Convective Instability in the Presence of A Catalytic Chemical Reaction:

Part II. Oscillatory Instability

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This is an extension of earlier work in which a linear analysis of hydrodynamic stability was applied to a fluid mixture contained between two horizontal planes. One species diffuses to the lower plane where it is destroyed by a rapid exothermic or endothermic catalytic reaction. In the present work, Galerkin's method has been used to study the importance of oscillatory instability in a system with the more dense species concentrated near the top plane. Results show that, for cases likely to be of physical interest, oscillatory instabilities are not expected.

In an earlier paper (*1*) the stability of a convective system in the presence of a catalytic chemical reaction was considered. In that article the principle of exchange of stabilities was assumed to be valid. In the present paper we show that this assumption can be removed and that the relevant differential equations can be solved by suitable modification of Galerkin's method and by application to slightly different boundary conditions than those of the previous work. Thus the present paper has a twofold purpose: 1. to show how the Galerkin technique can be modified to apply to problems with heterogeneous boundary conditions, and 2. to show that the assumption of a stationary mode of instability, that is, validity of the principle of exchange of stabilities is reasonable for many cases of practical interest. This conclusion removes an important restriction from our earlier results.

In the previous paper (*1*) we described in some detail the fundamental and practical motivation for study of stability in buoyant reacting systems. It is made clear there that transport to and from a catalyst surface can be profoundly affected by coupling between thermal and concentration fields in the presence of gravitation and that the coupling can have serious implications for the designer of chemical reactors. These arguments also apply to the present extension, which, as mentioned above, significantly broadens the scope of the earlier work.

The governing differential equations and the stationary solution to these equations are independent of the assumption of exchange of stabilities. Consequently these items, though essential to an understanding of the nature of the problem, are not rederived but are drawn directly from (*1*).

BACKGROUND

Though our earlier work did not allow for the possibility of oscillatory instabilities, the degree to which they may be important in problems of heat and mass transfer is not readily apparent. For example, Veronis (*2*) and Baines and Gill (*3*), employing boundary conditions different from those used here, found that oscillatory instability can occur but only for certain combinations of Prandtl and Schmidt numbers. From their results one can show that when heat transfer is stabilizing and mass transfer is destabilizing, oscillatory instability occurs only for combinations of Prandtl and Schmidt numbers which are not physically meaningful. For systems with stabilizing mass transfer and destabilizing heat transfer oscillatory instability would be expected (*2*, *3*). Such systems would probably require large heats of reaction, however, and hence were not considered here or in our previous work.

The effect of boundary conditions on the occurrence of oscillatory instability is not completely clear. In particular the differences between homogeneous and heterogeneous boundary conditions are not firmly established. If one is content to generalize from published results for homogeneous boundary conditions, certain predictions can be made. For example, Chandrasekhar (*4*) has found that alteration of the velocity boundary condition from a fixed to a free boundary had only a small numerical effect on the neutral stability curve, and oscillatory instability was more likely to occur for the case of a free velocity than for a fixed velocity boundary condition. This result is consistent with the small effect which is found when, under the assumption of exchange of stabilities, the boundary conditions are changed from fixed to free (*4* to *6*). The effect of other boundary conditions can be more important, however. It has been found (*5*, *6*) that changing the boundary conditions for temperature and concentration can have a substantial effect on the shape of the neutral stability curves when exchange of stabilities is assumed.

To simplify the analysis of oscillatory instability of the present problem we have considered stability with free velocity boundary conditions. In view of the results presented above, it seems reasonable to believe that this procedure should result in a conservative estimate of the ranges of Prandtl and Schmidt numbers for which oscillatory instability will occur for fixed velocity boundary conditions.

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THEORY

Statement of the Problem

We consider as before (*1*) a Newtonian fluid bounded by infinite horizontal planes spaced a distance d apart. Reactant A diffuses from the top plane ($z = d$) through an inert fluid layer (component B) to the bottom plane ($z = 0$) where the reaction

$$A \rightarrow \text{Products}$$

occurs. Assumptions about the physical nature of the system remain unchanged from the earlier work. These assumptions are:

1. Instantaneous reaction.
2. Irreversible reaction.
3. Products are passive.
4. A is passive within the flow space, except for its effect upon fluid density.
5. Density is a linear function of temperature and of concentration of A.
6. The Boussinesq approximation is valid.
7. The mass transfer rate is so small that it does not contribute appreciably to the bulk velocity.

