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MATHEMATICAL MODEL FOR HEAT OR MASS TRANSFER AT THE BUBBLE-STIRRED INTERFACE OF TWO IMMISCIBLE LIQUIDS

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Abstract—A mathematical model is proposed to describe the mechanism of heat or mass transfer at the bubble-stirred interface of two immiscible liquids. On the assumption of unsteady-state condition (diffusion) between the arrival of successive bubbles, an expression is derived for the overall transfer coefficient.

One practical application of the proposed model is the interpretation of slag–metal interactions in an open-hearth furnace.

NOMENCLATURE

A_B,	cross-sectional area of the bath;
A_b,	surface area swept by one bubble;
C,	concentration;
C_1, C_2,	instantaneous concentrations in phases 1 and 2 respectively;
$\bar{C}_1$, $\bar{C}_2$,	uniform bulk concentrations in phases 1 and 2 respectively;
C_{AW},	concentration of component W in phase A;
C_{BW},	concentration of component W in phase B;
C^*,	equilibrium concentration;
C_p,	specific heat;
D_1, D_2,	diffusivities in phases 1 and 2 respectively;
D_W,	diffusivity of component W;
h_{DWA}, h_{DWB},	mass-transfer coefficient relating to component W, in phases A and B respectively;
h,	overall heat-transfer coefficient;
h_D,	overall mass-transfer coefficient;
k_1, k_2,	thermal conductivity in phases 1 and 2 respectively;
K,	partition coefficient defined by equation (3A);
$\bar{N}_W$,	mean mass flux of component W;
$N(t)$,	instantaneous mass flux;
$\bar{N}_{t_e}$,	mean value of mass flux over time interval t_e;
$Q(t)$,	instantaneous heat flux;
$\bar{Q}_{t_e}$,	mean value of heat flux over time t_e equation (11);
T_1, T_2,	instantaneous temperatures in phases 1 and 2 respectively;
$\bar{T}_1$, $\bar{T}_2$,	uniform bulk temperatures in phases 1 and 2 respectively;
t,	time;
t_e,	time interval between the arrival of consecutive bubbles;
x,	distance from the interface;
κ_1, κ_2,	thermal diffusivities in phases 1 and 2.

INTRODUCTION

THE TRANSFER of heat or mass across the interface of two immiscible liquids has attracted a large number of investigations, both theoretical and practical. This paper is restricted to the discussion of a rather specialized case, namely transport across the interface of two immiscible liquids periodically disturbed by gas bubbles rising in a direction perpendicular to the plane of separation, a problem on which no comprehensive theoretical treatment is available at present.

The system to be discussed is of practical interest for the interpretation of slag-metal interactions in an open-hearth furnace.

MODEL FOR HEAT TRANSFER

Temperatures are known more accurately than concentrations in open-hearth practice, therefore derivation of the model will be given for heat transfer. It can be shown however, that similar considerations apply for mass transfer. (For final results see Appendix).

Consider two immiscible liquid phases, 1(slag) and 2(metal), extending from $x = 0_{(+)}$ to $x = +\infty$, and $x = 0_{(-)}$ to $x = -\infty$, respectively.

Assign values to the respective uniform bulk temperatures, $\bar{T}_1$, $\bar{T}_2$, thermal diffusivities κ_1, κ_2 and thermal conductivities k_1, k_2 of the two phases.

Finally consider that the instantaneous temperature near the surface is given by T_1 and T_2 for the two phases respectively.

The proposed model suggests that when a large bubble crosses the interface it destroys instantaneously the temperature gradients on both sides of the $x = 0$ plane. Thus at this instant $T_1 = \bar{T}_1$ for $x > 0$ and $T_2 = \bar{T}_2$ for $x < 0$. Thereafter there exists unsteady state conduction of heat from phase 1 to phase 2 with a corresponding re-establishment of temperature gradients, until the arrival of the next bubble, when the whole cycle starts again.

The problem is to calculate point values and time-averaged values of the heat flux crossing the $x = 0$ plane during the time interval between the arrival of successive bubbles.

