Analysis of Phase-Boundary Motion in Diffusion-Controlled Processes:

Part I. Solution of the Diffusion Equation with a Moving Boundary

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Three general methods are developed for solving moving-boundary problems which are governed by diffusional processes such as heat and mass transfer. Examples of such problems include melting, evaporation, and ablation. A method based upon a Riemann-Volterra integration of the diffusion equation leads to nonlinear integrodifferential equations for the boundary motion that are in terms of definite integrals involving Green's functions. An analytical method, which is more convenient for problems involving phase motion, is based on the method of intermediate integrals. A numerical method based on finite difference approximations is implemented on the differential analyzer (analogue computer).

The analysis of a diffusion process frequently involves consideration of the motion of a phase boundary. Familiar examples include melting or freezing which is controlled by heat conduction and evaporation which is controlled by mass diffusion. The mathematical models describing these processes are known as *moving-boundary problems, free-boundary problems, or Stefanlike problems* and are generally regarded by engineers and mathematicians alike as being problems of considerable difficulty.

The published literature on this subject is profuse and dates back at least to 1840 and the work of Neumann (*1*). A bibliography of many of the more significant contributions may be found in reference 2. Notable among the more recent contributions are those of Danckwerts (*3*), of Scriven (*4*) in the engineering literature, and of Kolodner (*5*) in the mathematics literature.

Most of the papers dealing with moving-boundary problems have consisted of analyses of particular problems, with the result that the methods which have been developed and the solutions obtained are quite specialized and difficult to generalize. The work of Danckwerts (*3*) seems to be the first organized general assault on problems of this type, but even it is restricted rather severely on boundary conditions and deals only with one-dimensional diffusion with a moving plane boundary separating two phases of semi-infinite extent.

The need for a more general and organized approach to the entire class of moving-boundary problems seems clear. This work, which will be presented in three parts, was undertaken to help fill this need. In the present paper, the mathematical bases for three general methods of solving the moving-boundary problems in a rigorous manner are developed. This is done without reference to specific problems, for the methods are more general than any particular problem which could serve as a suitable example.

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In Part II, the methods developed here will be applied in detail to the problem of evaporation from a flat surface, in order to illustrate their explicit application. In Part III, these methods will be applied to more difficult problems in cylindrical and spherical coordinates, with the penetration of metal ions into cellulose xanthate fibers and the growth of sulfuric acid droplets in humid air used as examples.

PROCEDURE FOR QUIESCENT PHASES

In the absence of bulk motion of the diffusion phase, the differential equation describing a diffusion process may be written as

$$D\nabla^2 u = \frac{\partial u}{\partial \theta} \qquad (1)^*$$

In this section, a method known as a *Riemann-Volterra integration* (*6*) is used to solve Equation (1) in an arbitrary volume subject to arbitrary boundary conditions, and the application to moving-boundary problems is indicated. This procedure leads to a class of nonlinear integrodifferential equations for the boundary motion.

The idea of concentrating the nonlinearity of a moving-boundary problem into an integrodifferential equation is not new. Kolodner (*5*), in 1956, was apparently the first to arrive at solutions of this type, but his approach is quite different from that taken here.† The Riemann-Volterra

* The simple, classical form of the diffusion equation as written here involves many assumptions, as the reader will realize. Constant phase densities and transport coefficients and absence of sources or sinks are assumed. Neglect of forced, pressure, and thermal diffusion in the case of mass transport, and viscous dissipation, mass diffusional transport, and radiation in the case of energy transport are also implicit. For detailed derivations including these effects, see reference 2.

† Kolodner constructed solutions for linear flow problems from a knowledge of the jump properties of fundamental (source) solutions of the diffusion equation. The present approach is considered more orderly and more readily applicable to problems in cylindrical and spherical coordinates and to cases involving variable boundary conditions.

integration, which is quite similar to what Carslaw and Jaeger (*1*) have employed for the solution of fixed-boundary problems and referred to as the method of Green's functions, was attempted in a slightly simpler form by Miles (*7*) in 1951. There have been numerous papers dealing with the existence and uniqueness of solutions of this type, and Miranker (*8*) has placed bounds on certain of these solutions by use of the law of the mean.

