NEL / REPORT 1393

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DIFFUSION IN HYDRODYNAMIC MODELS OF STRATIFIED FLUIDS

How quickly are discrete layers obliterated?
A theoretical solution is presented and confirmed by experiment

A. A. Hudimac,
R. B. Rayment, and R. R. Smith • Research Report • 26 July 1966

U. S. NAVY ELECTRONICS LABORATORY, SAN DIEGO, CALIFORNIA 92152

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PROBLEM

Develop the theory of internal waves in the ocean. Design and perform experiments to obtain needed military information. Specifically, investigate the diffusion of a solute through discrete layers of solvent, and develop a technique for measuring diffusion without disturbing the diffusion process.

RESULT

A technique was developed for determining the rate of diffusion between two discrete layers by taking minute samples at regular intervals with a microcell and determining concentration by conductivity measurement.

RECOMMENDATION

Apply the technique to investigation of the effect of wave action on diffusion between discrete layers.

ADMINISTRATIVE INFORMATION

Work was performed under SR 104 03 01, Task 0594 (NEL L41171) between January and September 1962. The report was approved for publication 26 July 1966.

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INTRODUCTION

Three approaches have been made to the study of internal waves and associated phenomena – theoretical analysis, model study, and field experiment. This report discusses the results of the first two applied to the specific problem of the diffusion of a solute between two discrete layers. The fluid model is prepared by dissolving different amounts of solute in successive layers of fluid.

THEORY

Attention is restricted to a two-layer liquid system with combined thickness l. Let the upper layer, of thickness h, initially be a pure solvent lying on a layer of thickness $(l - h)$ of solution having a concentration C_0. If the diffusion coefficient k can be taken as constant for concentrations ranging from 0 to C_0, the concentration satisfies the simple diffusion equation, subject to the appropriate initial and boundary condition. This problem, as a special case of a more general treatment, is treated in the appendix. The solution is

$$e = \frac{C_0}{2} \left\{ \operatorname{erf}\left(\frac{z-h}{\sqrt{4kt}}\right) - \operatorname{erf}\left(\frac{z-l}{\sqrt{4kt}}\right) + \operatorname{erf}\left(\frac{z+l}{\sqrt{4kt}}\right) - \operatorname{erf}\left(\frac{z+h}{\sqrt{4kt}}\right) \right\} \tag{1}$$

where erf stands for the error function, z is distance from surface, the absolute value of the argument is used, and the sign of the term is changed if the argument is negative.

EXPERIMENTAL VALIDATION

The experiment checks the results of the theory for a special case. If the agreement between theory and experiment is good, the theory and the assumptions on which it is based can then be applied with confidence to a range of similar problems.

The basic problem is to get experimental curves. A sequence of measurements of concentration at several points along a vertical line is required, repeated several times, preferably at times varying exponentially. It is clear that the instrument used in making the measurement or extracting a sample should disturb the layer system as little as possible, both in moving into the fluid and in extracting a sample. The measurement of the chlorinity of very small samples of NaCl solution was considered. However, as this method would have required many insertions of the sampling probe, and as adequate microchemical techniques were not available, it was decided instead to make conductivity measurements with a special microcell that allows a given sequence of measurements to be made with a single insertion.

Since both the diffusion coefficient and the conductivity of the salt solution depend on temperature, a temperature bath was required.

Apparatus

MICROCELL AND BRIDGE

The microcell was built into a capillary tube with an enlarged lenticular chamber. The microcell consists of two platinized platinum electrodes firmly spaced within a glass chamber with a volume of 0.01115 ml between its tip and a level ½ inch above the upper electrode.

The bridge is basically an ac Wheatstone bridge excited with a low-voltage, single-stage vacuum-tube amplifier and equipped with a null-indicator tube. The bridge is provided with a vacuum-tube oscillator circuit supplying 1000-c/s bridge current. The operator balances the bridge by selecting a ratio and adjusting a resistance in the fourth arm of the bridge. A capacitor in parallel with this resistance can be varied to nullify the effect of the capacitance of the leads from the bridge to the cell.

A Hamilton Model 0010 Micro-Syringe was used to withdraw the liquid through the conductivity cell. It consisted of a 1-cc syringe with a plunger in which a lead screw was mounted to withdraw minute amounts of liquid; as the lead screw was turned, the inner part of the plunger moved in or out, changing the volume of the liquid in the syringe. The change in volume per turn of the lead screw was 0.0076 ml ±1.3 percent.

The lead screw could be withdrawn a total of 33 turns. In practice, it was turned three turns at each new position to insure that the sample was first thoroughly flushed from the chamber. Thus, 11 samples could be obtained for each insertion of the probe assembly. The assembly was securely attached to a feeding mechanism, a 12-cm, 28-threads-to-the-inch screw on which was attached a turn knob divided into 100 equal parts. The probe assembly could be advanced to each position with a least count of 0.01 mm.

A cathetometer was used to determine depth (refer to Procedure).

TANK AND BATH

A 5-gallon tank contained the two-layer system. Temperature was maintained at 21.00 (±0.1) °C by immersion of the 5-gallon tank in a 15-gallon aquarium tank. The latter tank was insulated from the atmosphere by a 3-inch layer of Microlite, a commercial form of spun glass. A window cut into the side of the Microlite enabled measuring with the cathetometer, and a small aperture cut into the top admitted the microcell. Water maintained at 21.00 (±0.02) °C flowed through copper tubing coiled around the 5-gallon tank. The temperature was regulated by a controlled-temperature bath manufactured by the Precision Scientific Company (fig. 1).