MATHEMATICAL FORMULATION

It can be shown [1] that the system described above can be represented by two simultaneous partial differential equations:

$$\frac{\partial^2 T_1}{\partial x^2} = \frac{1}{\kappa_1}\frac{\partial T_1}{\partial t}, \quad \text{for } x > 0 \tag{1}$$

and

$$\frac{\partial^2 T_2}{\partial x^2} = \frac{1}{\kappa_2}\frac{\partial T_2}{\partial t}, \quad \text{for } x < 0. \tag{2}$$

The initial and boundary conditions are as follows:

$$T_1 = T_2, \text{ at } x = 0,\ t > 0 \tag{3}$$

$$k_1 \frac{\partial T_1}{\partial x} = k_2 \frac{\partial T_2}{\partial x}, \quad \text{at } x = 0,\ t > 0 \tag{4}$$

$$T_1 = \bar{T}_1, \text{ at } t = 0 \quad \text{for } x > 0 \tag{5}$$

$$T_2 = \bar{T}_2, \text{ at } t = 0 \quad \text{for } x < 0. \tag{6}$$

The solution of equations (1) and (2) is given by Carslaw and Jaeger [1] as,

$$T_1 - \bar{T}_2 = \frac{k_1\kappa_1^{-1/2}(\bar{T}_1 - \bar{T}_2)}{k_1\kappa_1^{-1/2} + k_2\kappa_2^{-1/2}} \left[1 + \frac{k_2\kappa_2^{-1/2}}{k_1\kappa_1^{-1/2}} \operatorname{erf} \frac{x}{2\sqrt{(\kappa_1 t)}}\right] \tag{7}$$

and

$$T_2 - \bar{T}_2 = \frac{k_1\kappa_1^{-1/2}(\bar{T}_1 - \bar{T}_2)}{k_1\kappa_1^{-1/2} + k_2\kappa_2^{-1/2}} \operatorname{erfc} \frac{|x|}{2\sqrt{(\kappa_2 t)}}. \tag{8}$$

Thus the instantaneous heat flux across the interface Q_t is given by:

$$Q_{(t)} = k_1 \left(\frac{\partial T_1}{\partial x}\right)_{x=0} \equiv k_2 \left(\frac{\partial T_2}{\partial x}\right)_{x=0}. \tag{9}$$

Thus by differentiating (7) and substituting $x = 0$ we obtain

$$Q_{(t)} = \frac{k_1(\bar{T}_1 - \bar{T}_2)}{\sqrt{(\pi\kappa_1 t)}} \left(\frac{k_2\kappa_2^{-1/2}}{k_1\kappa_1^{-1/2} + k_2\kappa_2^{-1/2}}\right). \tag{10}$$

A time averaged value of the flux Q_{t_e} for the period t_e, (i.e. time interval between the arrival of two successive bubbles) is given by:

$$\bar{Q}_{t_e} = \frac{1}{t_e}\int_0^{t_e} Q(t)\,\mathrm{d}t \tag{11}$$

that is,

$$\bar{Q}_{t_e} = 2\frac{k_1(\bar{T}_1 - \bar{T}_2)}{\sqrt{(\pi\kappa_1 t_e)}} \left(\frac{k_2\kappa_2^{-1/2}}{k_1\kappa_1^{-1/2} + k_2\kappa_2^{-1/2}}\right) \tag{12}$$

hence the overall heat transfer coefficient h is given by:

$$h \equiv \frac{\bar{Q}_{t_e}}{\bar{T}_1 - \bar{T}_2} = \frac{2k_1}{\sqrt{(\pi\kappa_1 t_e)}} \frac{k_2\kappa_2^{-1/2}}{k_1\kappa_1^{-1/2} + k_2\kappa_2^{-1/2}}. \tag{13}$$

It can be seen that if $k_2\kappa_2^{-1/2} \gg k_1\kappa_1^{-1/2}$ equation (13) reduces to

$$h = \frac{2k_1}{\sqrt{(\pi \kappa_1 t_e)}} \tag{14}$$

i.e. there is "negligible resistance to transfer in phase 2". Furthermore, it can be shown that for

$$k_1 \kappa_1 \gg k_2 \kappa_2,$$

$$h = \frac{2k_2}{\sqrt{(\pi \kappa_2 t_e)}} \tag{14b}$$

i.e. there is "negligible resistance in phase 1". The expressions given in (14) and (14b) are analogous to that given by the penetration theory [2] for mass transfer. Moreover, it is noted that the transfer coefficient shows a proportionality with the bubble frequency $h \propto 1/\sqrt{(t_e)}$ which is consistent with observations that higher rates of transfer exist during more violent bubbling.

