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SOME CALCULATIONS OF DIFFUSION CONTROLLED
THERMAL GRADIENT MASS TRANSFER

ABSTRACT

Calculations are presented of the amount of mass transfer to be expected in liquid alkali metal-alloy systems for two assumed diffusion mechanisms.

In the first of these, the boundary layer is assumed saturated and the rate of transfer limited by the rate of diffusion of solute into the liquid. A general expression is derived including effects of variation in temperature and liquid composition around a closed loop, which simplifies under certain conditions (see page 17) to

$$W(\theta) = k_L S(t_H - t_C)\theta$$

$W(\theta)$ is the amount of mass transferred in time θ , k_L is the conventional mass transfer coefficient, S is the solubility of solute (Ni) in liquid (Na or NaK), t_H and t_C are the hot and cold zone temperatures, respectively. For typical forced convection loop conditions ($t_H = 1500^\circ\text{F}$, $t_C = 1200^\circ\text{F}$, $\theta = 1000$ hrs), this mechanism predicts W to be between 400 and 1000 grams whereas the observed value is about 13 grams. Furthermore, this mechanism predicts W to be only slightly temperature dependent, in contradiction to observation.

In the second mechanism, the rate of mass transfer is assumed to be limited by the rate of diffusion of a component (e.g., Ni) of the solid alloy (e.g., Inconel) up to the solid surface, for which an approximate expression for $W(\theta)$ is derived:

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$$W(\theta) \cong 2\rho_w (D_w \theta/\pi)^{1/2} X(t_H - t_C)/t_H$$

where D_w is the diffusivity of Ni in the solid wall; X , the bulk solid composition; ρ_w , the solid density.

Based on extrapolation of some data for diffusivity of Ni reported in the literature, $W(\theta)$ is estimated to be between 2 and 25 grams for $t_H = 1500^\circ\text{F}$, $t_C = 1200^\circ\text{F}$, $\theta = 1000$ hrs. Note that this is in better agreement with observed mass transfer accumulation than is the calculation based on liquid diffusion. On the other hand, the wall diffusion mechanism does not account for the observed dependence of W on flow rate and time, and does not predict a sharp break in mass transfer at about 1350°F . Table IV, page 33, summarizes the calculations and data.

It is concluded that a more general hypothesis is needed combining the three mechanisms of hot zone attack with a nucleation-deposition mechanism in the cold zone.

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NOMENCLATURE

c	specific heat, Btu/lb-°F
d	inside tube diameter, ft
D	diffusivity, ft ² /hr or cm ² /sec
G	mass velocity, lb/hr-ft ²
h	film heat transfer coefficient, Btu/hr-ft ² -°F
k, k _s	solution rate constants, lb/hr-ft ² -mf
k _L	film mass transfer coefficient, lb/hr-ft ² -mf
k _w	thermal conductivity of solid wall, Btu/hr-ft-°F
L	length, ft
q	activation energy, k-cal/g-mol
Q	heat flux, Btu/hr
R(θ)	mass removal (transfer) rate at θ hrs, lb/hr-ft ²
S	solubility constant, 1/°F
t	temperature, °F
W(θ)	gross mass removal in θ hrs, lb/ft ² (hot surface)
x	weight fraction of transferring component in liquid
x'	weight fraction of transferring component in solid
X	bulk (initial) solid composition, weight fraction
y	distance from entrance to heater, ft
y'	distance from entrance to cooler, ft
θ	time, hr
ρ	density, lb/ft ³
μ	viscosity, lb/ft-hr

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Subscripts

C cold zone
H hot zone
L liquid
W wall
I heater
II cooler

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INTRODUCTION

Thermal gradient mass transfer is the process whereby solute atoms, originating at a solid-liquid interface at a given temperature, precipitate from solution at another interface where the temperature is lower. The consequences of this process in terms of hot-zone attack and cold-zone plugging are too well known to bear repetition. The problem is particularly acute with Ni-base alloys in contact with liquid alkali metals above $1350^{\circ}\text{F}^{1, 2}$. However, little is known regarding the fundamental mechanisms involved, although considerable experimental data have been obtained at ORNL on the influence of the more important variables. It is the purpose of this summary to present some analytical expressions for the rate of mass transfer and for the cumulative build-up for two assumed diffusion mechanisms and to compare the calculations with some available data on Na. Unfortunately, neither mechanism is in satisfactory quantitative agreement with experiment for the range of variables studied, although qualitative agreement with the observed effect of some of the significant variables is noted.

Two basically different mechanisms have been postulated in the literature to explain observed mass transfer rates³. The first of these assumes that the rate of solution of solute is so much lower than the rate of diffusion into the solvent that the interface is never saturated, and the process is thus "solution rate limited." Epstein⁴ has stated his belief that mass transfer of Fe in Na is an example of this.

A second mechanism, postulated to explain the observed mass transfer of Fe in Hg, assumes that the interface is always saturated with solute, and

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that the rate of diffusion into the solvent is the limiting factor. DeVan⁵ has combined the first and second mechanisms into a single expression which is valid for intermediate cases.

Still a third mechanism, which might be considered as a special case of the first or solution rate limited process, can be postulated when the solid wall material is an alloy. Suppose one constituent of the alloy is selectively removed; i.e., its rate of removal relative to that of the other constituents is greater than would be expected based on the composition. Then it is conceivable that the over-all rate of transfer could be limited by the rate of diffusion toward the interface of the constituent in the alloy wall, since the rate of diffusion in liquids is many times greater than in solids, and thus the surface would tend to be depleted more and more as mass transfer proceeds. Figure 1 illustrates this point.