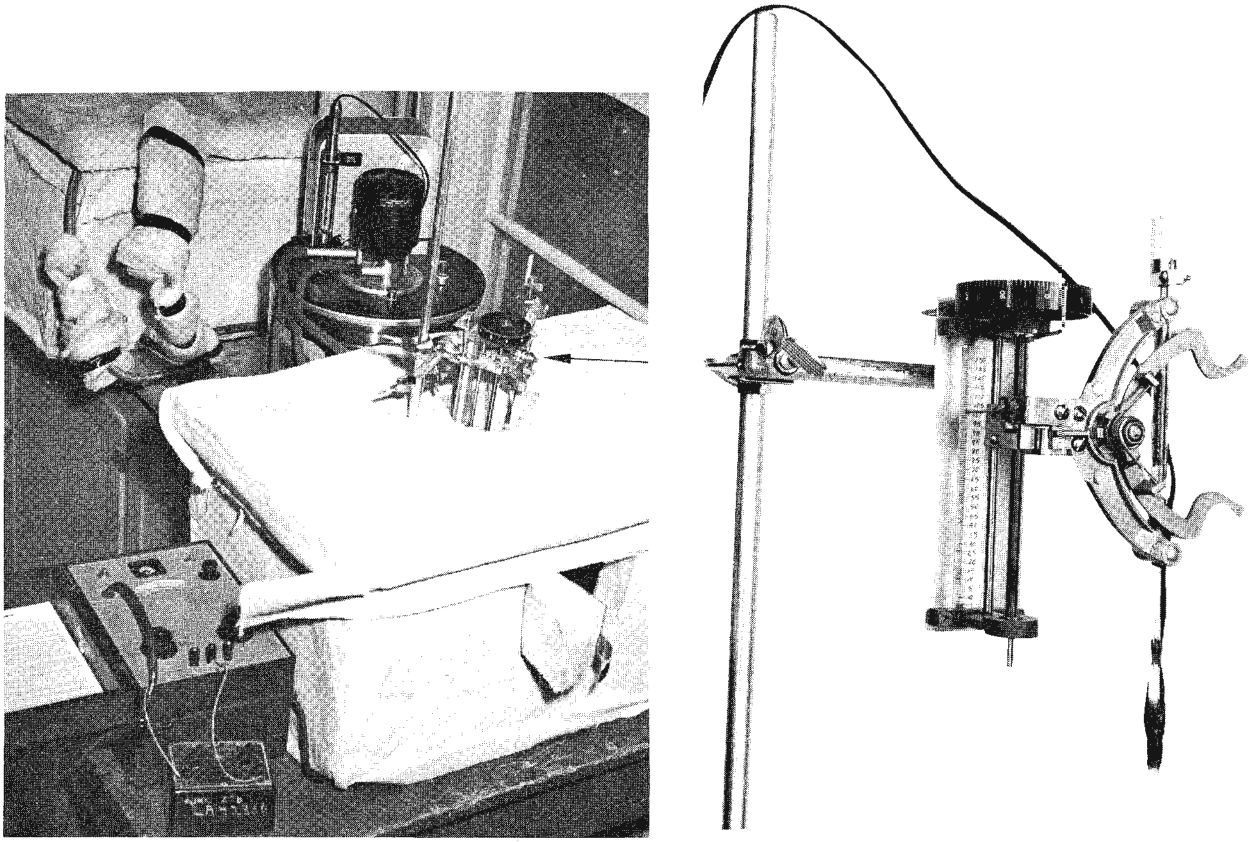

Figure 1. Test setup, showing insulated aquarium tank, temperature control system, and conductivity cell. Photograph at right shows conductivity cell in entirety.

Procedure

The specific conductance bears the following relationship to the resistance: $C = K_c/R$. The cell constant K_c is determined with a solution of known specific conductance. In this instance, KCl with normality of 0.01000 was used at 21.0°C. The specific conductance is 0.001305 mho/cm. The specific conductance of nine different NaCl solutions with normalities ranging from about 0.001 to 1 normality was measured; the temperature was maintained at 21.0 (±0.1) °C. The cell constant did not actually have to be determined; the resistance of the cell corresponding to the normality would have sufficed. The results are plotted in figure 2.

A sharp discontinuity in the concentration was achieved at the interface by laying a sheet of Dow Handi-Wrap over a layer of 1.05-normality NaCl. Distilled water at 21.0°C was poured over the sheet until the desired thickness was obtained, and the sheet was carefully withdrawn from between the two layers over a 5-minute period. Diffusion started at the time the trailing edge of the sheet passed the region in which the microcell would operate. The depth l of the two layers and the thickness h of the upper layer, together with the position of the microcell, were determined by means of the cathetometer.

A short time after the layers were laid down, the first sequence of measurements of conductivity was made at points straddling the initial interface. The microcell was initially filled with distilled water and then inserted into the liquid to the first position, which was determined with the aid of the cathetometer. The screw knob on the micro-syringe was given three turns to withdraw 23 microliters of solution, on which the specific conductance was measured. The time was noted. The microcell was then advanced to the second position by means of the lead screw feed mechanism, and the measurement procedure was repeated. Eleven measurements were taken in a given sequence. The tip position was checked again with the cathetometer at the last position; then the microcell was removed slowly from the body of the liquid, flushed out with distilled water, and made ready for the next run.

