

TABLE 1. COMPARISON OF TEST CONDITIONS

	Biggs	L-P
Type of system	Flow	Batch
Position studied	Exit stream	Total tank contents
Impeller clearance	Impeller diameter	Tank mid height
Tank diameter	2.4 to 3.9	3.0 to 10.0
Impeller diameter	1	1
Baffle width	10	12
Tank diameter	9.62	8.25, 13.0, 21.6
Tank diameter (in.)		

fles. The exact method or position of tracer addition by Landau and Prochazka was not indicated for all tests, but it was noted by them that a variety of positions of addition as well as dif-

ferent volumes and concentrations of tracer seemed to have little effect on their mixing times.

An overall comparison of the mixing times as calculated by the *L-P* correla-

tion with $X = 0.05$ and the observed $t(0.05)$ data of Biggs is presented in Figure 1. Included in Figure 1 are the data points and the calculated values of the *L-P* correlation for three blade square pitch propeller (Biggs did not present a correlation for the propeller data in his article); the *L-P* values are in agreement with the limited data of Biggs. The Biggs data for feed stream passing through the tank in upflow and for the six blade 45 deg. inclined paddle are not included in Figure 1 since they did not match any of the *L-P* correlation lines. It should be mentioned that the six blade 45-deg. paddle data fell about midway between the four blade 45-deg. paddle and six blade fan disk turbine correlations of (Continued on page 182)

Suitability of the Nernst-Planck Equations for Describing Electrokinetic Phenomena

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The quantitative description of electrokinetic phenomena requires the use of reliable rate expressions to relate mass fluxes and the concentration, electrical, and pressure driving forces tending to cause mass movement. Present indications are that (7), at least for systems of simple ionic constituents, the linear flux relations of irreversible thermodynamics (5) are adequate for this purpose. There appear, however, to be very few cases in which the full set of phenomenological coefficients required by these relations has been determined. This is because of the large amount of information required for such a description: $n(n-1)/2$ diffusivities and associated activity data for a system of n diffusing species.* The most complete available treatments appear to be those of Spiegler (13) and Mackay and Meares (11).

In practice, most experimental investigations have been interpreted on the basis of the much simpler pseudo-binary Nernst-Planck equations (6), even though these are known to be incapable of providing a complete description of the systems concerned, and even though their use has often led to poor agreement between prediction and experiment (1). They are known to be useful only for correlation of data taken for individual electrokinetic proc-

esses, for example ion exchange (9) or electrodialysis (1).

Tests of the usefulness of the Nernst-Planck equations are as yet inconclusive, and no generally sound method has been suggested for making such tests. The purpose of this paper is to provide such a method and to demonstrate the need for tests. This is done by rewriting the complete set of linear flux expressions in the form of the Nernst-Planck equations so that the

differences between the two sets of flux expressions may be seen clearly. Procedures for testing the Nernst-Planck equations are then developed and some examples are given. The approach is similar to that used for estimating pseudo-binary diffusivities in ideal gases.

The Nernst-Planck equations are presented in such a wide variety of forms that the use of a single name for all is misleading. In this paper the

TABLE 1†

Friction coefficients $\left(\frac{\text{Joule sec.}}{\text{sq. cm. mole}} \right)$	Diffusivities $\left(\frac{\text{sq. cm.}}{\text{sec.}} \right)$
$^{\circ}F_M^{+W} = 4.93 \times 10^8$	$\overline{D}_M^{+W} = 4.92 \times 10^{-6}$
$^{\circ}F_A^{-W} = 3.32 \times 10^8$	$\overline{D}_A^{-W} = 7.30 \times 10^{-6}$
$^{\circ}F_{WR}^{-} = 4.65 \times 10^6$	$\overline{D}_{WR}^{-} = 5.86 \times 10^{-6}$
$^{\circ}F_M^{+R^{-}} = 2.13 \times 10^8$	$\overline{D}_M^{+R^{-}} = 1.28 \times 10^{-7}$
$^{\circ}F_A^{-R^{-}} = 2.0 \times 10^6$	$\overline{D}_A^{-R^{-}} = 1.35 \times 10^{-5}$
$F_M^{*+M^{+}} = 2.0 \times 10^6$ (estimated)	$\overline{D}_M^{*+M^{+}} = 1.35 \times 10^{-5}$ (estimated)