Let v be a function satisfying the adjoint of Equation (1):

$$D\nabla^2 v = -\frac{\partial v}{\partial \theta} \qquad (2)$$

One may combine Equations (1) and (2) to obtain

$$D(v\nabla^2 u - u\nabla^2 v) = \frac{\partial (uv)}{\partial \theta} \qquad (3)$$

Integrating Equation (3) over an arbitrary volume region (which may be time dependent) and time, and applying the well-known Green's identity (9) to the left side, one obtains

$$\iiint D(v\nabla u - u\nabla v)\cdot \bar{n}\, dS'\, d\theta' = \iiiint \frac{\partial (uv)}{\partial \theta'} dV'\, d\theta' \qquad (4)$$

In accordance with the Leibnitz rule for volume integrals (*10*)

$$\frac{d}{d\theta}\iiint (uv)\, dV = \iiint \frac{\partial (uv)}{\partial \theta} dV + \iint uv\, (\bar{v}_S\cdot \bar{n})\, dS \qquad (5)$$

Introducing Equation (5) into Equation (4) to eliminate the right side of the latter, performing the indicated time integration, and rearranging terms one obtains

$$\iiint (uv)\, dV'\Big|_{\theta'=\theta} = D\iiint \left(v\nabla u - u\nabla v + \frac{uv(\bar{v}_S)}{D}\right)\cdot \bar{n}\, dS'\, d\theta' + \iiint (uv)\, dV'\Big|_{\theta'=0} \qquad (6)$$

Treatment of Boundary Conditions

Let P denote a point in space. Most boundary conditions of physical interest may be written in terms of the mixed Dirichlet-Neumann type

$$u + F(P,\theta)\,\nabla u\cdot \bar{n} = G(P,\theta) \quad \text{for } P \text{ on } S \qquad (7)$$

and the initial condition

$$u = i(P) \quad \text{for } \theta = 0 \qquad (8)$$

The auxiliary function v has been introduced as a matter of mathematical convenience, and since it has no physical obligations to fulfill, the boundary conditions on v may be chosen in any way convenient to the analysis. In what follows, it will be convenient to write x_1', x_2', x_3', and θ' as integrating variables and to consider x_1, x_2, x_3, and θ as particular values of these variables. The point (x_1', x_2', x_3') is denoted as P'.

Let it now be required that

$$v(P',\theta) = 0 \quad \text{for } P' \neq P \qquad (9)$$

and

$$v(P,\theta) = \infty \qquad (10)$$

the singularity being of such order that

$$\lim_{\epsilon\to 0} \int_{x_3-\epsilon}^{x_3+\epsilon}\int_{x_2-\epsilon}^{x_2+\epsilon}\int_{x_1-\epsilon}^{x_1+\epsilon} v(P',\theta)\, dx_1'\, dx_2'\, dx_3' = 1 \qquad (11)$$

Under these conditions, it follows that

$$\iiint (uv)\, dV'\Big|_{\theta'=\theta} = u(P,\theta) \qquad (12)$$

Let it also be required that

$$\frac{v}{F} = -\left(\nabla v - \frac{v\,\bar{v}_S}{D}\right)\cdot \bar{n} \quad \text{on } S \qquad (13)$$

Equations (6), (7), (8), (12), and (13) may be combined to produce

$$u = D\iiint \left(\frac{vG}{F}\right)_{S'} dS'\, d\theta' + \iiint (iv)\, dV'\Big|_{\theta'=0} \qquad (14)$$

and Equation (14) represents the solution of the physical problem in terms of the auxiliary problem.

The Auxiliary Problem

For a portion of the bounding surface which is moving ($\bar{v}_S \neq 0$), it is possible to write two physical boundary conditions: an equilibrium relationship involving u, and a continuity relationship (energy or mass balance for example) involving ∇u. When both u and ∇u are specified on a portion of S, Equation (7) is not sufficient to define both arbitrary functions F and G, and one must be specified independently. This may be done by allowing Equation (13) to define F, whereupon Equation (7) defines G, and the moving-boundary conditions will be satisfied.

For the determination of v, one has the differential equation, Equation (2), and the initial condition, Equations (9), (10), and (11). Since the surface condition, Equation (13), is to be taken as the definition of F on a moving portion of S, it is clear that another, independent restriction upon v must be specified in order to define the auxiliary function uniquely. This is done as follows. The domain of the auxiliary problem is extended across the moving boundary, and the auxiliary function is required to be bounded throughout the extended domain. For example, if the physical problem involved spherically symmetrical diffusion in the region bounded externally by a spherical moving boundary, the auxiliary function would be required to be bounded for infinite radius.