The boundary conditions on the velocity are different from those used earlier. This change greatly simplifies application of the Galerkin technique. We previously employed a combination of fixed and free boundary conditions on the velocity. The present discussion is limited to free boundary conditions for velocity, meaning that at $z = 0$

$$w = \frac{\partial u}{\partial z} = \frac{\partial v}{\partial z} = 0$$

$$C = 0 \tag{1}$$

$$l_R \mathfrak{D} Q \frac{\partial C}{\partial z} = -k \frac{\partial T}{\partial z}$$

while at $z = d$

$$w = \frac{\partial u}{\partial z} = \frac{\partial v}{\partial z} = 0$$

$$C = C_d \tag{2}$$

$$T = T_d$$

The stationary state solution is identical to that given earlier [Equations (6) and (7) of (*1*)] and the Rayleigh numbers for heat and mass transfer are again related by

$$R_T = R_C \, N_{Le}{}^2 \, N_H{}^2 \tag{3}$$

when a reaction occurs.

The normal mode analysis is similar to our earlier treatment except that no specific form for time dependence is used. One eventually obtains the following equations

$$\left.\begin{aligned} \left[\frac{\partial^2}{\partial z^2} - a^2\right] \frac{\partial w}{\partial \tau} &= \left[\frac{\partial^2}{\partial z^2} - a^2\right]^2 w - R_T{}^{1/2} \, a\theta - sR_C{}^{1/2} \, a\xi \\ N_{Pr} \frac{\partial \theta}{\partial \tau} &= \left[\frac{\partial^2}{\partial z^2} - a^2\right] \theta + l_T R_T{}^{1/2} \, aw \\ N_{Sc} \frac{\partial \xi}{\partial \tau} &= \left[\frac{\partial^2}{\partial z^2} - a^2\right] \xi + l_C R_C{}^{1/2} \, aw \end{aligned}\right\} \tag{4}$$

with boundary conditions

$$z = 1: \quad w = \frac{\partial^2 w}{\partial z^2} = \theta = \xi = 0 \tag{5}$$

$$z = 0: \quad w = \frac{\partial^2 w}{\partial z^2} = \xi = 0; \quad -N_H l_R \frac{\partial \xi}{\partial z} = \frac{\partial \theta}{\partial z} \tag{6}$$

Galerkin Method

We wish to find those conditions for which a solution to Equations (4), subject to boundary conditions (5) and (6), is possible. Application of the Galerkin method (*7* to *9*) to such problems is usually restricted to systems which have homogeneous boundary conditions, boundary conditions which can be made homogeneous by a simple transformation of variables, or boundary conditions of the general form $Y' + KY = 0$, in which case nonspatial effects of the variable Y cancel. In the present problem the reaction boundary condition is heterogeneous and does not fit into any of the above three categories. However, the Galerkin procedure is readily adapted to this condition by assuming that the solution to Equations (4) to (6) can be expressed in the form

$$\begin{aligned} w(z, \tau) &= \sum_{i=1}^{M} A_i(\tau) \, w_i(z) \\ \theta(z, \tau) &= \sum_{i=1}^{M} B_i(\tau) \, \theta_i(z) + \sum_{i=1}^{M} C_i(\tau) \, \hat{\theta}_i(z) \\ \xi(z, \tau) &= \sum_{i=1}^{M} C_i(\tau) \, \xi_i(z) \end{aligned} \tag{7}$$

where the $\xi_i(z)$, $\theta_i(z)$, $\hat{\theta}_i(z)$ and $w_i(z)$ are drawn from complete systems of functions and are chosen so that

At $z = 0$	At $z = 1$
$w_i = D^2 w_i = 0$	$w_i = D^2 w_i = 0$
$\xi_i = 0$	$\xi_i = 0$
$D\theta_i = 0$	$\theta_i = 0$
$l_R N_H D\xi_i = -D\hat{\theta}_i$	$\hat{\theta}_i = 0$

With this choice all boundary conditions are satisfied and any solution can in principle be approximated. The $A_i(\tau)$, $B_i(\tau)$, and $C_i(\tau)$ represent undetermined time-dependent coefficients.