PRACTICAL APPLICATION OF THE MODEL: INTERPRETATION OF SLAG–METAL INTERACTIONS IN AN OPEN-HEARTH FURNACE

(a) *General description of the process*

The mathematical model derived in the preceding sections can be used for the interpretation of slag–metal interactions in an open-hearth furnace.

The function of an open-hearth furnace is to convert various types of iron into steel of given composition and quality. The process depends on the oxidation of impurities such as carbon, silicon, manganese. During the so-called refining period when the ferrous charge is melted and is covered with molten slag, the rate of these oxidation processes has been stated to depend on the rate at which oxygen can diffuse through the slag–metal interface [3, 4, 5]. Similarly, the rate of heat flow from slag to metal during the refining period—required to offset heat losses through the hearth pan—is said to be limited by a resistance at the slag metal interface [6].

The product of reaction between carbon and oxygen in the steel bath is gaseous carbon monoxide. It has been generally assumed [3, 4, 5] that the metal phase is super-saturated with carbon monoxide and that CO bubbles form at nucleation points at the bottom of the bath. These bubbles were found by observation [3, 7] to grow to a considerable size before passing through the interface prior to leaving the system via the slag surface. The vigorous stirring afforded by these bubbles in both phases is thought to account for the relatively high rate of transfer that exists at the slag–metal interface.

The physical picture of the open-hearth process discussed above is consistent with the assumptions made for derivation of the model for transfer at the bubble stirred interface of two immiscible liquids.

Moreover, the following brief review of previous work on the kinetics of slag–metal interactions will show that the model proposed is a more realistic representation of the process than those available up to the present.

(b) *Previous work*

Previous workers [3, 4, 5] when interpreting the kinetics of slag–metal interactions have either restricted themselves to calculating wholly empirical transfer coefficients defined as

$$h_{DW} = \frac{\bar{N}_W}{C_W - C_W} \tag{15}$$

or have postulated that the transfer coefficient h_D is of the form of

$$h_{DW} = \frac{D_W}{\delta} \tag{15b}$$

where δ is an arbitrarily chosen boundary layer thickness, selected to fit empirical results.

All these authors assumed:

(a) steady state conditions,
(b) that the resistances to transfer were linearly additive (i.e. that conditions of the two-film theory applied).

$$\begin{aligned} \bar{N}_W &= h_{D_{AW}}(C_{AW} - C^*_{AW}) \\ &= h_{D_{BW}}(C^*_{BW} - C_{BW}). \end{aligned} \tag{16}$$

This previous work, although it helped the understanding of a very complicated system, can be criticized on grounds that—

(a) the approach does not allow predictions to be made for different operating conditions (effect of different stirring rates by bubbles on h_D),

(b) a model assuming steady-state transfer through a thin uninterrupted layer hardly agrees with observations which would indicate an interface that is periodically disrupted by the bubbles.

The model presented in the preceding sections gives a more realistic picture and also allows predictions to be made as to the magnitude of the transfer coefficients and their dependence on certain process variables.

(c) *Calculation of the heat-transfer coefficient*

Equation (13) derived previously, can be used for calculating h the overall heat-transfer coefficient at the slag–metal interface. Estimates can be made for the property values k_1, k_2 and κ_1, κ_2. The main problem is to assign specific numerical values to t_e. As a first approximation this can be done by assuming that all the carbon monoxide given up by the bath in unit time (which can be calculated from the change in carbon concentration of the charge) is uniformly divided into bubbles of equal size. Under these conditions t_e is given by,

$$t_e = \frac{A_B}{A_b N_o}, \tag{17}$$

where A_B = cross-sectional area of the bath,
A_b = surface area swept by one bubble,
N_o = number of bubbles produced in unit time.

A detailed numerical example is worked out as follows.