The first mechanism is not amenable to quantitative calculation at this time, due to the absence of information from which solution rate constants for the systems of interest could be estimated¹. This report is concerned, therefore, with the two diffusion mechanisms for which some reasonable estimates of the controlling diffusivities can be made, in the absence of direct experimental measurement.

Epstein^{3, 4} has presented an analysis of the rate of mass transfer for a simple two-temperature loop, for which diffusion of solute in the liquid (second mechanism) is assumed limiting. Since this treatment neglects variation in temperature and solution composition, a more general solution was obtained as outlined herein for the same basic mechanism. In addition, some

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Fig. 1. Variation in Concentration with Time for Wall Diffusion Limited Mass Transfer.

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calculations, assuming diffusion within the solid wall to be limiting (third mechanism) are described. The two diffusion mechanisms are then compared with some experimental measurements for Inconel alloy in contact with Na.

It is worthy of note that, in the analysis of the two diffusion mechanisms, the liquid leaving the cold zone is assumed to be just saturated. If supersaturation were to occur, and this is a good possibility as shown by the calculations, page 17, the rates of mass transfer would be appreciably lower than calculated herein.

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SUMMARY OF ANALYSIS

I. Liquid Diffusion Limited Mass Transfer

Consider a simple heat transfer loop:

Assumptions:

1. Isothermal between heater and cooler.
2. Heater and cooler geometrically identical diameter d and length L .
3. Heat input and removal constant per unit length.

$$\frac{dQ}{dy} = \frac{-dQ}{dy'} = \frac{Q}{L} = h\pi d(t_w - t)$$

where h is the heat transfer coefficient between tube wall and fluid;
 t_w , the wall temperature at any position; and t , the corresponding
 bulk fluid temperature.

4. Mass- and heat-transfer coefficients and fluid properties are evaluated at a mean bulk temperature:

$$\frac{t_C + t_H}{2}$$

and are therefore independent of position.

5. Effect of mass transfer on heat transfer is negligible.
6. Mass velocity G is constant.
7. Tube wall is assumed to be pure metal; e.g., Ni.
8. Solubility relationship of the form $x = S(t)$ where x is the mole fraction of solute dissolved in the fluid, S is the solubility constant.
9. The circulating fluid is assumed saturated at the tube wall:
- $$x_w = S(t_w)$$
10. Axial diffusion is negligible.
11. All of the mass picked up in the hot region is removed before leaving the cold region; i.e., no supersaturation. This is probably the least likely of the eleven assumptions.

General Solution

(See Appendix I for derivation of the following equations.)

The variation of solute concentration x in the heater with respect to distance y from the inlet and time θ is given by:

$$x_I(y, \theta) = \left[1 - \frac{1}{1 + \frac{G\theta}{\rho y}} \right] \left\{ \exp \left(-\frac{4k_L y}{Gd} \right) \left[x_I(0, \theta) - SE \right] + S \left(E + \frac{y \Delta t}{L} \right) \right\} \quad (1)$$

$$E = t_C + \frac{Q}{c\pi d \cdot L} \left(\frac{c}{h} - \frac{1}{k_L} \right)$$

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A similar expression is obtained for the cooler.

Note that the time dependence takes the form

$$\left(1 - \frac{1}{1 + \frac{G\theta}{\rho L}}\right) \text{ for } y = L \quad (2)$$

If the equilibrium time θ_e is defined as the time for this factor to reach 0.99 (i.e., 99% of the ultimate value of $x_I(L, \infty)$),

$$\theta_e \cong 100 \frac{\rho L}{G} = \frac{100 L}{V}$$

where V is the linear velocity through the loop. For a typical forced circulation loop L may be of the order of 10 ft; and V , 7 ft/sec, giving $\theta_e = 144$ sec (~ 2.5 hr). Thus a system of this type should reach steady-state operation rapidly regardless of the diffusivity of solute or its solubility. Consequently, further discussion will assume steady-state operation.

Steady-State

For θ sufficiently long (see above).

Heater

$$x_I(y) = \exp\left(-\frac{4k_L y}{Gd}\right) x_I(0) + S \left\{ \frac{y\Delta t}{L} + E \left[1 - \exp\left(-\frac{4k_L y}{Gd}\right) \right] \right\} \quad (3)$$

Cooler

$$x_{II}(y') = \exp\left(-\frac{4k_L y'}{Gd}\right) x_I(L) + S \left\{ -\frac{y\Delta t}{L} + F \left[1 - \exp\left(-\frac{4k_L y'}{Gd}\right) \right] \right\} \quad (4)$$

From equations (3) and (4):

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$$x_I(\theta) = x_{II}(L) =$$

$$\frac{S \left[\exp \left(-\frac{4k_L L}{Gd} \right) \left\{ \Delta t + E \left[1 - \exp \left(-\frac{4k_L L}{Gd} \right) \right] \right\} - \Delta t + F \left[1 - \exp \left(-\frac{4k_L L}{Gd} \right) \right] \right]}{1 - \exp \left(-\frac{8k_L L}{Gd} \right)} \quad (5)$$

The rate of mass transfer per unit cross-sectional area is:

$$G\Delta x = G \left[x_I(L) - x_I(0) \right] =$$

$$G \left[\left[\exp \left(-\frac{4k_L L}{Gd} \right) - 1 \right] x_I(0) + S \left\{ \Delta t + E \left[1 - \exp \left(-\frac{4k_L L}{Gd} \right) \right] \right\} \right] \quad (6)$$

$$E = t_C + \frac{Q}{c\pi d \cdot L} \left(\frac{c}{h} - \frac{1}{k_L} \right) \quad (7)$$

$$F = t_H - \frac{Q}{c\pi d \cdot L} \left(\frac{c}{h} - \frac{1}{k_L} \right) \quad (8)$$

Refer to page 18 for a simplification of equation (6).