The next step is to substitute the trial functions, as given by Equations (7), into Equations (4). The residuals are made orthogonal to the trial functions by multiplying Equation (4a) by w_j, (4b) by $(\theta_j + \hat{\theta}_j)$, and (4c) by ξ_j. The equations are then integrated between $z = 0$ and $z = 1$. This procedure provides the advantage of coincidence with the usual Galerkin method (*7*) when $N_H = 0$. The resulting equations, which are listed in the Appendix, can be represented in matrix form by

$$\underline{\underline{E}} \frac{d\underline{F}}{d\tau} = \underline{\underline{G}} \, \underline{F} \tag{8}$$

Since, as will be shown later by direct computation, $\underline{\underline{E}}$ is nonsingular we write

$$\frac{d\underline{F}}{d\tau} = \underline{\underline{E}}^{-1} \underline{\underline{G}} \, \underline{F} = \underline{\underline{H}} \, \underline{F} \tag{9}$$

where $\underline{\underline{H}} = \underline{\underline{E}}^{-1} \underline{\underline{G}}$. The argument employed by Finlayson (*9*) can also be used here to show that if one expresses the time dependence of $\underline{F}$ in the form exp $(\sigma\tau)$, then the eigenvalues of $\underline{\underline{H}}$ correspond to permissible values for σ. Thus our approach is to search for the eigenvalues of $\underline{\underline{H}}$ and to ascertain the behavior of real and imaginary parts of σ for different parameters of the physical problem. This in turn will tell one whether or not an oscillatory instability is controlling.

In order to obtain numerical answers specific approximating functions must be used. The choice of approximating functions is guided by literature sources (7 to 9) and by the known solution when exchange of stabilities is invoked. Unfortunately, the choice may require considerable trial and error in order to find functions which converge satisfactorily. The following approximating functions satisfy the boundary conditions and converge reasonably well.

$$\begin{aligned} w_n(z) &= \sqrt{2}\sin n\pi z \\ \xi_n(z) &= \sqrt{2}\sin n\pi z \\ \hat{\theta}_n(z) &= -\sqrt{2}\, l_R N_H \sin n\pi z + N_H \theta_n(z) \qquad (10) \\ \theta_n(z) &= \sqrt{2}\left[\frac{\cos n\pi z}{\cos n\pi} - \frac{\cosh n\gamma z}{\cosh n\gamma}\right] \end{aligned}$$

where γ is arbitrary. Note that although the functions chosen are complete and satisfy the boundary conditions, there is no assurance that they are the best possible approximating functions.

Eigenvalues σ of $\underline{\underline{H}}$ are now found after substitution of Equation (10) into (9) and subsequent inversion and solution for eigenvalues by standard computer subroutines. For a given choice of N_{Pr}, N_{Sc}, and N_H, the critical value of R_C or, alternatively, R_T was found by finding that wave number a for which $\sigma_r = 0$; that is, a state of neutral stability. If at this condition $\sigma_i \neq 0$, then the neutral state is one which divides oscillatory disturbances which grow or decay in amplitude with time.

RESULTS AND DISCUSSION

No Reaction

Comparison with work of other authors is possible if one considers the limiting case of no reaction ($N_H = 0$), in which case R_T and R_C are independent. Nield (5) has reported the following results.

Case A	Case B
$N_H = R_T = 0$	$N_H = R_C = 0$, fluid heated from below
Boundary Conditions	Boundary Conditions:
at $z = 0,\ 1$:	$w = \dfrac{\partial^2 w}{\partial z^2} = \theta = 0$ at $z = 1$
$w = \dfrac{\partial^2 w}{\partial z^2} = \theta = 0$	$w = \dfrac{\partial^2 w}{\partial z^2} = \dfrac{\partial \theta}{\partial z} = 0$ at $z = 0$
$(R_C)_{\text{critical}} = 657.5$	$(R_T)_{\text{critical}} = 384.7$
$a = 2.22$	$a = 1.76$

In a physical sense Case B is not a special case of the system discussed here because Nield is considering a fluid which is heated from below by an external source. However, the perturbation equations for Case B correspond to a special case of the perturbation equations for the system discussed in this paper.