Fixed Boundaries

When the bounding surface consists of fixed portions in addition to moving ones, as would be the case if a control surface or an impenetrable wall were involved, a somewhat more difficult auxiliary problem is faced. Let FBS denote the portion of S which is stationary, and MBS denote the moving-boundary surface. For a fixed-boundary surface, one generally encounters a boundary condition of one of the three following forms: u specified; ∇u specified;

or $\nabla u \cdot \bar{n} + K(u - u') = 0$, where u' is the counterpart of u in the adjacent phase. For these three conditions, respectively, one has, from Equations (7) and (13)

$$F = 0 \quad ; \quad v = 0 \qquad \text{on FBS} \quad (15)$$

$$F = \infty \text{ and } \frac{G}{F} = \bar{n} \cdot \nabla u \; ; \quad \nabla v = 0 \qquad \text{on FBS} \quad (16)$$

$$F = \frac{1}{K} \text{ and } G = u' \quad ; \quad Kv = -\bar{n} \cdot \nabla v \qquad \text{on FBS} \quad (17)$$

Equations (15) and (16) are clearly special cases of Equation (17), so that all of these conditions may be represented in terms of a single fixed-boundary auxiliary boundary equation:

$$Kv + \bar{n} \cdot \nabla v = 0; \quad 0 \leqq K \leqq \infty \quad \text{on FBS} \quad (18)$$

Derivation of the Auxiliary Functions

The auxiliary problem may now be stated, putting $t(\theta) = \theta - \theta'$, as follows:

$$D \nabla^2 v = \frac{\partial v}{\partial t} \quad (19)$$

$$v(P', 0) = 0, \quad P' \neq P \quad (20)$$

$$v(P, 0) = \infty \quad (21)$$

$$\int\int\int v(P', 0)\, dV' = 1 \quad (22)$$

$$Kv + \bar{n} \cdot \nabla v = 0, \quad 0 \leqq K \leqq \infty \quad \text{on FBS} \quad (23)$$

$$v \text{ bounded for extension across MBS} \quad (24)$$

The entire problem has now been maneuvered onto familiar territory as may be recognized. The auxiliary problems for systems involving no fixed boundaries are satisfied by the well-known source solutions of heat conduction, while those involving fixed boundaries are solved by the so-called *Green's functions* for the parabolic equation. A number of auxiliary functions, covering most geometries and boundary conditions likely to be encountered in practice, are presented in tabular form in reference *2*. Others may be found in reference *1*. The functions are, however, not difficult to derive directly for any case needed. The auxiliary problem, consisting of Equations (19) through (24), is readily solved by the method of Laplace Transforms.

Calculation of the Boundary Motion

When the auxiliary function has been determined, the group $(vG/F)_{s'}$ which appears in Equation (14), may be deduced from Equations (7) and (13). Then, if $P_s(\theta)$ denotes the locus of points on the moving-boundary surface, the solution for the intensive variable profile may be written from Equation (14). The solution is, functionally, as follows:

$$u = \int_0^\theta fct\, [P, P_s(\theta'), \bar{v}_s(\theta'), \theta, \theta']\, d\theta' \quad (25)$$

Since the boundary values of u and ∇u on the moving-boundary surface are determined by the physics of the process

$$u = f\,[P_s(\theta), \theta] \quad (26)$$

and on MBS

$$\bar{n} \cdot \nabla u = g\,[P_s(\theta), \theta] \quad (27)$$

equations for the moving boundary may be obtained by applying either, or combinations of these types of conditions to Equation (25). For example, one may write an equation corresponding to the following functional equation:

$$f\,[P_s(\theta), \theta] = \int_0^\theta fct\, [P_s(\theta), P_s(\theta'), \bar{v}_s(\theta')\, \theta, \theta']\, d\theta' \quad (28)$$

These equations describing the boundary motion are nonlinear Volterra integrodifferential equations. Their explicit formulation and solution are considered by example in a following paper.