II. Wall Diffusion Limited Mass Transfer

Refer to Figure 1 and Appendix II for derivations. For this case, as has been indicated, the concentration of diffusing constituent in the solid at the interface, x_w' , decreases with time from the initial value x representing the bulk concentration. As shown in Appendix II, page 40, the assumption of large solution rate constants, k and k_s in equation (b) (Appendix II) leads to a proportionality between x_w' and the liquid concentration at the interface given by equation (f):

$$x_w' = kx_w'/k_s \quad (9)$$

so that x_w' will also decrease with time, as will the mass transfer rate.

This latter relationship is expressed quantitatively by equation (j)

(Appendix II) for any value of θ :

$$R(\theta) = (k_L k / k_s) \exp(k_W^2 D_W \theta) \operatorname{erfc}(k_W \sqrt{D_W \theta}) (X - k_s x / k) \quad (10)$$

where:

$R(\theta)$ = mass transfer rate per unit surface area

k_L = liquid mass transfer coefficient

$k_W = k_L k / \rho_w D_w k_s$

ρ_w = wall density

D_w = diffusivity of solute in the wall (hot zone)

x = bulk fluid concentration

θ = time from initial contact.

For θ sufficiently long,* the result simplifies to:

$$R(\theta) = \frac{\rho_w D_w^{1/2}}{\sqrt{\pi \theta}} (X - k_s x / k) \quad (11)$$

Since

$$R(\theta) = \frac{dW(\theta)}{d(\theta)},$$

where $W(\theta)$ is the total mass transferred per unit area of hot surface

(assumed to be at constant temperature, t_H) in time θ^* :

$$W(\theta) \cong 2 \rho_w (D_w \theta / \pi)^{1/2} (X - k_s x / k) \quad (12)$$

In apply equations (10), (11), or (12), it is necessary to estimate

* It can be shown that equation (11) holds for $\theta \geq 0.05$ hours. Since the total mass transferred for $\theta \leq 0.05$ is small, equation (12) is a good approximation for any θ .

the term $k_s x/k$. x , the bulk liquid concentration, has been assumed constant in the derivations. That this is permissible is seen from two considerations:

- (a) It can be shown that the solution will quickly reach saturation, and if supersaturation is neglected, the concentration leaving the cooler will be $x = St_C$ where S is the solubility constant and t_C the liquid temperature. This is also found to be the case for solution diffusion limited mass transfer (see page 12).
- (b) While no rigorous solution of the dynamics of a closed loop with wall diffusion limiting has been obtained due to the analytical complexity, it can be shown that the concentration of bulk liquid will change very little from the saturation level, St_C , anywhere in the loop.

Accordingly, one can set $x = St_C$.

k_s/k , the ratio of two unknown solution rate constants, can be eliminated from the equations by recognition of the fact that when $t_C = t_H (\Delta t = 0)$ there should be no mass transfer; and $W(\theta) = 0$.*

From equation (12):

$$X - k_s/k St_H = 0$$

$$\therefore \frac{k_s}{k} = \frac{X}{St_H}$$

The final result for $W(\theta)$ becomes, accordingly:

$$W(\theta) \cong 2\rho_w (D_w \theta/\pi)^{1/2} X(t_H - t_C)/t_H \quad (13)$$

*There is an indication that $W(\theta) = 0$ for $\Delta t \neq 0$ (Figure 4). However, for present purposes this anomaly is of secondary importance.

Note that a constant heater temperature, t_H , has been assumed in calculation of $W(\theta)$. For this same assumption, the corresponding mass transfer for liquid diffusion limiting is:

$$W(\theta) = k_L(x_w - x)\theta = k_L S(t_H - t_C)\theta \quad (14)$$

Equations (13) and (14) illustrate the fundamental differences in the two postulates, as amplified in the Summary and Table IV.

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DISCUSSION OF THE ANALYTICAL RESULTS

I. Liquid Diffusion Limiting

For practical conditions it can be shown that equation (5), page 13, reduces to:

$$x_I(0) = x_{II}(L) \cong S(t_C + t_H)/2$$

which is above the composition corresponding to saturation at the cooler exit. Thus, it is seen that the liquid will tend toward supersaturation. If it is assumed, however, that this cannot occur:

$$x_I(0) = x_{II}(L) = St_C$$

This equation thus replaces equation (5), page 13. Also, it is apparent that equation (4) can hold only up to the point of saturation in the cooler.

Simplification

The following additional assumptions are made to simplify equation (6):

1. The heat-transfer coefficient, h , is large compared with the mass-transfer coefficient, k_L . This assumption is equivalent to neglecting the film temperature drop.
2. The increase in composition in the heater, $x_I(L) - x_I(0)$, is small compared with the driving force, $x_w - x$.
3. $4k_L/Gd$ is small enough that the exponential can be represented by a three term series expansion.

Under these assumptions equations (6) and (7) combine to give:

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$$G\Delta x \approx \frac{2L}{d} k_L S(t_H - t_C) \quad (15)$$

and the gross mass transfer becomes

$$W(\theta) \approx \frac{k_L}{2} S(t_H - t_C)\theta \quad (16)$$

Comparing the latter result with equation (14), page 16, it is seen that, as an approximation, the amount of transfer is reduced by a factor of two when the temperature of the heater varies linearly from t_C to t_H , as might be expected.