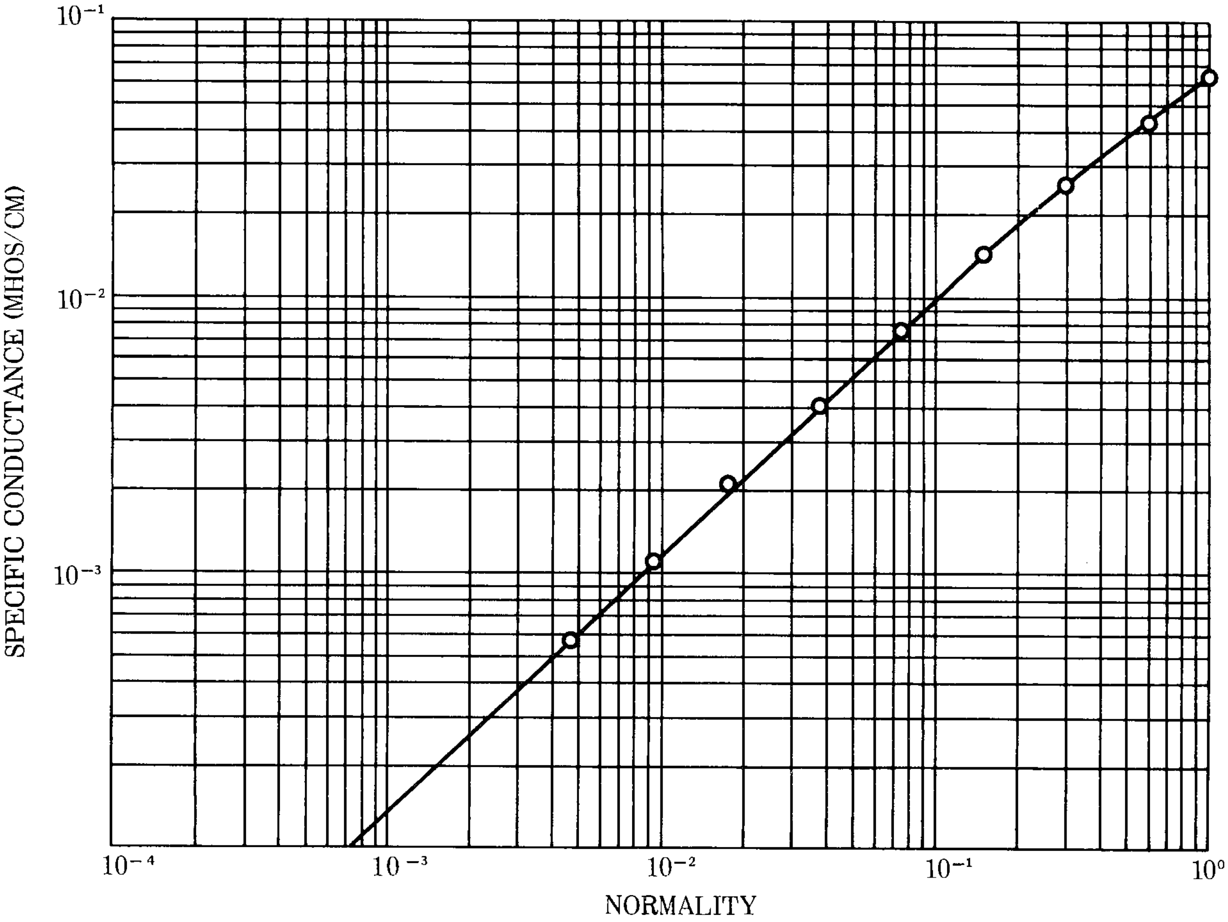

Figure 2. Calibration of conductivity cell.

RESULTS AND CONCLUSION

Five sequences of readings were taken at approximately exponentially increasing times. For each sequence of readings, equation (1) in THEORY was used to determine a theoretical curve. The curve took into account the fact that an appreciable time was required to complete a sequence of readings. The normalized normality is plotted versus the normalized depth, with time a parameter along the curve. The conductance measurements at a given depth were entered into figure 2 to determine the normality, which was then plotted on the same figure as the theoretical curve (figs. 3-7). The agreement between theoretical predictions and experimental results is good, and it is concluded that the theory is valid, and can be applied to a wide range of similar problems.

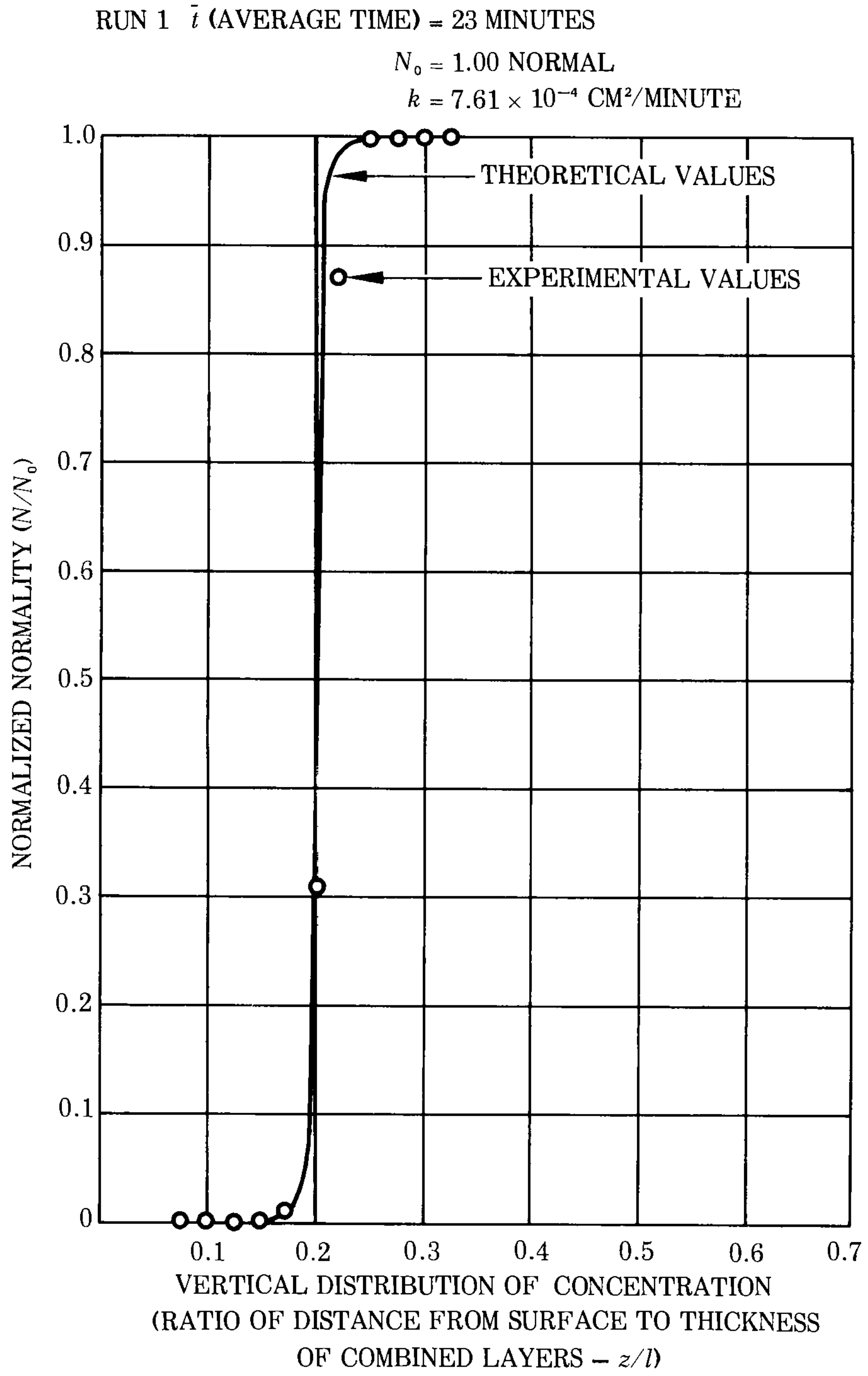

Figure 3. Run one -- diffusion profile at 23 minutes.