Concentration in membrane $\left(\times 10^{-3} \frac{\text{mole}}{\text{liter}} \right)$	Equivalent fraction in membrane	
	With A ⁻	Without A ⁻ (estimated)
$^{\circ}C_M^{+} = 0.468$	$x_M^{+} = 0.0113$	$x_M^{+} = 0.011$
$^{\circ}C_A^{-} = 0.0124$	$x_A^{-} = 0.0003$	$x_A^{-} = 0$
$^{\circ}C_W = 40.7$	$x_W = 0.9775$	$x_W = 0.978$
$^{\circ}C_R^{-} = 0.4556$	$x_R^{-} = 0.0109$	$x_R^{-} = 0.011$

† Values of the F_{ij} and the c_i are those of Mackay and Meares except $F_M^{*+M^{+}}$. This was estimated to be equal to $F_A^{-R^{-}}$ because of lack of specific information. All \overline{D}_{ij} except \overline{D}_A^{-W} and $\overline{D}_A^{-R^{-}}$ are calculated on the basis of the A^{-} -free compositions. A^{-} = anion, M^{+} = cation, M^{*+} = cation isotope, R^{-} = resin.

* In this paper the membrane is considered to be one of the diffusing species.

TABLE 2. CATION EXCHANGER MEMBRANE WITH M^+ (M^{*+}), R^- , AND W

Operation	$\alpha_i = 0$		$\alpha_i = 1$		$\alpha_i > 0$			
	\mathcal{D}_{M^+I}	\mathcal{D}_{WI}	\mathcal{D}_{M^+m}	\mathcal{D}_{Wm}	$\mathcal{D}_{M^+, \text{eff}}$	$\mathcal{D}_{W, \text{eff}}$	α_{M^+}	α_W
Self-diffusion	3.50×10^{-6}	—	3.50×10^{-6}	—	$3.46 \times 10^{-6}\dagger$	5.35×10^{-6}	—	1.02
	$3.51 \times 10^{-6}\ddagger$	—	$3.51 \times 10^{-6}\ddagger$	—	$3.47 \times 10^{-6}\ddagger$			
Conductivity	5.66×10^{-6}	—	2.59×10^{-6}	—	3.47×10^{-6}	5.35×10^{-6}	0.71	1.00
Streaming current	—	2.43×10^{-4}	4.92×10^{-6}	5.35×10^{-6}	3.47×10^{-6}	5.35×10^{-6}	0.706	1.00

$\ddagger \mathcal{D}_{M^{*+}M^+}$ assumed equal to infinity.

\dagger with $x_{M^{*+}} = 0.0055$.

following definition will be used:

$$N_i = -c D_{im} \nabla X_i + \alpha_i x_i \sum_{j=1}^n N_j \quad (1a)$$

$$= -c D_{im} \nabla X_i + \alpha_i c_i \mathbf{v}^* \quad (1b)$$

Here

$$\nabla X_i = \nabla x_i + x_i \nabla \ln f_i + \frac{x_i v_i F}{RT} \nabla \phi + \frac{x_i}{RT} \left(\bar{V}_i - \frac{M_i}{\rho} \right) \nabla p \quad (2)$$

Equation (1) is convenient for several reasons.

1. It is consistent with the accepted form of Fick's first law for the limiting case of a binary system (2) with only concentration diffusion when $\nabla X_i =$

$$\frac{\partial \ln f_i x_i}{\partial \ln x_i} \text{ and } \alpha_i = 1.$$

2. It allows for the sometimes important effects of convection, through inclusion of the last term on the right.

3. The use of the molar-average velocity, \mathbf{v}^* , rather than the mass-

average velocity, \mathbf{v} , of Helfferich (8) results in a simpler set of flux expressions.

4. The introduction of α_i , usually considered to be unity or zero in existing correlations of experimental data, will be shown to be useful in the discussion below.

A completely analogous set of equations may be written in terms of mass rather than molar quantities, and it is in such an equation that the mass-average velocity is best used.