It is further evident from equation (16) that, for liquid diffusion limited mass transfer, the amount should vary approximately linearly with:

- (a) mass transfer coefficient
- (b) equilibrium solubility
- (c) temperature difference (assuming t_H constant)
- (d) time.

Furthermore, since k_L varies approximately as $N_{Re}^{0.83*}$, W should vary as the 0.83 power of the velocity. According to Treybal*, k_L and thus W should vary as $D_L^{0.67}$ where D_L is the diffusivity of solute in liquid.

The dependence of mass transfer on temperature in the hot zone is contained in the expression for k_L , the temperature sensitive terms being D_L and μ neglecting density variations. k_L varies as $D_L^{0.67}/\mu^{0.5}$, according to Treybal. As an approximation for D_L , the Stokes-Einstein equation (6)

*Treybal, Mass Transfer Operations, p 51 gives:

$k_L d/D_L = 0.023 N_{Re}^{0.83} N_{Sc}^{0.33}$, where $N_{Re} = dG/\mu$; $N_{Sc} = \mu/\rho D_L$
(units of $k_L = \text{lb-mole/hr-ft}^2 - (\text{lb-mole/ft}^3)$).

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can be used:

$$D_L = \frac{RT}{N} \frac{1}{6\pi\mu r}$$

$$\therefore k_L \text{ varies approximately as } (t_H + 460)^{0.67}/\mu^{1.17}.$$

From a log-log plot, it is found that k_L , and thus $W(\theta)$, will vary as $t_H^{1.3}$ for this case.

Note that the geometrical dependence is expressed by

$$N_{Re}^{0.83}/d \sim \frac{1}{d^{0.17}}.$$

The conclusions reached in this section are in substantial agreement with those of Epstein, and are summarized in Table IV, page 33.

II. Wall Diffusion Limiting

The basic equation is:

$$W(\theta) = 2\rho_w(D_w\theta/\pi)^{1/2} (X - k_s St_C/k) \quad (12')$$

The dependence of $W(\theta)$ on temperature difference for this case is directly related to the value of k_s/k , for if this term is zero it is apparent that there can be no dependence on Δt . On the other hand, if $k_s/k = X/St_H$ as assumed on page 15, $W(\theta)$ becomes directly proportional to Δt (equation (13)).

It is interesting to note that equation (12') does not rule out the possibility that $W(\theta)$ will be zero for $\Delta t \neq 0$, for if t_C' is the cold zone temperature for which no mass transfer occurs ($t_C' \neq t_H$) then

$$W(\theta) = 2\rho_w(D_w\theta/\pi)^{1/2} X(t_C' - t_C)/t_C'$$

the amount of mass transfer will depend on the temperature difference

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$t_C' - t_C$. Since t_C' may depend on the manner in which the temperatures are varied, the above equation has no general applicability, and equation (13) will be employed in the discussion and calculations to follow:

$$W(\theta) \cong 2\rho_w (D_w \theta/\pi)^{1/2} X(t_H - t_C)/t_H \quad (13)$$

Note that, for constant hot zone temperature, the amount of mass transfer will vary directly with Δt ; and with the initial (bulk) wall composition, X .

On the other hand, for constant Δt , W will vary as $D_w^{1/2}/t_H$. D_w can usually be expressed by an Arrhenius expression of the form:

$$D_w = A \exp(-q/RT) \quad .$$

As indicated on page 27, q , the activation energy for solid diffusion of Ni, may be between 28 and 67 k-cal/g-mol. From a log plot, $W(\theta)$ is found to vary approximately as $t_H^{3.5}$ for $q = 28$ k-cal, and t_H^{10} for $q = 67$ k-cal.

Note that $W(\theta)$ as expressed by equation (13) is independent of the mass velocity, G (or N_{Re}) of the equilibrium solubility, S , and the system geometry. These conclusions are summarized in Table IV.

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CALCULATIONS

I. Liquid Diffusion Limiting

It is desired to estimate the amount of mass transfer for conditions approximating forced-convection corrosion loop operation. Equations (6) and (7), page 13, will be utilized to compute $G\Delta x$ for the following typical operating conditions.*

Loop Type: "Figure eight" sodium pump loop, considering the economizer annulus as the heater.

$$G = 1.33 \times 10^6 \text{ lb/hr-ft}^2$$

$$N_{Re} = 31,000 \text{ (equivalent)}$$

$$d = 0.01 \text{ ft (equivalent)}$$

$$L = 6.3 \text{ ft}$$

Cross-sectional area

$$= 7.36 \times 10^{-4} \text{ ft}^2$$

$$Q = 7.63 \times 10^4 \text{ Btu/hr}$$

$$t_C = 1200^\circ\text{F}$$

$$t_H = 1460^\circ\text{F}$$

Estimate of k_L

One estimate is obtained from the Treybal Equation (footnote, page 18):

$$k_L d / D_L = 0.023 N_{Re}^{0.83} N_{Sc}^{0.33}$$

*Unless otherwise stated, all of the operating conditions and data used herein have been obtained as personal communication from J. H. DeVan, Dynamic Corrosion Group, ORNL Metallurgy Division; or from the ANP Quar. Prog. Rpts.