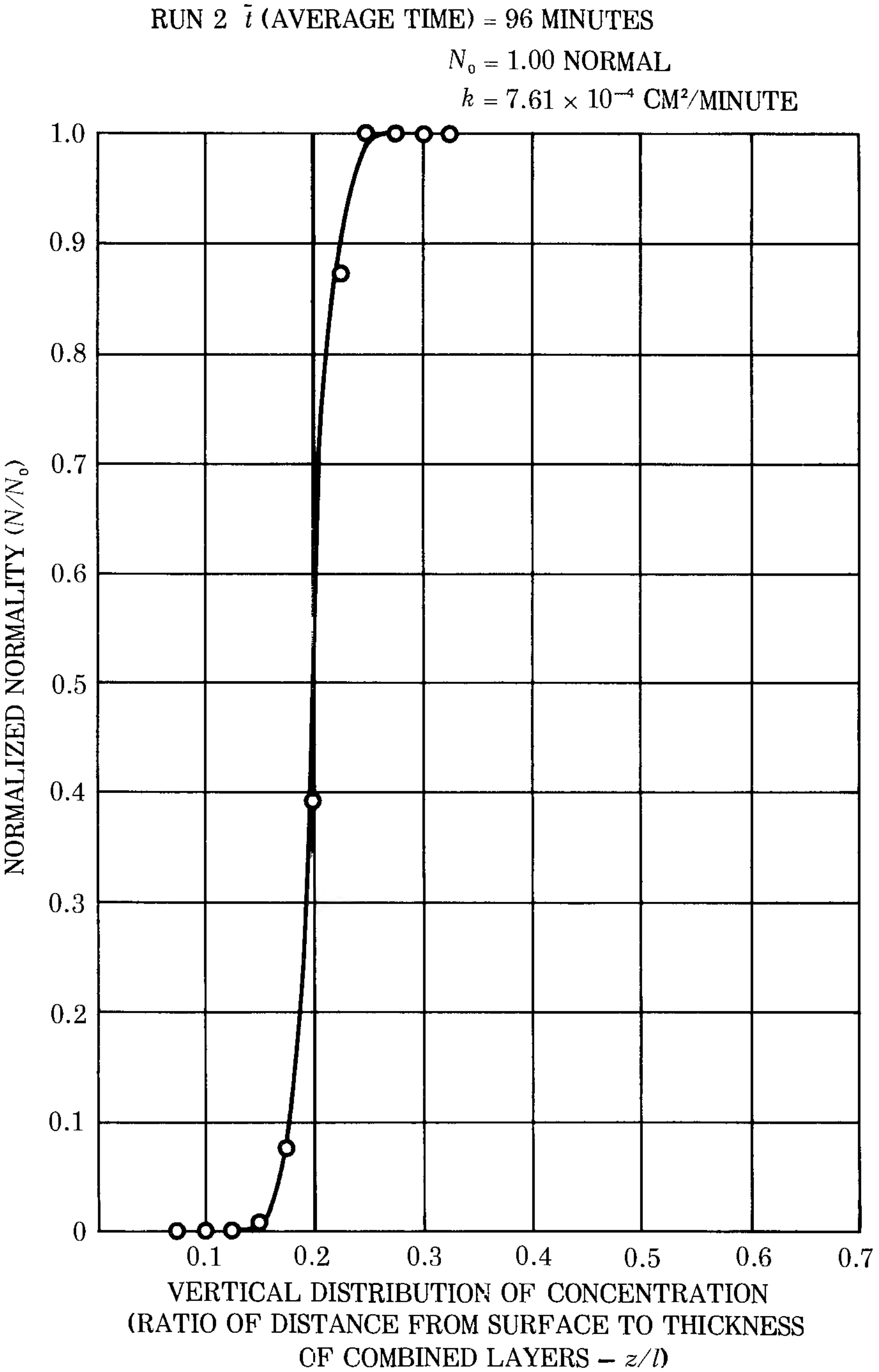

Figure 4. Run two – diffusion profile at 96 minutes.