It only remains to put the more reliable flux equations of irreversible thermodynamics in a similar form. To do this one begins with the generalized Stefan-Maxwell equation (10):

$$\nabla X_i = \sum_{j=1}^n \frac{x_i x_j}{\bar{D}_{ij}} (\mathbf{v}_j - \mathbf{v}_i) \quad (3a)$$

$$= \sum_{j=1}^n \frac{1}{c \bar{D}_{ij}} (x_i N_j - x_j N_i) \quad (3b)$$

Equations (3a) and (3b) are equivalent to the expressions introduced

earlier by Spiegler (13)* but are more convenient for the present purpose. First, they are of the same form as the Stefan-Maxwell equations for ideal gases, thus permitting immediate use of the pseudo-binary expressions that have been developed for these better-understood systems. Second there is reason to believe (10) that the \bar{D}_{ij} in Equations (3a) and (3b) are less concentration dependent than most other phenomenological coefficients now in use (see Table 1).

There are many ways to force Equations (3a) and (3b) into the form of Equation (1), but three seem especially promising.

The method used most frequently is (Continued on page 185)

* The friction coefficients defined by Spiegler, which are also consistent with the principles of irreversible thermodynamics, are given by

$$\frac{RT \nabla X_i}{x_i} = \sum_{j=1}^n F_{ij} (\mathbf{v}_j - \mathbf{v}_i)$$

so that

$$F_{ij} = \frac{RT x_j}{\bar{D}_{ij}}$$

with

$$x_i F_{ij} = x_j F_{ji}$$

Distillation Studies — Viscous Drag on a Gas Bubble Rising in a Liquid

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This study forms part of an investigation into the fluid mechanics of a perforated plate distillation column at its minimum load. In particular, the seal point (6), at which stage liquid is just flowing over the downcomer weir, is considered. Here the gas is bubbling, singularly or otherwise, through some of the holes, while liquid is dumping through the others. One equation of motion can be obtained from a momentum balance on a rising bubble:

$$(\text{Net Buoyance Force} + \text{Excess Pressure} + \text{Rate of Loss of Gas Momentum}) = (\text{Viscous}$$

Drag + Surface Tension Drag + Inertial Force of the Rising Bubble)

For a single gas bubble, the viscous drag term is usually represented by Drag Force = Drag Coefficient \times Inertial Stress \times Projected Area or

$$F_D = C_D \cdot \frac{1}{2} \rho_L U^2 \cdot \frac{\pi}{4} D_e^2$$

The drag coefficient may be correlated experimentally by

$$C_D = a/N_{Re}$$

where

$$N_{Re} = D_e U \rho_L / \mu_L$$

PREVIOUS WORK

Datta (1) and Gorrington (3) have studied various proposed relations for the value of a :

$$\text{Stokes' equation: } a = 24$$

$$\text{Moore's equation: } a = 32$$

Hadamard's equation:

$$a = 24 \left(\frac{2 \mu_L + 3 \mu_G}{3 \mu_L + 3 \mu_G} \right)$$

(Continued on page 188)

(Continued from page 176)

to define an effective binary diffusivity \mathcal{D}_{im} for each species by

$$N_i = -c \mathcal{D}_{im} \nabla X_i + x_i \sum_{j=1}^n N_j \quad (4)$$

$$\alpha_i = 1, \text{ all species and operations} \quad (5)$$

It then follows (3) that

$$\frac{1}{\mathcal{D}_{im}} = \frac{\sum_{j=1}^n \frac{1}{D_{ij}} (x_j N_i - x_i N_j)}{N_i - x_i \sum_{j=1}^n N_j} \quad (6)$$

Here the α_i are set equal to unity, to conform to usual practice, and in agreement with the corresponding expression for binary systems. Then \mathcal{D}_{im} corresponds to the D_{im} of Equation (1). This is an approach long used with success in work with ideal gases (3). It suffers from the defect that \mathcal{D}_{im} is not a system property. That is, it depends upon the flux ratios (N_i/N_k) as well as upon system temperature, pressure, and composition. It will be shown below that this dependence upon flux ratios can be a severe limitation to the usefulness of Equation (4).