In order to estimate D_L , the Stokes-Einstein Equation is employed, which at an average heater temperature of 1375°F gives

$$D_L = 1.32 \times 10^{-3} \text{ ft}^2/\text{hr}^*$$

Also

$$N_{Sc} \text{ at } 1375^\circ\text{F} = 7$$

$$\therefore k_L = 31 \text{ mol/hr-ft}^2 (\text{mol/ft}^3) = 3800 \text{ lb/hr-ft}^2\text{-mol ft}$$

Another estimate of k_L is obtained from the Martinelli-Boelter Equation (7):

$$\frac{k_L/G}{f/2} = \frac{1/\Delta c_m}{5 \sqrt{\frac{f}{2}} \left[N_{Sc} + \ln(1 + 5N_{Sc}) \right] + \frac{1}{2} \ln \left(\frac{N_{Re}}{60} \sqrt{\frac{f}{2}} \right)}$$

Using $f = 0.046/N_{Re}^{0.2}$ (Blasius Equation) and $1/\Delta c_m \cong 1$:

$$k_L = 1280 \text{ lb/hr-ft}^2$$

Estimation of h

The Martinelli Equation (8) gives $hd/k = 8.5$ for $N_{Pr} = 0.004$.

Since

$$k = 33 \text{ Btu/hr-ft}^2 (^\circ\text{F/ft}) \text{ for Na}$$

$$\therefore h = 28,000 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F}$$

Using the higher value of $k_L = 3800$, equations (6) and (7) give

$$G\Delta x = 1.52 \times 10^8 \text{ S lb/hr-ft}^2$$

*Note that this is in agreement with the self-diffusion coefficient of Na, reported to be $1.21 \times 10^{-3} \text{ ft}^2/\text{hr}$ at the same temperature (J. Chem. Phys., 23, 1951).

S, the solubility coefficient, is taken to be 2.0×10^{-8} for concentrations in weight fraction, based on data of Rodgers* and assuming applicability in the range from 1200°F to 1600°F.

$$\therefore G\Delta x = 3 \text{ lb/hr-ft}^2, \text{ and}$$

$$W(\theta) = 3 \times 7.36 \times 10^{-4} (\theta) = 2.2 \times 10^{-3} (\theta) \text{ lb.}$$

For $(\theta) = 1000 \text{ hr}$, the amount of mass transfer predicted is 2.2 lb (1000 g).

Using the lower value of $k_L = 1280$, equations (6) and (7) give

$$G\Delta x = 1.28 \text{ lb/hr-ft}^2$$

$$\therefore W(\theta) = 1.28 \times 7.36 \times 10^{-4} (\theta) = 9.4 \times 10^{-4} (\theta) \text{ lb.}$$

For $(\theta) = 1000 \text{ hr}$, the amount of mass transfer predicted is 0.94 lb (420 g).

Since the observed mass transfer accumulation under the conditions assumed above is about 13 grams, it is seen that the assumption of liquid diffusion limiting overestimates the amount of mass transfer by a factor of 30 to 80, depending on the value of k_L used. It is thus apparent that this mechanism is not a satisfactory one.

Equation (4) may be employed to estimate the point at which deposition would begin to occur in the cooler, that is the point at which $x_{II} = St_C$. Thus, for $N_{Re} = 100,000$, it is found that the liquid should begin to deposit crystals at $y'/L = 0.65$; and that saturation would be reached at $y'/L = 0.85$ (i.e., 65% and 85% of the distance through the cooler). Actually, it is observed in Na pump loops that deposition begins at $y'/L = 0.25$. Of course, the geometry of the pump loop is far from the idealized case assumed, so it is probably not valid to draw conclusions from the apparent discrepancy.

* Rodgers, S., MSA TR-27, June 30, 1954, gives solubility data for Ni in Na range 400 to 1000°F.

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II. Wall Diffusion Limiting

Estimation of Diffusivity D_w for Ni

In deriving equation (13) for $W(\theta)$ it was assumed that Fick's law of diffusion could be applied; and that D_w , the solid wall diffusivity, was a constant. That these are not rigorous assumptions is apparent from a review of diffusion theory and data in the literature, and are employed here as a first, albeit poor, approximation. In particular, the contribution of grain boundary diffusion is neglected.

A brief review of the literature on diffusion of Ni was made, and the pertinent data are summarized in Table I and Figure 2. Note that for diffusion in Fe, D_w varies only slightly as the Ni concentration is reduced from ~100% to 80%. It will be assumed that this is true also with Inconel, whose composition is about 78% Ni, so that the data for very pure Ni are taken as valid in this alloy and the variation in D_w with composition neglected. Three extrapolations from the data are made, Figure 2, corresponding to activation energies of 28, 50, and 67 k-cal/g-mole to represent what is believed to be maximum limits for D_w . Table II is a tabulation of the extrapolated values for D_w in the temperature range of interest.

Table III summarizes the calculated results for the amount of mass transfer, W (g/ft² hot surface), as a function of the principal variables, employing equation (13):

$$W(\theta) = 2\rho_w(D_w\theta/\pi)^{1/2} X(t_H - t_C)/t_H$$

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TABLE I

SUMMARY OF DIFFUSION DATA FOR NICKEL

Wt. % Ni	Alloy Material	D_w cm ² /sec	t, °C	q, k-cal/g-mole	Ref.
99.99	Fe	19×10^{-10}	1300	58*	9
80.00	Fe	18.5×10^{-10}	1300	61*	9
20.00	Fe	2.5×10^{-10}	1300	66*	9
0.01	Fe	1.5×10^{-10}	1300	68*	9
99.99	Cu	1.6×10^{-10}	1052		10,11
99.99	Cu	1.0×10^{-10}	1052		11
99.99	Au	3.7×10^{-11}	900		11
99.999+	Self Diffusion	2.4×10^{-13}	878	67	12
99.999+	"	6.7×10^{-13}	905	67	12
99.999+	"	1.9×10^{-12}	965	67	12
99.999+	"	8.8×10^{-12}	1035	67	12
99.999+	"	7.3×10^{-11}	1163	67	12
99.999+	"	3.7×10^{-10}	1240	67	12

* Calculated values

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Fig. 2. Summary of Data for Diffusion of Very Pure Nickel.