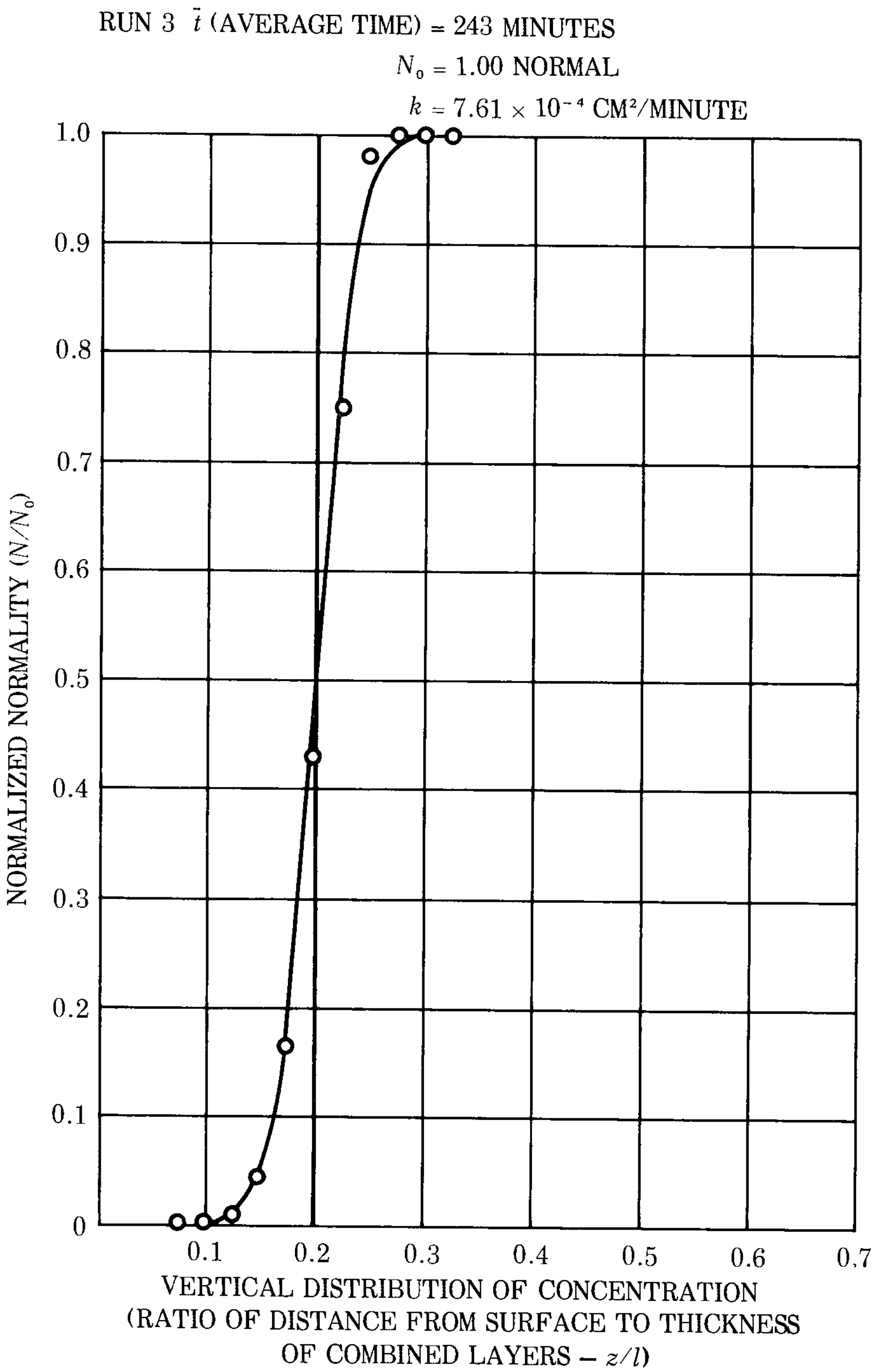

Figure 5. Run three – diffusion profile at 243 minutes.

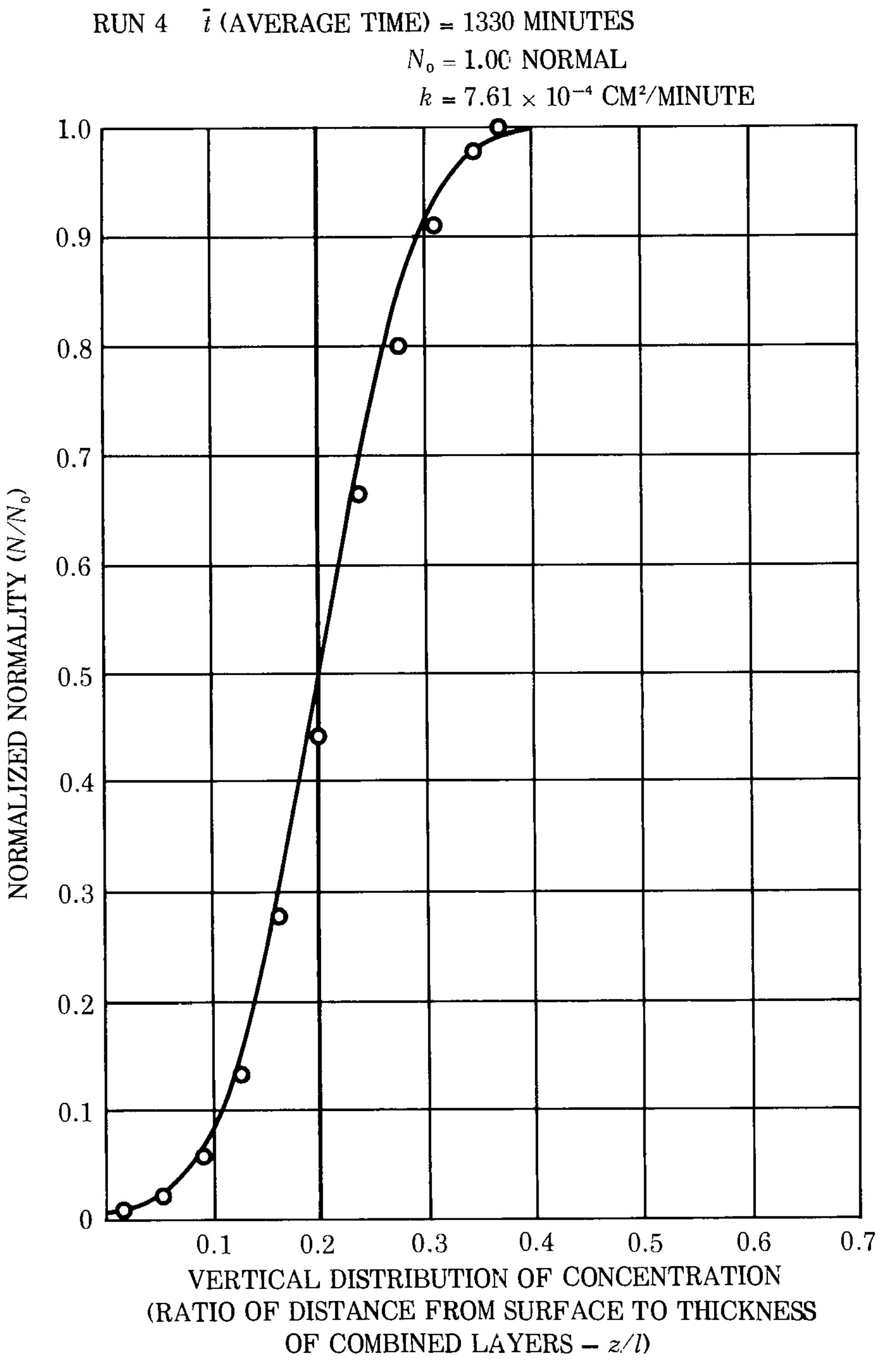

Figure 6. Run four – diffusion profile at 1330 minutes.