Consider a 100 ton furnace, with a bath cross-sectional area of 480 ft² [8].
Further, consider "normal boil" conditions when the decrease in carbon concentration is approximately 0·14 wt per cent/h.
Assume a metal temperature of 1555°C and a slag temperature of 1600°C. Finally assume that the bubbles are of a spherical cap shape, having a volume of $14{\cdot}5\ \text{cm}^3 = 5{\cdot}13 \times 10^{-4}\text{ft}^3$ and that the area swept by one bubble is $19{\cdot}6\ \text{cm}^2 = 2{\cdot}11 \times 10^{-2}\text{ft}^2$. Justification for these latter assumptions will be discussed later.

The total volume of carbon monoxide evolved at the bath temperature is given as

$$\frac{0{\cdot}14 \times 2240}{12} \times \frac{359 \times 1828}{273} = 6{\cdot}27 \times 10^4\text{ft}^3/\text{h}.$$

Thus the number of bubbles

$$\frac{6{\cdot}27 \times 10^4}{5{\cdot}13 \times 10^{-4}} = 1{\cdot}23 \times 10^8/\text{h}$$

and t_e is given by substitituting these values into equation (17).

$$t_e = \frac{480}{1{\cdot}23 \times 10^8 \times 2{\cdot}11 \times 10^{-2}} = 1{\cdot}85 \times 10^{-4}/\text{h}.$$

For the property values of materials consider [4].

Slag (phase 1)

$k_1 = 0{\cdot}5$ Btu/h ft degF
$Cp_1 = 0{\cdot}28$ Btu/lb degF
$\rho_1 = 170$ lb/ft³

i.e. $$\kappa_1 = \frac{0{\cdot}5}{0{\cdot}28 \times 170} = 1{\cdot}05 \times 10^{-2}\text{ft}^2/\text{h}.$$

Metal

$k_2 = 10$ Btu/h ft degF
$Cp_2 = 0{\cdot}1$ Btu/lb degF
$\rho_2 = 450$ lb/ft³

i.e. $$\kappa_2 = \frac{10}{0{\cdot}1 \times 450} = 0{\cdot}22\ \text{ft}^2/\text{h}.$$

Thus substitution into equation (13) gives

$$h = \frac{2 \times 0{\cdot}5}{(3{\cdot}14 \times 1{\cdot}05 \times 10^{-2} \times 1{\cdot}85 \times 10^{-4})^{1/2}} \times \frac{\dfrac{10}{0{\cdot}47}}{\dfrac{0{\cdot}5}{0{\cdot}103} + \dfrac{10}{0{\cdot}47}}, \tag{13b}$$

$h = 321$ Btu/h ft² degF.

This value is of the same order of magnitude, although about one and a half times larger than those measured in practice [6].

DISCUSSION

Agreement between transfer coefficients predicted as first approximation by the proposed model and those measured experimentally in practice is not unreasonable; especially if it is noted that practical measurements were not very accurate and that a number of simplifying assumptions had to be made to formulate the model in a comparatively simple mathematical form. Critical examination of these assumptions may lead towards some refinement.

(a) The assumption of uniform temperature fields in the bulk of both phases is probably correct and is supported by experimental evidence [6]. In any case small deviations from uniformity should have no great effect on the value of the transfer coefficient.

(b) Assumption with regard to bubble size was based on visual observations and may be in error to a considerable extent. Indeed, the value of bubble volume is the major uncertainty in the numerical calculations. Once it is accepted that the bubble volume is larger than about 1 cm^3 it is well established [9, 10] that the shape of bubbles is that of a spherical cap and that there exists a mass of fluid of approximately equal volume associated with this spherical cap. Thus the assumption that on hitting the interface the bubble destroys temperature or concentration gradients within an area that equals its own projected area in a plane perpendicular to its motion, does not appear to be unreasonable.

(c) The assumption that the bubbles are evenly distributed over the bath area may be in error but this error is not thought to be too great as compared with the uncertainty of bubble size, [e.g. if it is assumed that the same number of bubbles pass over half the bath area, the other half being unaffected, the transfer coefficient would be reduced by a factor of $\sqrt{(2)}/2$].

CONCLUSIONS

Assuming unsteady state transfer between the arrival of successive bubbles and expression has been derived for the rate of heat or mass transfer across the bubble-stirred interface of two immiscible liquids.

The equations derived have a practical application for interpreting slag–metal interactions in an open-hearth furnace. For this case the value obtained for the transfer coefficient was shown to be of the same order of magnitude as those observed experimentally for similar conditions. The main uncertainty was thought to be due to the fact that bubble sizes were not known with sufficient accuracy. Further work, possibly statistical study of bubble sizes in open-hearth furnaces and also some model work would be necessary to produce data for a more refined treatment.

