

FIG. 5. Moisture distribution in radial direction.

liquid layers. This phenomenon results in accelerating the removal of gel and capillary water in concrete. Therefore, the drying rate and the moisture loss at any time are functions of temperature; the drying rate increases rapidly with increase of temperature. Similar results have been reported previously [4].

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## DIFFUSION AND INFORMATION

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(Received 18 February 1983)

#### NOMENCLATURE

|            |                               |
|------------|-------------------------------|
| $a_1, a_2$ | normalizing constants         |
| $c$        | concentration                 |
| $c_0$      | initial concentration         |
| $c_e$      | final concentration           |
| $D$        | diffusivity                   |
| $FI$       | functional information        |
| $H$        | differential entropy          |
| $L$        | length of diffusion cell      |
| $p$        | density distribution function |
| $t$        | time                          |
| $x$        | distance                      |

AS A MEASURE of the disorder of a continuous distribution with the density distribution function  $p(x)$ , the differential entropy can be calculated in the following way [1]:

$$H = -a_1 \int_{-\infty}^{\infty} p(x) \log p(x) dx, \quad (1)$$

where  $a_1$  is a constant.

In distributions with a large number of particles, e.g. as is usually present in diffusion processes, the density distribution of the residence probability  $p(x)$  in equation (1) can be replaced by that of an appropriately normalized concentration  $c(x)$ .

On the other hand, for time-dependent concentration

distributions  $c(x, t)$  between two layers of equal size of a diffusant, the following equation is obtained [2]:

$$c(x, t) = (c_0/2) + (2c_0/\pi) \sum_{n=0}^{\infty} (-1)^n (2n+1)^{-1} \times \exp[-(2n+1)^2 \pi^2 Dt/L^2] \cos[(2n+1)\pi x/L], \quad (2)$$

with the initial conditions

$$c = c_0 \quad \text{in} \quad 0 \leq x < L/2,$$

and

$$c = 0 \quad \text{in} \quad L/2 < x \leq L.$$

Substitution of equation (2) into equation (1) gives the very result which is to be expected from the thermodynamic viewpoint: entropy is increasing with time, attaining a maximum at equilibrium, i.e. at equipartition.

When information and negentropy is equated, this means that the initial state contains the most and the final state the least amount of information. Statistically this is obvious, since the initial distribution of the diffusant is least probable, whilst equipartition is most probable.

In the colloquial sense, gain in information is understood as gain in knowledge. Applied to the simple case of diffusion, this would mean to perform experiments to measure diffusivities, and the knowledge of these and of the laws of diffusion would result in the prediction of time-dependent changes in arbitrary distributions. In this kind of consideration the least knowledge about the process is available from the initial and final states—both being independent of time—since knowledge of these contains nothing about the preceding or impending processes. Therefore, the most informative would be the intermediate state which is equidistant from the initial and the final state. The least squares principle lends itself to a mathematical description of this 'functional information' in the process:

$$FI(t) = a_2 \int_{-\infty}^{\infty} [\Delta_{0,e}^2 - (\Delta_{0,i}^2 + \Delta_{e,i}^2)] dx, \quad (3)$$

where  $a_2$  is a normalizing constant, suited for comparison of different magnitudes;  $\Delta_{0,e} = c_0(x) - c_e(x)$  is the concentration difference between the initial and final distributions;  $\Delta_{0,i} = c_0(x) - c(x, t)$  and  $\Delta_{e,i} = c_e(x) - c(x, t)$  are the concentration differences between the initial (0) and instantaneous (i) distribution and the final (e) and instantaneous (t) distribution, respectively.

Substitution of equation (2) into equation (3) and integration from 0 to  $H$  gives

$$FI(t) = 4a_2 c_0^2 L/\pi^2 \sum_{n=0}^{\infty} (2n+1)^{-2} \exp[-(2n+1)^2 \pi^2 Dt/L^2] \times \{1 - \exp[-(2n+1)^2 \pi^2 Dt/L^2]\}. \quad (4)$$

Equation (4) exhibits the expected properties, vanishing at  $t = 0$  and  $t = \infty$ , and attaining a maximum at

$$\sum_{n=0}^{\infty} \exp[-(2n+1)^2 \pi^2 Dt/L^2] = 1/2,$$

because of the quadric premises (3), the summation in equation (4) in principle covers an expression of the form  $y = z(1-z)$  which obviously attains a maximum at  $z = 1/2$ . Figure 1 presents the time course of the function  $FI(t)$  as defined by equations (3) and (4) in diffusion between two layers of equal size. The maximum value of  $FI(t)$  amounts to ca.  $a_2 c_0^2 L/\pi^2$  and is assumed at  $t \approx 0.07L^2/D$ .

This result shows a remarkable analogy with theoretical calculations [3]. Unnam and Tenney [3] determine the effect of zone size on the convergence of exact solutions for diffusion in single phase systems. With planar geometry they find a cross-over in the number of series terms required by double

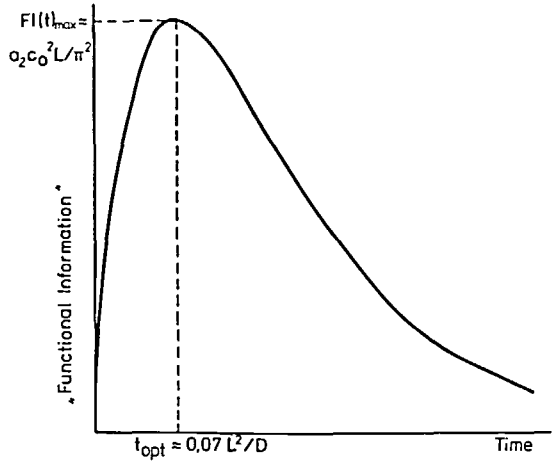


FIG. 1. Time course of the 'functional information'  $FI(t)$  in arbitrary units in a diffusion process.

error function series and Fourier series to occur at  $t \approx 0.111L^2/D$  (with a change of notation).