ALTERNATE ANALYTICAL PROCEDURE FOR PROBLEMS INVOLVING CONVECTION

When the diffusion phase involves convection, Equation (1) must be replaced by

$$D \nabla^2 u - \bar{v} \cdot \nabla u = \frac{\partial u}{\partial \theta} \quad (29)$$

For the remainder of this work, it is convenient to restrict the problems under consideration to those which may be described in terms of two independent variables, a space coordinate variable s and time θ. Thus the two- and three-dimensional problems to be considered feature cylindrical and spherical symmetry. For the two-variable problem, Equation (29) becomes

$$D\, s^{1-d} \frac{\partial}{\partial s}\left[s^{d-1} \frac{\partial u}{\partial s} \right] - v_s \frac{\partial u}{\partial s} = \frac{\partial u}{\partial \theta} \quad (30)$$

The continuity equation for a constant-density phase in terms of these coordinates is

$$v_s\, s^{d-1} = h(\theta) \quad (31)$$

where $h(\theta)$ is some function of time.

Let $s = B(\theta)$ denote the position of the moving boundary. The flux of total material with respect to this boundary is

$$\rho(v_s - \dot{B}) \quad (32)$$

In most problems of practical interest, at most one of the phases in contact at the moving boundary has a nonzero velocity. Let the adjacent phases be denoted by *I* and *II*, and suppose that the velocity of phase *I* is zero. A total material balance at the moving boundary may be written as follows:

$$-\rho_I \dot{B} = \rho_{II}\, (v_{II,B} - \dot{B}) \quad (33)$$

from which

$$v_{II,B} = \epsilon \dot{B} \quad (34)$$

where ϵ is a function of the phase densities:

$$\epsilon = \frac{\rho_{II} - \rho_I}{\rho_{II}} \quad (35)$$

Eliminating $h(\theta)$ from Equation (31) by evaluation for $s = B$, one has

$$v_s\, s^{d-1} = v_B\, B^{d-1} \quad (36)$$

From Equations (34) and (35)

$$v_s = \epsilon \left(\frac{B}{s} \right)^{d-1} \dot{B} \quad (37)$$

and Equation (30) becomes

$$D\, s^{1-d} \frac{\partial}{\partial s}\left[s^{d-1} \frac{\partial u}{\partial s} \right] - \epsilon \left(\frac{B}{s} \right)^{d-1} \dot{B} \frac{\partial u}{\partial s} = \frac{\partial u}{\partial \theta} \quad (38)$$

In the course of solution of a moving-boundary problem, it is clear that one must be concerned with both the differential equation and the boundary conditions. The present section is concerned with only the first of these items, and the objective is to evolve a method of generating particular solutions to Equation (38). One may then examine any such particular solution for its behavior on the boundaries to determine if it is a suitable solution to the problem at hand.

The Method of Intermediate Integrals

It is usually possible to transform the independent variables of a problem and eliminate s and θ by transformation equations:

$$x = x(s, \theta); \quad y = y(s, \theta) \tag{39}$$

Leaving the choice of transformation open, one may write the transformed differential equation corresponding to Equation (38) as

$$\beta_1 \bar{r} + \beta_2 \bar{s} + \beta_3 \bar{t} + \beta_4 p + \beta_5 q = 0 \tag{40}$$

where

$$\bar{r} = \frac{\partial^2 u}{\partial x^2}; \; \bar{s} = \frac{\partial^2 u}{\partial x \, \partial y}; \; \bar{t} = \frac{\partial^2 u}{\partial y^2} \tag{41}$$

$$p = \frac{\partial u}{\partial x}; \; q = \frac{\partial u}{\partial y}$$

$$\beta_1 = D\left(\frac{\partial x}{\partial s}\right)^2; \beta_2 = 2D\frac{\partial x}{\partial s}\frac{\partial y}{\partial s}; \beta_3 = D\left(\frac{\partial y}{\partial s}\right)^2$$

$$\beta_4 = D\frac{\partial^2 x}{\partial s^2} + \left[\frac{D(d-1)}{s} - \epsilon(B/s)^{d-1}\dot{B}\right]\frac{\partial x}{\partial s} - \frac{\partial x}{\partial \theta}$$

$$\beta_5 = D\frac{\partial^2 y}{\partial s^2} + \left[\frac{D(d-1)}{s} - \epsilon(B/s)^{d-1}\dot{B}\right]\frac{\partial y}{\partial s} - \frac{\partial y}{\partial \theta}$$