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TABLE II

EXTRAPOLATED DIFFUSIVITIES			
t, °F	D_w cm ² /sec		
	q = 67 k-cal (Self Diffusion)	q = 50 k-cal	q = 28 k-cal
1200	3.0×10^{-16}	2.0×10^{-14}	1.4×10^{-12}
1300	2.0×10^{-15}	1.0×10^{-13}	3.0×10^{-12}
1400	1.3×10^{-14}	4.0×10^{-13}	7.0×10^{-12}
1500	6.0×10^{-14}	1.4×10^{-12}	1.4×10^{-11}
1600	3.0×10^{-13}	5.0×10^{-12}	2.6×10^{-11}

TABLE III

SUMMARY OF CALCULATIONS FOR WALL DIFFUSION LIMITED MASS TRANSFER

$\theta = 1000$ hr

Hot Zone Temp., t_H (°F)	Cold Zone Temp., t_C (°F)	Temp. Diff., Δt (°F)	W (g/ft ² hot surface)		
			q = 67	50,	28 k-cal/g-mole
1500	1500	0	0	0	0
1500	1350	150	0.4	2.0	6.2
1500	1200	300	0.8	4.0	12.4
1500	1100	400	1.1	5.3	16.5
1200	900	300	0.0	0.5	4.0
1300	1000	300	0.2	1.1	5.7
1400	1100	300	0.4	2.1	8.8
1500	1200	300	0.8	4.0	12.4
1600	1300	300	1.9	7.4	17.0

Numerically: $\rho_w = 530 \text{ lb/ft}^3$
 $X = 0.78$
 $\theta = 1000 \text{ hr}$

Figures 3 and 4 compare the calculations for wall diffusion limited mass transfer with some forced-convection corrosion loop data obtained by the Dynamic Corrosion Group of the ORNL Metallurgy Division¹³. An effective hot zone area of 2 ft^2 was used in the calculations as typical of these loops. The extrapolations of D_w as given in Figure 2 and Table II were used to establish the range of estimation given by the top and bottom solid lines in Figures 3 and 4.

Note that order of magnitude agreement is found at the upper temperatures (Figure 3) for $q = 28$ and 50 k-cal/g-mole but that the observed mass transfer decreases more rapidly with temperature than calculated. For $q = 67 \text{ k-cal/g-mole}$ as obtained for self-diffusion of Ni, the calculations underestimate W , although the temperature dependence is more nearly in agreement with data.

It should be pointed out that the calculations have assumed only Ni to be transferring, whereas some Cr also transfers. This would have the effect of decreasing the rate of depletion of the Ni concentration at the wall surface and thus tend to make the calculations low as compared to the measurements. Consequently, it is felt that the correct activation energy probably lies between 50 and 67 k-cal/g-mole , and that the low estimates may be a result of neglecting Cr diffusion.

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Fig. 3. Comparison of Calculations for Wall Diffusion Limited Mass Transfer With Data From Corrosion Loops.

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Fig. 4. Comparison of Calculations for Wall Diffusion Limited Mass Transfer With Corrosion Loop Data .

SUMMARY

Comparison of the Three Mechanisms with Data

As discussed in the Introduction, three controlling mechanisms have been postulated to explain observed mass transfer rate in liquid metal - solid alloy systems:

- (1) Solution rate.
- (2) Liquid diffusion rate.
- (3) Solid wall diffusion rate.

In order to compare these, Table IV is included, in which the calculations presented in this report are used for mechanisms (2) and (3), and some qualitative remarks of Epstein^{3, 4} for (1).

It is apparent that the only variable whose predicted influence is the same for the three mechanisms is Δt , and indeed the experimental evidence indicates a linear relationship as predicted. With respect to gross mass transfer, W , it is also apparent that the liquid and wall diffusion mechanisms predict vastly different amounts. The wide spread in diffusivity data for the range of W makes it difficult to draw any quantitative conclusions. However, there is better agreement with data using either extreme of W as calculated for wall diffusion limiting than for liquid diffusion limiting. Furthermore, the temperature dependence predicted by the wall diffusion mechanism is in fairly good agreement with observation at the higher temperatures. Surprisingly, the liquid diffusion mechanism predicts the observed dependence of flow (N_{Re}) more accurately than do the other two. The