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APPENDIX

Equations describing mass transfer in accordance with the proposed model can be derived similarly to those for heat transfer [11]. Expressions for the time averaged mass flux are given below:

$$\bar{N}_{t_e} = 2\sqrt{\left(\frac{D_1}{\pi t_e}\right)} \times (\bar{C}_1 - \bar{C}_2/K) \frac{K\left(\frac{D_2}{D_1}\right)^{1/2}}{1 + K\left(\frac{D_2}{D_1}\right)^{1/2}} = \qquad \text{(A1)}$$

$$\equiv 2\sqrt{\left(\frac{D_2}{\pi t_e}\right)} \times K(\bar{C}_1 - \bar{C}_2/K) \frac{1}{1 + K\left(\frac{D_2}{D_1}\right)^{1/2}} \qquad \text{(A2)}$$

where $\bar{N}_{t_e}$ is the time averaged mass flux across the interface, D_1 and D_2 are the diffusivities in phases 1 and 2 respectively, and

$$K = (C_2/C_1) \text{ at } x = 0. \qquad \text{(A3)}$$

It can be seen that if $K(D_2/D_1)^{1/2} \ll 1$, resistance in phase 2 is controlling and conversely, for $K(D_2/D_1) \gg 1$ resistance in phase 1 is the limiting factor.

For the transfer of oxygen from slag (1) to metal (2)

$$K = 2{\cdot}2 \times 10^{-2}$$
$$D_1 = 2 \times 10^{-6}\text{cm}^2/\text{s}$$
$$D_2 = 1 \times 10^{-4}\text{cm}^2/\text{s}$$

i.e. $$K\left(\frac{D_2}{D_1}\right)^{1/2} = 0{\cdot}15.$$

Thus the resistance is largely in the metal phase in spite of the higher diffusivity of oxygen in that phase, because of the low value of the partition coefficient.

Numerical value of the mass-transfer coefficient can be calculated from equation (2A),

i.e.

$$h_D \equiv \frac{N_{t_e}}{K(\bar{C}_1 - \bar{C}_2/K)} = 2\sqrt{\left(\frac{D_2}{\pi t_e}\right)} \times \frac{1}{1 + K\left(\frac{D_2}{D_1}\right)^{1/2}} \qquad \text{(A4)}$$

taking $t_e = 1{\cdot}85 \times 10^{-4}\text{h} = 0{\cdot}67$ s as in the problem for heat transfer we have,

$$h_D = 1{\cdot}2 \times 10^{-2}\ \text{cm/s}$$

which is of the same order of magnitude as experimental values of about (3×10^{-2}) quoted by Darken [4].

Résumé—Cet article propose un modèle mathématique pour décrire le mécanisme de transport de chaleur et de masse à l'interface turbulent de deux liquides non miscibles en mouvement. En faisant l'hypothèse d'un état transitoire (diffusion) entre l'arrivée de bulles successives, on trouve une expression générale du coefficient de transport.

Une application pratique du modèle proposé est l'interprétation des interactions métal-laitier dans un haut-fourneau à foyer ouvert.

Zusammenfassung—Um den Mechanismus des Wärme- und Stoffüberganges an der blasen-gerührten Trennfläche zweier unmischbarer Flüssigkeiten zu beschreiben, wird ein mathematisches Modell vorgeschlagen. Mit der Annahme instationärer Bedingungen (Diffusion) zwischen dem Auftreffen aufeinanderfolgender Blasen lässt sich ein Ausdruck für den Gesamtübergangskoeffizienten finden. Als praktische Anwendung des vorgeschlagenen Modells sind die Schlacke-Metall Wecheslwirkungen im offenen Schmelzofen zu deuten.

Аннотация—Предлагается математическая модель для описания механизма тепло-и массообмена на границе двух несмешивающихся жидкостей, приведенной в движение пузырьками. В предположении нестационарности режима (диффузия) в период между поступлением последующих пузырьков выведено выражение для общего коэффициента переноса.

Практическим применением предложенной модели является объяснение взаимодействия шлака-металла в подовой печи открытого типа.