It is now assumed that for any problem under consideration, there exists an intermediate (first) integral function (*11*):

$$G\,[x, y, p(x, y), q(x, y)] = 0 \tag{42}$$

If this is so, it follows that*

$$\left(\frac{\partial G}{\partial x}\right)_y = G_x + G_p \bar{r} + G_q \bar{s} = 0 \tag{43}$$

and

$$\left(\frac{\partial G}{\partial y}\right)_x = G_y + G_p \bar{s} + G_q \bar{t} = 0 \tag{44}$$

Then, solving Equations (43) and (44) for $\bar{r}$ and $\bar{t}$, and substituting into Equation (40), one has

$$\left(-\frac{G_x}{G_p}\beta_1 - \frac{G_y}{G_q}\beta_3 + \beta_4 p + \beta_5 q\right) + \left(-\beta_1\frac{G_q}{G_p} + \beta_2 - \beta_3\frac{G_p}{G_q}\right)\bar{s} = 0 \tag{45}$$

For this identity to be satisfied, it is necessary that

$$F_1(G_x, G_y, G_p, G_q, x, y, p, q) = \beta_1\frac{G_x}{G_p} + \beta_3\frac{G_y}{G_q} - \beta_4 p - \beta_5 q = 0 \tag{46}$$

and either

$$F_2(G_x, G_y, G_p, G_q, x, y, p, q) = \beta_1\frac{G_q}{G_p} - \beta_2 + \beta_3\frac{G_p}{G_q} = 0 \tag{47}$$

or

$$\bar{s} = \frac{\partial^2 u}{\partial x \, \partial y} = 0 \tag{48}$$

* The subscripts in Equations (43) and (44) indicate partial derivatives, with the other three independent variables held constant. For example, $G_x = \partial G/\partial x$, y, p, and q held constant.

Which of these possibilities is to be realized may be determined by computing

$$(F_1, F_2) = \frac{\partial(F_1, F_2)}{\partial(x, G_x)} + \frac{\partial(F_1, F_2)}{\partial(y, G_y)} + \frac{\partial(F_1, F_2)}{\partial(p, G_p)} + \frac{\partial(F_1, F_2)}{\partial(q, G_q)} \tag{49}$$

where each term on the right side of Equation (49) is a Jacobian determinant. If (F_1, F_2) is zero either identically or by virtue of either Equation (46) or (47) or both, then Equations (46) and (47) are consistent. One then has two simultaneous first-order partial differential equations in four independent variables. If (F_1, F_2) is not zero by the above-mentioned mechanisms, it may be possible to set $(F_1, F_2) = F_3 = 0$, so that (F_1, F_3) and (F_2, F_3) are both zero by the same mechanisms, and so on. If (F_1, F_2) cannot be made to be zero by any of these methods, then Equations (46) and (47) are inconsistent (have no common solution), so that if an intermediate integral indeed exists, Equation (48) is true while Equation (47) is false. Equation (46) is true in either case.

When Equation (48) holds, partial integrations show that the solution is of the form

$$u = f(x) + g(y) \tag{50}$$

One could introduce this form into Equation (40), note the form of $B(\theta)$ necessary for variables to be separable, and so proceed to possible useful results along these lines. A different, more unified approach to all of these cases is followed here, however.

The First-order Equations

It has been noted above that when an intermediate integral exists, Equation (46) holds. The problem may therefore be approached first by finding particular solutions of this first-order equation. One way of approaching this problem is to generate additional functions, $F_i = 0$, analogous to Equation (46) and mutually consistent [that is $(F_i, F_j) = 0$], until a total of four such equations are obtained. These functions should be such that, when solved simultaneously for G_x, G_y, G_p, and G_q in terms of x, y, p, and q, they render the differential expression

$$dG = G_x dx + G_y dy + G_p dp + G_q dq \tag{51}$$

exact and therefore integrable. It can be shown (*11*) that such functions are given by particular integrals of the following set of ordinary differential equations subsidiary to Equation (46):

$$\frac{dG_x}{\left(\frac{\partial F_1}{\partial x}\right)} = \frac{dG_y}{\left(\frac{\partial F_1}{\partial y}\right)} = \frac{dG_p}{\left(\frac{\partial F_1}{\partial p}\right)} = \frac{dG_q}{\left(\frac{\partial F_1}{\partial q}\right)} = \frac{-dx}{\left(\frac{\partial F_1}{\partial G_x}\right)} = \frac{-dy}{\left(\frac{\partial F_1}{\partial G_y}\right)} = \frac{-dp}{\left(\frac{\partial F_1}{\partial G_p}\right)} = \frac{-dq}{\left(\frac{\partial F_1}{\partial G_q}\right)} \tag{52}$$

In view of the discussion following Equation (49), it follows that from zero to three integrals may be required.