TABLE IV

SUMMARY OF CALCULATIONS AND COMPARISON WITH DATA FOR
THREE POSTULATED MECHANISMS - MASS TRANSFER IN Na-INCONEL SYSTEMS

Variable	Calculations			Data (Forced- Circulation Corrosion Loop Program)
	(1) Solution Rate Limiting (Epstein)	(2) Liquid Diffusion Limiting	(3) Wall Diffusion Limiting	
W: gross transfer				
(a) $\theta = 1000$ hr $t_H = 1500^\circ\text{F}$ $\Delta t = 300^\circ\text{F}$	No estimate available	400 - 1000 grams	2 - 25 grams	13 grams
(b) $\theta = 1000$ hr $t_H = 1200^\circ\text{F}$ $\Delta t = 300^\circ\text{F}$	No estimate available	300 - 700 grams	0 - 8 grams	< 0.5 gram
t_H : hot zone temperature	Depends on value of the activation energy for the solution mechanism; most likely this is large and W is strongly dependent on t_H	$W \approx t_H^{1.3}$	$W \approx t_H^4$ to t_H^{10} ($28 < q < 67$ k-cal)	t_H^6 ($t_H > 1350$) t_H^{30} ($t_H \leq 1350$)
Δt : temp. difference	$W \approx \Delta t$	$W \approx \Delta t$	$W \approx \Delta t$ (t_H constant)	$W \approx (\Delta t - 60)$
N_{Re} : fluid flow	Independent	$W \approx N_{Re}^{0.83}$	Independent	Definite dependence
L,d: geometry	Independent	$W \approx d^{-0.17}$	Independent	Some dependence noted
θ : time	$W \approx \theta^n$ ($n < 1$)	$W \approx \theta$	$W \approx \theta^{0.5}$	$W \approx \theta^{0.67}$ (approx.) ($\theta \leq 2000$ hr)
s: solubility of Ni in Na	-	$W \approx s$	Independent	Dependence not known
X: bulk solid concentration	-	Independent	$W \approx X$	W decreases as Ni content of alloy decreases

observed time dependence is seen to lie between that predicted by the two diffusion mechanisms. Note that none of the mechanisms would indicate a significant geometrical dependence.

There is some evidence that decreasing the bulk concentration X reduces the mass transfer nearly proportionally (e.g., Incoloy vs Inconel), which is in agreement with the wall diffusion mechanism.

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CONCLUSIONS

It is apparent from Table IV that no one limiting mechanism of the three postulated for mass transfer corrosion in liquid metals is adequate to explain the observed results. Not much can be concluded as to the validity of the solution rate mechanism in the absence of information on the solution rate constant. Of the two diffusion mechanisms, wall diffusion limiting appears to give the best agreement with gross mass transfer amounts. On the other hand, this mechanism fails to predict the observed flow dependence; and also predicts greater than observed fall-off in mass transfer rate with time.

The assumption that the liquid leaves the cooler saturated is possibly the weakest one in the analysis. Supersaturation may decrease the average driving force for liquid diffusion in the hot zone to the point where this mechanism could be controlling. For this reason, it is recommended that work be continued to explore the mechanism of deposition in the cooler and to determine the dynamic composition of circulating liquid in a forced-convection corrosion loop.*

One important piece of information in connection with the wall diffusion hypothesis would be the determination of surface concentration of Inconel after prolonged exposure to Na or NaK under Δt conditions.

Because of the similarity of the solution rate and wall diffusion mechanisms with respect to the predicted influence of the important variables (Table IV), it is not possible at this time to delineate the independent

* Such a program was initiated in ARED Experimental Engineering, June 1956, but soon terminated.

contributions of each. Solution rate data are thus badly needed.

In the over-all analysis, it may be necessary to combine all three mechanisms; as for example equation (d), Appendix II, such that none is unequivocally controlling.

APPENDIX I

Derivation of Equations for Liquid Diffusion Limiting

Consider differential volume element at any point in header or cooler:

x_w is the wall concentration, assumed saturated at t_w . k_L is the mass transfer coefficient between tube wall and fluid.

From a material balance, equating net input to accumulation:

$$\frac{4k_L}{d} [x_w(y, \theta) - x(y, \theta)] = G \frac{\partial x(y, \theta)}{\partial y} + \rho \frac{\partial x(y, \theta)}{\partial \theta} \quad (a)$$

θ = time, ρ = density

The complete integral of this equation by the method of LaGrange is:

$$x(y, \theta) \exp\left(\frac{4k_L y}{Gd}\right) - \left[\int \frac{4k_L S(A + By)}{Gd} \exp\left(\frac{4k_L y}{Gd}\right) dy \right] = \alpha(Gd - \rho y) + \beta \quad (b)$$

where $B = \frac{\Delta t}{L}$, $A = t_C + \frac{q}{h\pi d \cdot L}$ for the heater;

$B = \frac{-\Delta t}{L}$, $A = t_H - \frac{q}{h\pi d \cdot L}$ for the cooler

α and β are constants of integration.

Considering the heating and cooling sections separately:

I. Heater ($0 \leq y \leq L$)

Boundary conditions:

$$\begin{aligned} x_I(y, 0) &= 0 \\ x_I(0, \theta) &= \text{constant to be determined} \end{aligned} \quad (c)$$

Substituting (c) in (b), performing the indicated integration, solving for α and β and substituting back in equation (3):

$$x_I(y, \theta) = \left[1 - \frac{1}{1 + \frac{G\theta}{\rho y}} \right] \left\{ \exp\left(\frac{-4k_L y}{Gd}\right) \left[x_I(0, \theta) - SE \right] + S(E + \Delta t \frac{y}{L}) \right\} \quad (d)$$

where

$$E = t_C + \frac{Q}{c\pi d \cdot L} \left(\frac{c}{h} - \frac{1}{k_L} \right) \quad (e)$$

c is the specific heat of the fluid.