Alternately one may define an intrinsic diffusivity \mathcal{D}_{iI} , by setting $\alpha_i = 0$:

$$\frac{1}{\mathcal{D}_{iI}} = \sum_{j=1}^n \frac{x_j}{D_{ij}} \left(1 - \frac{x_i}{x_j} \frac{N_j}{N_i} \right) \quad (7)$$

Third, one may define an effective diffusivity, $\mathcal{D}_{i, \text{eff}}$, which is a system property, and thereby fix the value of α_i for each species and set of flux ratios. Here

$$N_i = -c \mathcal{D}_{i, \text{eff}} \nabla X_i + \alpha_i x_i \sum_{j=1}^n N_j \quad (8)$$

with

$$\frac{1}{\mathcal{D}_{i, \text{eff}}} = \left(\sum_{\substack{j=1 \\ j \neq i}}^n \frac{x_j}{D_{ij}} \right) / (1 - x_i) \quad (9)$$

It then follows that

$$\alpha_i = \frac{N_i + \sum_{\substack{j=1 \\ j \neq i}}^n N_j \frac{\mathcal{D}_{i, \text{eff}}}{D_{ij}}}{\sum_{k=1}^n N_k} \quad (10)$$

One now finds that α_i is different for each species, is dependent upon the flux ratios, and is not in general unity. The value of α_i will become unimportant when $x_i \ll 1$ and will approach a value of unity when $\sum_{\substack{j=1 \\ j \neq i}}^n N_j \ll N_i$. In

Hydrates at high pressures: Part III. methane-argon-water and argon-nitrogen-water systems, Saito, Shozaburo, and Riki Kobayashi, *A.I.Ch.E. Journal*, 11, No. 1, p. 96 (January, 1965).

Key Words: Hydrates-1, Clathrates-1, Inclusion Compounds-2, Phase Behavior-8, Gas-Liquid-Solid Equilibria-8, Methane-Argon-Water-1, Argon-Nitrogen-Water-1, High-Pressure Apparatus-10.

Abstract: Equilibrium gas hydrate data were obtained for binary gas mixtures of argon with methane and argon with nitrogen in water by a method introduced previously (see Part I) up to 15,000 lb./sq. in. abs. The data are presented in tabular and graphical form. The general hydrate theory of van der Waals and Platteeuw, after modifications, proved accurate.

Stability criteria for miscible displacement of fluids from a porous medium, Schowalter, W. R., *A.I.Ch.E. Journal*, 11, No. 1, p. 99 (January, 1965).

Key Words: Fluid Mechanics-8, Porous Media-9, Petroleum-9, Petroleum Production-8, Secondary Recovery-8, Hydrodynamic Stability-8, Flow-8, Numerical Analysis-10, Miscible Displacement-8, Crude Oil-9, Darcy's Law-10, Eigenvalue-10, Viscosity-6, Density-6.

Abstract: A model has been formulated for miscible displacement of a fluid from a porous medium. The model consists of two pure fluids separated by a region of constant thickness in which the two fluids interpenetrate. Stability criteria have been found for this model when the flow is vertically downward and the less dense fluid is displaced by the more dense fluid. Differences in pure-component densities, viscosity variation in the mixing zone, and displacement velocity all have major effects on the stability criteria for the model.

Thermal conductivity of porous catalysts, Butt, John B., *A.I.Ch.E. Journal*, 11, No. 1, p. 106 (January, 1965).

Key Words: Conductivity-8, Physical Properties-9, Silver Catalysts-7, Alumina Catalysts-7, Heat Conduction-9, Temperature-6, Catalytic Pore Structure-6.

Abstract: A random pore model of catalytic pore structure is applied to the determination of the thermal conductivity of porous catalytic solids. The low thermal conductivities reported for such materials are explained by considering the microporous particles to consist of a packed bedlike assembly of smaller particles; heat transfer through the solid fraction occurs by conduction through individual small particles in contact with other particles. An expression for the effective thermal conductivity of a gas in the region between ordinary thermal conduction and free molecule conduction is used in determination of heat conduction through the void volume. Agreement of the theory with experimental data on alumina and silver catalysts over a wide range of conditions is good.

Solution of problems involving conventional and complex columns at unsteady state operation, Waggoner, R. C., and C. D. Holland, *A.I.Ch.E. Journal*, 11, No. 1, p. 112 (January, 1965).

Key Words: Distillation-1, Transient-1, Separations-6, Computer-10, Computations-10, Temperature-6, Enthalpy-7, Boiling Points-7, Compositions-7, Pressure-6.