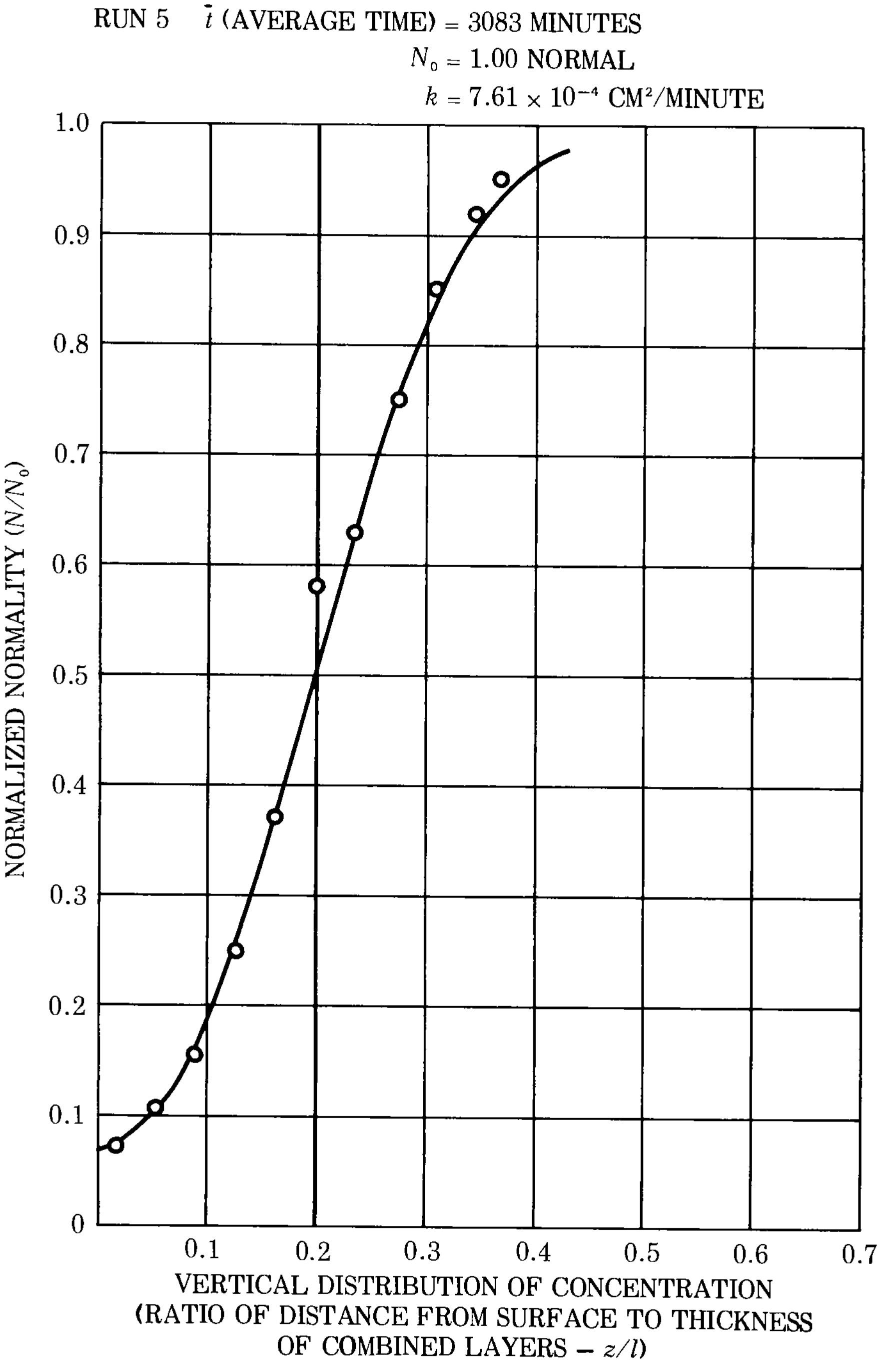

Figure 7. Run five – diffusion profile at 3083 minutes.

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APPENDIX: ANALYSIS

The analysis follows somewhat that in Sommerfeld.*

The diffusion equation in one dimension is

$$\frac{\partial^2 u}{\partial z^2} = \frac{1}{k}\frac{\partial u}{\partial t} \tag{1}$$

where u is the concentration of solute, t is time, and k is the diffusion coefficient. Assume that the variables are separable; that is

$$u = Z(z)\ T(t) \tag{2}$$

Use $-\left(\frac{\pi n}{l}\right)^2$ as the separation coefficient (where $2l$ is the length of the specimen). Then:

$$Z'' + \left(\frac{\pi n}{l}\right)^2 Z = 0,\ T' + \left(\frac{\pi n}{l}\right)^2 kT = 0 \tag{3}$$

The solutions become

$$Z_n = a_n \cos\left(\frac{\pi nz}{l}\right) + b_n \sin\left(\frac{\pi nz}{l}\right),\ T = e^{-\left(\frac{\pi n}{l}\right)^2 kt} \tag{4}$$

A general solution is obtained from the superposition of a number of such solutions for a proper set of values of n.

Case of the Ring

Take $z = 0$ at some point on the ring, and assume that the initial concentration $f(z)$ is arbitrary but symmetrical about $z = 0$. The Fourier expression** for $f(z)$ becomes

$$f(z) = \sum_{n=0}^{\infty} A_n \cos\left(\frac{\pi nz}{l}\right),\ A_0 = \frac{1}{2l}\int_{-l}^{l} f(z)\,dz,\ A_n = \frac{1}{l}\int_{-l}^{l} f(z)\cos\left(\frac{\pi nz}{l}\right)dz \tag{5}$$

The solution for u becomes

$$u(z,t) = \sum_{n=0}^{\infty} A_n e^{-\left(\frac{\pi n}{l}\right)^2 kt} \cos\left(\frac{\pi nz}{l}\right) \tag{6}$$

Now consider $f(z)$ to be a "δ function" situated at $z = \pm\xi$. The expression for f becomes the theta function.*** Thus