The function  $FI(t)$  is neither restricted to diffusion processes nor to one dimension. It generally gives a criterion for the optimal moment in the measurement of time-dependent distribution processes. As far as distributions can be described by Fourier series, the squaring in equation (3) does not present substantial difficulties because of the orthogonality of the coefficients. In the particular case of diffusion between two layers of the same size  $FI(t)$  yields plausible results:

(a) Diffusion experiments are all the more informative, i.e. measured concentration differences are all the more significant, the greater the initial concentration and the dimension of the measuring cell.

(b) The optimal moment of measurement is directly proportional to the square of the cell length and inversely proportional to the diffusivity.

Moreover, the definition of  $FI(t)$  entails some philosophical implications. The symbol  $FI(t)$  has been chosen to point to the time-dependent course of distributions—as complementary to equation (1) which rather stresses the structural aspects of distributions. Besides, equation (3) demands modesty in statements on processes with unknown initial and final states, for  $FI(t)$  is then not defined. Indeed, one never knows exactly how informative the present state of such processes may be, and independent facts must be looked for to obtain adequate extrapolations. The problem of mass distribution in the universe may for instance be recalled as a prominent paradigm. Similar considerations will apply to events in the steady state.

On the other hand, as can be inferred from the above diffusion example, the asymptotic approach to a final state does not raise computational difficulties. The connection between entropy, negentropy and information is all the same broadly speaking. Recently Sato [4, 5] has suggested a definition of negentropy as a thermodynamical quantity, aiming at compatibility of the concepts of order, information, non-equilibrium and work. He also made use of the difference between the final and the present state of a thermodynamical system: "Negentropy in the system is the difference between the entropy in the final equilibrium state where it is assumed to be isolated from the exterior and to undergo an irreversible transformation within the system, and the entropy of the system at present state."

The present paper tries to supplement the thermodynamic and the statistic by kinetic considerations.

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*Int. J. Heat Mass Transfer.* Vol. 26, No. 11, pp. 1712–1715, 1983  
Printed in Great Britain

0017-9310/83 \$3.00 + 0.00  
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## AN APPROXIMATE ANALYTICAL SOLUTION TO THE FREEZING PROBLEM SUBJECT TO CONVECTIVE COOLING AND WITH ARBITRARY INITIAL LIQUID TEMPERATURES

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### NOMENCLATURE

|               |  |
|---------------|--|
| $c$           | heat capacity                                      |
| $h$           | heat transfer coefficient                          |
| $k$           | thermal conductivity                               |
| $L$           | latent heat of fusion                              |
| $Ste$         | Stefan number, $C_s(T_m - T_a)/L$                  |
| $Ste'$        | modified Stefan number, $C_l(T_0 - T_m)/L$         |
| $t$           | time   |
| $T$           | temperature  |
| $V$           | parametric variable                                |
| $y$           | space coordinate                                   |
| $Y$           | dimensionless distance, $hy/k_l$                   |
| Greek symbols |  |
| $\alpha$      | thermal diffusivity                                |
| $\epsilon$    | solid thickness                                    |
| $\eta$        | dimensionless solid thickness, $he/k_s$            |
| $\theta$      | dimensionless temperature, $(T - T_m)/(T_0 - T_m)$ |
| $\rho$        | density  |
| $\tau$        | dimensionless time, $\alpha_l h^2 t/k_s^2$         |

### Subscripts

|   |              |
|---|--------------|
| a | ambient      |
| f | liquid phase |
| m | freezing     |
| 0 | initial      |
| s | solid phase  |
| t | surface      |

### 1. INTRODUCTION

THE ONE-DIMENSIONAL moving boundary problems associated with freezing and melting have always been of great interest to engineers and scientists. The problem has wide application, e.g. in freezing and melting of lake ice, cooling of large masses

of igneous rock, solidification of castings and purification of materials.

The problem is characterized by the existence of a moving boundary resulting from phase change. Analytical solutions are possible only for a few special classes of boundary and initial conditions, for example, Stefan or Neumann's problems [1]. For other more complicated boundary conditions, different assumptions have to be used. Series solutions have been attempted [2–5]. More recently, Foss [6] presented a simple approximate solution to an important class of moving boundary problems; the freezing and melting of lake ice. A convective boundary was applied to the air–ice interface. The solution compared closely with Westphal's more accurate series solution [5]. However, the initial water temperature in both cases was assumed to be at the fusion temperature, thus, ignoring conduction in the liquid phase. This limits the usefulness of the solutions in many practical applications.

This paper presents an approximate analytical solution to the moving boundary problem associated with the freezing of a semi-infinite phase-change medium due to convective cooling in the fixed boundary and with an initial temperature which can be higher than the fusion temperature.

### 2. STATEMENT OF THE PROBLEM

The problem considers a semi-infinite body of phase change medium extending from  $y = 0$  to  $\infty$ . The initial temperature of the liquid ( $T_0$ ) is assumed to be uniform and higher than the fusion temperature ( $T_m$ ). Convective cooling is applied to the fixed boundary ( $y = 0$ ) at  $t \geq 0$  with a constant heat transfer coefficient ( $h$ ) and a constant sub-freezing ambient temperature ( $T_a$ ) (Fig. 1). The problem can be divided into two: before and after freezing at the free surface.

Before freezing ( $t < t_m$ ), the problem is relatively simple and