ \simeq (1 - 2A + A^2) + t[2Ak(1 - A)] + t^2[Ak^2(2A - 1)] \quad (\text{Eq. A2})$$

where the exponential expansion $e^x \simeq 1 + x + x^2/2$ has been truncated for third and higher power terms. The requirements for the approximation in Eq. A2 is that like power terms have equal coefficients, *i.e.*:

$$1 - 2A + A^2 \simeq 1 \quad (\text{Eq. A3})$$

$$(F/l)^2 = 2Ak(1 - A) \quad (\text{Eq. A4})$$

$$t^2Ak^2(2A - 1) \simeq t2Ak(1 - A) \quad (\text{Eq. A5})$$

The last requirement is that $t^2Ak^2(2A - 1)$ be zero or negligible, and this is equivalent to it being significantly smaller than the t term.

The requirement in Eq. A3 is fulfilled when $A \simeq 1$ or $A \simeq 2$. Table II shows that $A \simeq \exp(-0.2) = 0.8$, *i.e.*, that $A \simeq 1$. The second requirement is that $(F/l)^2 = 2Ak(1 - A) \simeq 0.32k$, which in logarithmic form becomes:

$$\ln k = 2 \ln(F/l) - 1.14 \quad (\text{Eq. A6})$$

which compares favorably with Eq. 12, and predicts that k should be proportional to l^{-2} as stated in the text.

The times for which the approximation can hold are deduced from Eq. A5 which can be rearranged to:

$$t < 2(1 - A)/k \quad (\text{Eq. A7})$$

where k is the range 0.1–0.4 in Table II (or 0.1–0.2 in Table I), and A is ~ 0.8 , so that:

$$t < (2 \cdot 0.8/0.1) \text{ to } (2 \cdot 0.8/0.4) = 4\text{--}16 \quad (\text{Eq. A8})$$

as upper limits. The t values in Table II correspond to this. The k , (F/l) , and t ranges used in Table II are within the bounds of the approximation.

APPENDIX II

The function:

$$y = \beta \sin \lambda x + \cos \lambda x \quad (\text{Eq. A9})$$

has the derivative:

$$\partial y / \partial x = \beta \lambda \cos \lambda x - \lambda \sin \lambda x \quad (\text{Eq. A10})$$

This equals zero when:

$$\tan \lambda x = \beta \quad (\text{Eq. A11})$$

β is at least 1 (since C is higher at the bottom of the tray than at the surface), so since $\tan(\pi/4) = 1$, it is necessary that in this limit:

$$\lambda l < \pi/4 \quad (\text{Eq. A12})$$

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