---

*Sommerfeld, A., Partial Differential Equations in Physics, Academic Press 1949

**The sine terms drop out because of symmetry.

***It is usually given for $\xi = 0$ and for $l = 1$.

$$u(z,t) = \frac{\theta(z,\xi;t) + \theta(z,-\xi;t)}{2l} = \frac{1}{2l}\left\{\left[1 + 2\sum_{n=1}^{\infty} e^{-\left(\frac{\lambda\pi}{l}\right)^2 kt} \cdot \cos \pi n \frac{(z-\xi)}{l}\right] + \left[1 + 2\sum_{n=1}^{\infty} e^{-\left(\frac{n\pi}{l}\right)^2 kt} \cdot \cos \pi n \frac{(z+\xi)}{l}\right]\right\} \tag{7}$$

Application to a Tank

Here instead of the ring, consider a rod of length l with the boundary conditions

$$\frac{\partial u}{\partial z} = 0; \; z = 0, \; l \tag{8}$$

expressing the fact that there is zero flow of solute at the ends. If the rod is reflected at $z = 0$, with the reflected part having the same initial distribution of solute and the free ends joined, it is seen that the boundary conditions (8) are automatically satisfied, and the problem is reduced to that of the ring. Thus, immediately

$$u = \int_0^l \frac{f(\xi)}{2l} \left[\theta(z,\xi;t) + \theta(z,-\xi;t)\right] d\xi \tag{9}$$

or

$$u = \frac{1}{l}\int_0^l f(\xi)\left\{1 + \sum_{n=1}^{\infty} e^{-\left(\frac{\pi n}{l}\right)^2 kt} \cdot \left[\cos \pi n \frac{(z-\xi)}{l} + \cos \pi n \frac{(z+\xi)}{l}\right]\right\} d\xi \tag{10}$$

The theta function given by expression (7) does not converge rapidly for very small values of kt. We shall transform it to a very rapidly converging series, but first we must put it into a suitable form. Consider

$$\theta(z,\pm\xi;t) = \frac{1}{2l}\left[1 + 2\sum_{n=1}^{\infty} e^{-\left(\frac{\pi n}{l}\right)^2 kt} \cdot \cos \pi n \frac{(z\mp\xi)}{l}\right] \tag{11}$$

Let

$$\tau = \frac{\pi i k}{l^2} t, \quad \chi_{\pm} = \frac{z\mp\xi}{2l} \tag{12}$$

Then

$$\theta(z,\pm\xi;t) = \frac{1}{2l}\left[1 + 2\sum_{n=1}^{\infty} e^{i\tau\pi n^2} \cos 2\pi n \chi_{\pm}\right] = \frac{1}{2l}\,\theta(\chi_{\pm}|\tau) \tag{13}$$

The famous transformation formula for the theta function gives

$$\theta(\chi|\tau) = \left(\frac{\tau}{i}\right)^{-\frac{1}{2}} \cdot e^{\frac{-\pi i \chi^2}{\tau}} \cdot \theta\left(\frac{\chi}{\tau} \,\middle|\, -\frac{1}{\tau}\right) \tag{14}$$

Substituting (12) into (14), (14) into (13), and (13) into (10) gives

$$u(z,t) = \frac{1}{\sqrt{4\pi kt}} \int_0^l f(\xi) \left\{ \sum_{-\infty}^{\infty} e^{-[(z-\xi)+2nl]^2/4kt} + \sum_{-\infty}^{\infty} e^{-[(z+\xi)+2nl]^2/4kt} \right\} d\xi \tag{15}$$

Each term represents a concentration pole at $\xi+2nl$ or $-\xi+2nl$, $n = \ldots, -2, -1, 0, 1, 2, \ldots$, and thus the initial concentration at ξ leads to an infinite sequence of concentration poles made up of the primary pole and all its images reflected at $z = 0, l$. The primary heat pole

$$\frac{1}{\sqrt{4\pi kt}}\, e^{\frac{-(z-\xi)^2}{4kt}}$$

could have been deduced initially (it is called the principal solution), and the images could then have been introduced to satisfy the boundary conditions.

Special Case

Suppose that initially in the tank there is a distribution of concentration

$$\begin{aligned} f(\xi) &= 0, & 0 \le z < h \\ f(\xi) &= C_0, & h < z \le l \end{aligned} \tag{16}$$

Then (15) becomes

$$u(z,t) = \frac{C_0}{\sqrt{4\pi kt}} \int_h^l \left\{ \sum_{-\infty}^{\infty} e^{-(z-\xi+2nl)^2/4kt} + \sum_{-\infty}^{\infty} e^{-(z+\xi+2nl)^2/4kt} \right\} d\xi \tag{17}$$

By letting

$$\frac{(z\mp\xi+2nl)^2}{4kt} = s_{\mp}^2 \tag{18}$$

we transform it into

$$u(z,t) = \frac{C_0}{2} \left\{ \sum_{-\infty}^{\infty} \frac{2}{\sqrt{\pi}} \int_{\frac{z-l+2nl}{\sqrt{4kt}}}^{\frac{z-h+2nl}{\sqrt{4kt}}} e^{-s_-^2}\, ds + \sum_{-\infty}^{\infty} \frac{2}{\sqrt{\pi}} \int_{\frac{z+h+2nl}{\sqrt{4kt}}}^{\frac{z+l+2nl}{\sqrt{4kt}}} e^{-s_+^2}\, ds \right\} \tag{19}$$

The error function is defined

$$\text{erf} = \frac{2}{\sqrt{\pi}} \int_0^{\chi} e^{-s^2}\, ds \tag{20}$$

Note that

$$\frac{2}{\sqrt{\pi}}\int_{a}^{b} e^{-s^2}\,ds = \frac{2}{\sqrt{\pi}}\int_{-\infty}^{b} e^{-s^2}\,ds - \frac{2}{\sqrt{\pi}}\int_{-\infty}^{a} e^{-s^2}\,ds = \operatorname{erf}(b) - \operatorname{erf}(a)$$

The error function is defined for a positive upper limit of integration. Here the upper limit may be negative. Note that

$$\frac{2}{\sqrt{\pi}}\int_{-\infty}^{-a} e^{-s^2}\,ds = 1 - \operatorname{erf} a,\ a > 0$$

Thus

$$u(z,t) = \frac{C_0}{2}\left\{\sum_{n=-\infty}^{\infty}\left[\operatorname{erf}\left(\frac{z-h+2nl}{\sqrt{4kt}}\right) - \operatorname{erf}\left(\frac{z-l+2nl}{\sqrt{4kt}}\right)\right] + \sum_{n=-\infty}^{\infty}\left[\operatorname{erf}\left(\frac{z+l+2nl}{\sqrt{4kt}}\right) - \operatorname{erf}\left(\frac{z+h+2nl}{\sqrt{4kt}}\right)\right]\right\} \quad (21)$$

where the sign of the term is changed if the argument is negative.