As a concluding word on analytical methods, it may be remarked that most of the closed-form analytical solutions for moving-boundary problems known to the authors (other than approximate solutions derived by the so-called *quasisteady* methods) may be found by the method of intermediate integrals. This includes, for example, the solutions of Arnold (*12*), Schwartz (*13*), Danckwerts (*3*), Crank (*14*), Jost (*15*), and Carslaw and Jaeger (*1*).

NUMERICAL SOLUTION OF MOVING BOUNDARY PROBLEMS

Perhaps the most efficient manner of solving a given moving-boundary problem involving convection is to adopt a numerical procedure at the outset. The present section is concerned with a general numerical method for solving Equation (38) subject to appropriate boundary conditions.

There have been special numerical methods developed for specific moving-boundary problems just as there have been special analytical ones. These include graphical methods based upon the Schmidt construction, relaxation methods, nomographic methods, digital computer methods, passive-element analogue computer methods, and electronic differential analyzer methods. These methods have been found in prominent papers including references *16, 17, 18, 19, 20,* and *21*.

The method to be described here is a generalization of the procedures used by Manci (*21*). In this method, the independent variables s and θ are transformed into new variables z and t in a manner such that the domain of the new variable z is independent of t. The domain of z is then discretized, and partial derivatives in z are replaced by their finite-difference approximations, thus reducing the partial differential equation in z and t to a set of simultaneous nonlinear ordinary differential equations, each member for a discrete value of z and continuous in t. This set of equations is then programed for solution with an electronic differential analyzer. The procedure will be illustrated in detail by example in Part II.

By obtaining solutions continuous in time, this procedure avoids the problems of numerical stability which may arise in the step-by-step graphical, relaxation, and digital computer methods. While the passive-element analogue methods share this advantage, they require somewhat specialized equipment, whereas the method described here may be implemented on the general purpose analogue computer.

SUMMARY

Three general mathematical approaches to the rigorous solution of moving-boundary problems have been shown. The first method is suitable for problems involving quiescent phases and leads to nonlinear integrodifferential equations for the boundary motion. The second analytical method is capable of solving problems involving convection normal to the moving boundary. This method offers no guarantee of success, but when it is successful, the solutions are likely to be obtained in a more convenient form than by the first method. Finally, a numerical method based upon finite-difference approximations and implemented on the differential analyzer was discussed. This method is capable of treating all moving-boundary problems considered to a degree of accuracy limited principally by the amount of computing equipment used.

In Parts II and III, the methods presented here will be applied to several example problems.

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NOTATION

B = moving-boundary position in the s coordinate system
$\dot{B}$ = boundary velocity
D = general diffusivity
d = dimension number; 1, 2, or 3 for one, two, or three dimensions respectively
F = function symbol
f = function symbol
G = function symbol
g = function symbol
h = function symbol
i = function symbol
K = general interphase transfer coefficient
$\bar{n}$ = unit outer normal vector from surface S
P = point in three-dimensional space
$P_S(\theta)$ = locus of points on the moving-boundary surface
p = first-order partial derivative
q = first-order partial derivative
$\bar{r}$ = second-order partial derivative
S = surface
$S(\theta)$ = surface of V
s = space coordinate
$\bar{s}$ = second-order partial derivative
t = transformed time variable
$\bar{t}$ = second-order partial derivative
u = intensive variable of a diffusion process
V = volume
$V(\theta)$ = volume of integration
v = auxiliary function
$\bar{v}$ = mass average velocity
$\bar{v}_S$ = surface velocity
v_s = component of mass average velocity in s direction
x = transformed independent variable
x_i = space coordinate
y = transformed independent variable
z = transformed independent variable
β_i = transformation coefficient
ϵ = phase density function
θ = time
ρ = mass density

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