II. Cooler ($0 \leq y' \leq L$)

Boundary conditions:

$$\begin{aligned} x_{II}(y', 0) &= 0 \\ x_{II}(0, \theta) &= x_I(L, \theta) \end{aligned} \quad (f)$$

From (f) and (b):

$$x_{II}(y', \theta) = \left[1 - \frac{1}{1 + \frac{G\theta}{\rho y'}} \right] \left\{ \exp\left(\frac{-4k_L y'}{Gd}\right) \left[x_I(L, \theta) - SF \right] + S(F - \Delta t \frac{y'}{L}) \right\} \quad (g)$$

where

$$F = t_H - \frac{Q}{c\pi d \cdot L} \left(\frac{c}{h} - \frac{1}{k_L} \right)$$

Combining equations (e) and (g):

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$$x_I(0, \theta) = x_{II}(L, \theta) =$$

$$\frac{S \left[\exp \left(-\frac{4k_L L}{Gd} \right) \left\{ E \left[1 - \exp \left(-\frac{4k_L L}{Gd} \right) \right] + \Delta t - \frac{F}{1 - \frac{1}{1 + \frac{G\theta}{\rho L}}} \right\} + \frac{F - \Delta t}{1 - \frac{1}{1 + \frac{G\theta}{\rho L}}} \right]}{\left(1 - \frac{1}{1 + \frac{G\theta}{\rho L}} \right)^2 - \exp \left(-\frac{8k_L L}{Gd} \right)} \quad (h)$$

Equations (d), (g), and (h) are the required solution including time dependence.

Steady-State Solution

Setting $\theta = \infty$ in equations (d), (g), and (h)

I. Heater

$$x_I(y, \infty) = \exp \left(-\frac{4k_L y}{Gd} \right) x_I(0, \infty) + S \left\{ \frac{y\Delta t}{L} + E \left[1 - \exp \left(-\frac{4k_L y}{Gd} \right) \right] \right\} \quad (i)$$

II. Cooler

$$x_{II}(y', \infty) = \exp \left(-\frac{4k_L y'}{Gd} \right) x_{II}(L, \infty) + S \left\{ -\frac{y'\Delta t}{L} + F \left[1 - \exp \left(-\frac{4k_L y'}{Gd} \right) \right] \right\} \quad (j)$$

$$x_I(0, \infty) = x_{II}(L, \infty) =$$

$$\frac{S \left[\exp \left(-\frac{4k_L L}{Gd} \right) \left\{ \Delta t + E \left[1 - \exp \left(-\frac{4k_L L}{Gd} \right) \right] \right\} - \Delta t + F \left[1 - \exp \left(-\frac{4k_L L}{Gd} \right) \right] \right]}{1 - \exp \left(-\frac{8k_L L}{Gd} \right)} \quad (k)$$

APPENDIX II

Derivation of Equations for Wall Diffusion Limited Mass Transfer

For the general case, the three mechanisms, page 32, must operate simultaneously. It is true, therefore that: $R_w = R_s = R_L$; where R_w and R_L represent the diffusion rates, per unit area, of solute through solid wall and liquid, respectively; and R_s is the rate of solution at the interface. Furthermore, assuming Fick's law and constant diffusivity:

$$R_w = D_w \rho_w \left. \frac{\partial x_w'}{\partial l} \right|_{l=0} \quad (a)$$

where D_w is the diffusivity in solid; ρ_w , the solid density; x_w' , the wall concentration; and l , the distance from the surface. The concentration gradient in (a) is evaluated at the surface, $l = 0$.

Also, as given by DeVan⁵ and others:

$$R_s = kx_w' - k_s x_w \quad (b)$$

where x_w is the concentration in the solution at the interface; k and k_s are solution rate constants; and

$$R_L = k_L (x_w - x) \quad (c)$$

where x is the bulk liquid concentration. Combining (a), (b), and (c):

$$D_w \rho_w \left. \frac{\partial x_w'}{\partial l} \right|_{l=0} = kx_w' - k_s x_w = k_L (x_w - x) \quad (d)$$

Therefore, from the pair of equations on the right:

$$x_w = \frac{kx_w' + k_L x}{k_L + k_s} \quad (e)$$

Let it be postulated that the solution rate constants, k and k_s , are large compared with the liquid mass transfer coefficient, k_L , so that equation (e) becomes:

$$x_w = \frac{kx'_w}{k_s} \quad (f)$$

$$\therefore D_w \rho_w \left. \frac{\partial x'_w}{\partial l} \right|_{l=0} = k_L \left(\frac{k}{k_s} x'_w - x \right) \quad (g)$$

Equation (g) is, therefore, a necessary boundary condition for solution of the differential equation

$$D_w \frac{\partial^2 x'_w}{\partial l^2} + \frac{\partial x'_w}{\partial \theta} = 0 \quad (h)$$

describing diffusion in the solid wall.

When x is constant, the solution of (h) and (g) can be deduced from a derivation of Carslaw¹⁴ for an analogous problem in heat flow, to give:

$$\frac{x'_w - k_s x/k}{X - k_s x/k} = \exp(K_w^2 D_w \theta) \operatorname{erfc}(K_w \sqrt{D_w \theta}) \quad (i)$$

where X is the initial (bulk) solid concentration, and $K_w = k_L k / \rho_w D_w k_s$.

Therefore, from (c) and (f) for the mass transfer rate, $R(\theta)$:

$$R(\theta) = (k_L k / k_s) \exp(K_w^2 \sqrt{D_w \theta}) (X - k_s x/k) \quad (j)$$

Equation (j) gives the mass transfer rate for $\theta \geq 0$.

For θ sufficiently long, equation (i) reduces to:

$$\frac{x'_w - k_s x/k}{X - k_s x/k} = \frac{\rho_w D_w^{1/2} k_s}{k_L k \sqrt{\pi \theta}} \quad (k)$$

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and the rate becomes:

$$R(\theta) = \frac{\rho_w D_w^{1/2}}{\sqrt{\pi \theta}} \left[X - k_s x/k \right] \quad (1)$$

$\theta > 0$

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