As an example of the special case, consider a layer of solvent of thickness h such that $h/l = 0.2$. Let this layer initially lie on a layer of solution with concentration C_0. The subsequent distribution of concentration C divided by C_0 (that is, C/C_0) is plotted as a function of z/l for the following values of the nondimensional parameter kt/l^2: 0.01, 0.0004, and 0.0001 (fig. A1).

The time required to reach the above indicated distributions from the initial state is given in table A1. Here $l = 50$ cm, solutions are aqueous, concentration is in gm-molecules per liter, and k is given in cm²/day.

TABLE A1. EXAMPLES OF DIFFUSION TIME

Material	Concentration	Temperature, ° C	Diffusion Coefficient (k)	kt/l^2	Time (t), days
NaCl	0.1 to 1	15	0.94	0.01	26.6
				0.0004	1.06
				0.0001	0.27
Sugar	1.0	12	0.254	0.01	98.4
				0.0004	3.94
				0.0001	0.984
$CdSO_4$	2.0	11.04	0.246	0.01	102.
				0.0004	4.06
				0.0001	1.02

It can be shown that the gradient at $h/l = 0.2$ is (for $\frac{kt}{l^2}$ not too large) proportional to $1/\sqrt{kt}$ and directly proportional to l. For a given gradient, the time required for a sharp interface to deteriorate to that gradient is inversely proportional to k.

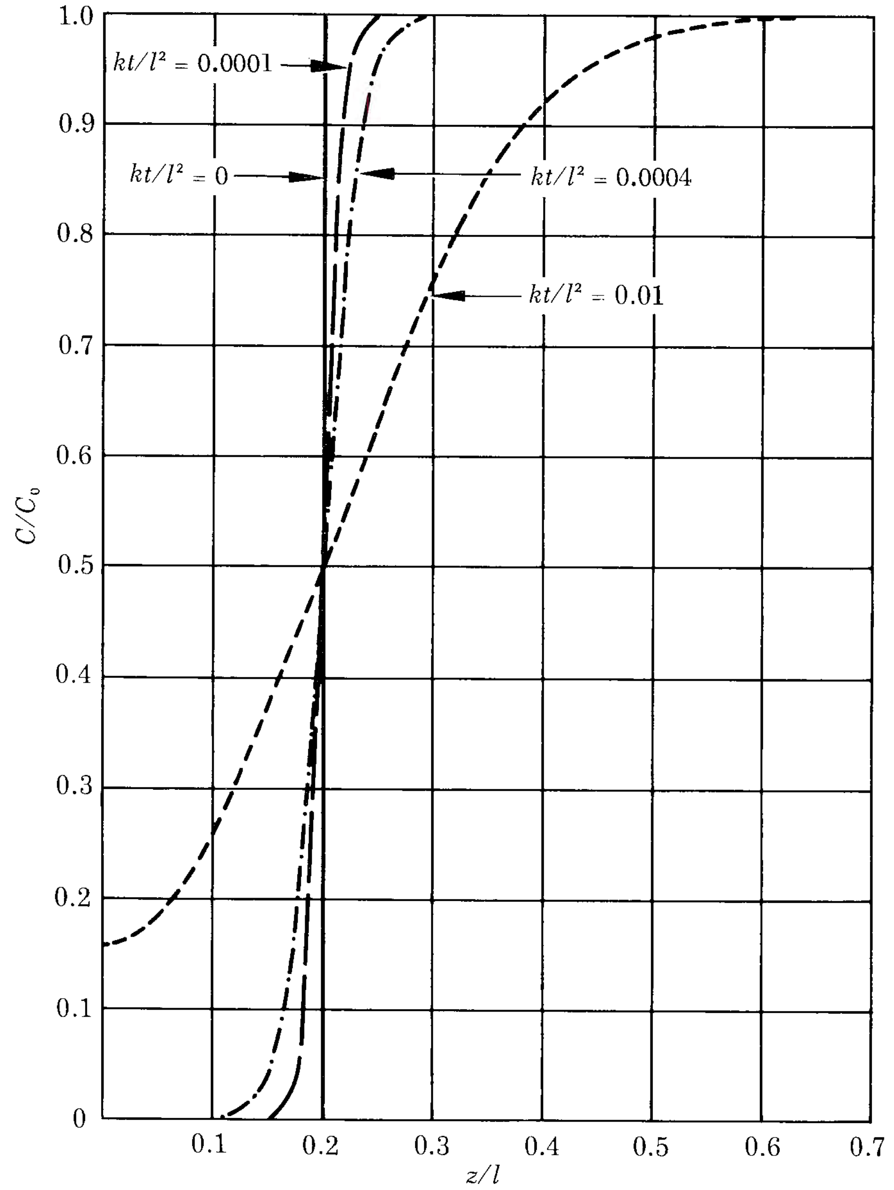

Figure A1. Vertical distribution of concentration as a function of time.

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DIFFUSION IN HYDRODYNAMIC MODELS OF STRATIFIED FLUIDS, by A. A. Hudimac, R. B. Rayment, and R. R. Smith, 21 p., 26 Jul 66.

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A solution is presented to the problem of the obliteration of discrete layers by diffusion. A technique is devised for measuring concentration at several points along a vertical line with minimum disturbance to the layer system. Conductivity measurements with a special microcell are employed. Results of the experiments are compared with theoretical curves, and close agreement validates the theory, which is applicable to a wide range of similar problems.

1. Oceans - Diffusion
2. Fluids - Diffusion
3. Solutions - Diffusion

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