Abstract: The θ method of convergence has been applied to conventional and complex columns at unsteady state operation. A different holdup for each stage may be specified in terms of molal, mass, or volumetric units. Plate efficiencies were used, and the K values and enthalpies were taken to be polynomials in temperature.

this latter case $D_{i,eff}$ and the D_{im} of Equation (6) become identical (4).

All of the sets of flux expressions introduced via Equations (4) through (10) have the desired property of being at the same time consistent with Equation (3a) and (3b) and of the form of Equation (1). They can thus be used to test the reliability of this latter equation if suitable \bar{D}_{ij} are available.

What are believed to be the best available data are shown marked by asterisks in Table 1 (11). Values of the corresponding \bar{D}_{ij}^* are also shown in this table. The diffusivity $\bar{D}_{M^++M^+}$ arises naturally from the Stefan-Maxwell equations but appears to have been neglected in the membrane literature. It was mentioned by Meares (12) who assumed it to be so large as to be negligible. It is interesting that the magnitude of the \bar{D}_{ij} in Table 1 fall into three groups:

1. Pairs of one charged and one neutral species, with \bar{D}_{ij} on the order of 5×10^{-6} sq.cm./sec.

2. Species of opposite charge, $\bar{D}_{M^+R^-} = 1.28 \times 10^{-7}$ sq.cm./sec.

3. Species of similar charge, $\bar{D}_{A^--R^-} = 1.35 \times 10^{-5}$ sq.cm./sec.

The relative magnitudes of \bar{D}_{ij} in these three groups are plausible on physical grounds. Thus species of opposite charge could be expected to interact most strongly and hence have a lower mutual diffusivity (which may be considered the inverse of a frictional coefficient).

Shown in Table 2 are calculated values of D_{im} , D_{ii} , $D_{i,eff}$, and α_i for some common electrokinetic processes. These were obtained from Equations (6), (7), (9), and (10), using the data of Table 1. In all cases the co-ion concentration was assumed to be zero. These results emphasize the weaknesses of all pseudo-binary treatments:

1. There are many situations for which the diffusivities, or the α_i , are indefinable.

2. The proper numerical value to be used for the diffusivities varies with the operation being considered.

The first of these weaknesses is frequently unimportant. Thus, in the case of self-diffusion, for the assumptions made here (identical properties for M^+ and M^{*+}), there will be no diffusional movement of water and no convective motion at all. The second weakness is much more serious: both D_{ii} and D_{im} show large variation from one process to another. This means that data taken under one set of conditions cannot safely be used to predict performance under others. The $D_{i,eff}$ seem

superior to the others, but can only be determined if all the \bar{D}_{ij} are known.

It does appear from the above calculations that $\bar{D}_{M^{*+}M^{+}}$ may be negligibly large, as usually assumed. Nevertheless an experimental check is desirable.

NOTATION

- c = total molar density of solution

$$= \sum_{i=1}^n c_i$$

 c_i = molar concentration of species i
 $D_{im}, \bar{D}_{ij}, \bar{D}_{im}, \bar{D}_{ii}, \bar{D}_{i,eff}$ = diffusivities
 F = Faraday's constant
 F_{ij} = frictional coefficient
 f_i = activity coefficient of species i
 M_i = molecular weight of species i
 N_i = molar flux of species i with respect to stationary coordinates
 n = number of species, including the membrane matrix
 p = pressure
 R = international gas constant
 T = absolute temperature
 \bar{V}_i = partial molal volume of species i
 v^* = molar average velocity
 v = mass average velocity
 v_i = velocity of species i with respect to stationary coordinates
 X_i = dimensionless total potential for diffusion
 x_i = mole fraction of species i
 α_i = dimensionless weighting factor for the convective term of the Nernst-Planck equation
 ∇ = del, or nabla, operator
 ν_i = charge of species i ; for example -1 for chlorine
 ρ = density of solution
 ϕ = electrostatic potential

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INFORMATION RETRIEVAL

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The influence of mass transfer on a countercurrent liquid-liquid fluidized system, Dunn, I., Leon Lapidus, and J. C. Elgin, *A.I.Ch.E. Journal*, **11**, No. 1, p. 158 (January, 1965).

Key Words: A. Mass Transfer-6, Holdup-7, Fluids-9, Liquid-Liquid Fluidized Systems-9, Countercurrent-0, Extraction-4, Solute-6, Droplets-9, Coalescing-7, Experimental-, Rate-6, Flow-9, Separation-4. B. Mass Transfer-6, Extraction-4, Solute-9, Operation-7, Columns (Process)-9, Separation-4. C. Holdup-8-7, Methyl Isobutyl Ketone-5, Water-5, Acetic Acid-1, Dodecyl Sodium Sulfate-6, Surface Active Agents-6, Aliphatic Compounds-1, Carboxylic Acid-1, Ketones-5. D. Holdup-8-7, Toluene-5, Water-5, Acetone-1, Aromatic Compounds-5, Dodecyl Sodium Sulfate-6, Surface Active Agents-6, Aliphatic Compounds-1, Hydrocarbons-5, Ketones-1.

Abstract: The effect of mass transfer on holdup of countercurrent fluidized systems and the effect of solute on droplet coalescence were determined experimentally. The effect of solute transfer on operation of extraction columns is discussed. Data were taken on two systems, methyl isobutyl ketone-water and toluene-water with acetic acid and acetone solutes, respectively. Dodecyl sodium sulfate was added to some of the runs to determine the effect of surface active agent.

The thermal conductivity and diffusivity of gases for temperatures up to 10,000°K., Mathur, G. P., and George Thodos, *A.I.Ch.E. Journal*, **11**, No. 1, p. 164 (January, 1965).

Key Words: High Temperatures-6, Self-Diffusivity-8, Mutual Diffusivity-8, Thermal Conductivity-8, Generalized-9, Dimensional Analysis-9, Critical Constants-10.

Abstract: A generalized method has been developed for the prediction of the diffusivity and thermal conductivity of nonpolar gases for temperatures up to 10,000°K. This method is applicable to monatomic gases and to diatomic gases existing in their molecular and dissociated states. Dimensional analysis has been used to obtain relationships for self- and mutual diffusivities and for the translational contribution to the thermal conductivity of these substances. For the internal contribution to thermal conductivity, a modified Eucken-factor relationship is presented. Furthermore, relationships have been developed for the calculation of these transport properties for the equilibrium mixture of a dissociating gas.

Particle flow patterns in a fluidized bed, Marsheck, R. M., and A. Gomezplata, *A.I.Ch.E. Journal*, **11**, No. 1, p. 167 (January, 1965).

Key Words: Flow Patterns-2, 8, Solid Particles-0, Fluidized Bed-9, Thermistor-0, Anemometer-0, Probe-10, Silica-Alumina-10, Velocity-6, Air-0, Bed Height-6, Radial-0.

Abstract: Solid particle flow patterns in a fluidized bed were studied with a modified thermistor anemometer probe. Radial flow patterns were found not to be symmetrical with the axis of the fluidized bed but were similar at angles of 60 deg. The percentage of the cross-sectional area of the bed through which the particle flow is upward was found to be 60%. The principal factors affecting the flow pattern were the superficial air velocity and the ratio of the distance above the support plate to the fluidized bed height.

bubble can be satisfactorily calculated despite the assumption of spherical bubbles when their real shape is that of an oblate spheroid.

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NOTATION

a	= constant
b	= constant
C_D	= drag coefficient in clear liquid based on D_e
D_e	= equivalent diameter (of sphere of same volume)
E	= kinetic energy
F_D	= drag force
Ir'	= volume integral
p_{ro}	= tangential stress
R	= $(D_e/2)$
R'	= region of liquid investigated
r	= bubble radius
t	= time
U	= velocity of rising bubble relative to liquid
u_r, u_θ, u_ω	= velocity components, spherical form
u, v, w	= velocity components, Cartesian form
n	= unit vector, normal to bubble surface
N_{Re}	= $(D_e U \rho_L / \mu_L)$
β	= coefficient of sliding friction
ϵ	= coefficient of surface viscosity
θ	= angle between r and the vertical
μ_G	= gas viscosity
μ_L	= liquid viscosity
ρ_L	= liquid density
σ	= standard deviation
ϕ	= velocity potential
ω	= angle in horizontal plane
∇	= the Laplacian operator

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