For Case A the approximating function for ξ [Equation (10b)] is an exact solution to the differential equation and boundary conditions. For Case B convergence of the Galerkin method was monotonic but very slow. However, for $M = 20$ we found $(R_T)_{\text{critical}} = 390.5$ and $a = 1.76$, the Rayleigh number differing from the exact result by approximately 1.5%.* Because of computer size and time limitations, a maximum value of $M = 20$ was used for all calculations.

It has been noted that γ in Equation (10d) is an arbitrary constant. Through experimentation the value $\gamma = 0.05$ was found to provide satisfactory results.

Computations for combined heat and mass transfer in the absence of chemical reaction ($N_H = 0$) were performed for the following combinations of dimensionless groups:

$$\left.\begin{array}{llll} N_{Pr} & 0.72 & 10 & 10^3 \\ N_{Sc} & 0.80 & 10^3 & 10 \end{array}\right\} -100 \leq l_T R_T \leq 390.5;$$

$$0 \leq s l_C R_C \leq 657.5$$

Results for the combination $N_{Pr} = 10^3$, $N_{Sc} = 10$ are shown in Figure 1. Note that if both concentration and temperature gradients are destabilizing, that is, both gradients contribute to $\partial\rho/\partial z > 0$, then exchange of stabilities is valid and oscillatory instabilities do not appear. However when the temperature gradient is stabilizing, oscillatory instabilities can occur. Occurrence of oscillatory instability in this region depends upon the values of N_{Pr} and N_{Sc}. For example, for $l_T R_T < 0$ oscillatory instability occurs for $N_{Pr} = 10^3$, $N_{Sc} = 10$ but does not occur for $N_{Sc} = 10^3$, $N_{Pr} = 10$ or for $N_{Pr} = 0.72$, $N_{Sc} = 0.80$. These results are in qualitative agreement with the findings of Baines and Gill (3) for slightly different boundary

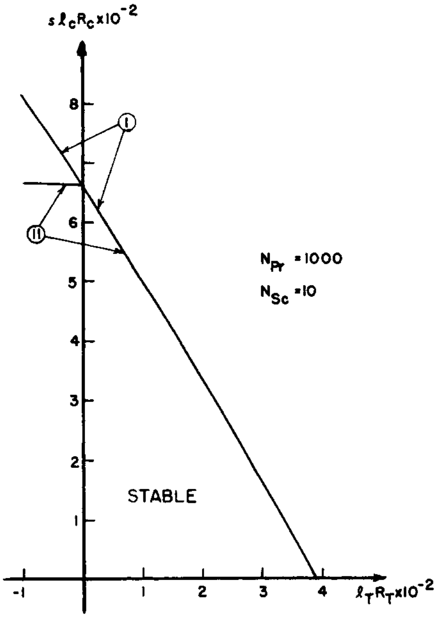

Fig. 1. Comparison of computations with and without assumption of exchange of stabilities for the case $N_H = 0$. Curve I: exchange of stabilities assumed. Curve II: exchange of stabilities not assumed.

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[*] Professor B. A. Finlayson pointed out to the authors that one can obtain better convergence properties with the trial function $\theta_n = \sqrt{2}\cos((2n-1)/2)\pi z$. The trial functions of Equation (10) are probably sufficient, however, for the task at hand, which is to ascertain those regions in which oscillatory instability may be important.

conditions. Since the case $sl_C R_C < 0$ was not considered in (*1*), we have omitted that region also from the present study.

Reaction

Recall that when a reaction occurs R_T and R_C are related by Equation (3). The methods of (*1*) may be used to show that if exchange of stabilities is valid, stability of the system is determined by A1 and A2, defined by

$$\begin{aligned} A1 &= l_T R_T + sl_C R_C \\ A2 &= l_C(l_R R_C{}^{1/2} R_T{}^{1/2} N_H - sR_C) \end{aligned} \tag{11}$$

Furthermore, the neutral curve relating A1 and A2 is immediately available from Figure 1. Replotting those results one obtains, as shown in Figure 2, the neutral curve for the free-free boundary conditions of the present problem. One could of course replot these results again in order to obtain the more physically meaningful stability envelopes analogous to those presented in (*1*). However, our present purpose is to investigate conditions under which oscillatory instability may exist. The Galerkin method described above was applied to eight combinations of N_{Pr}, N_{Sc}, and N_H, where the temperature gradient is caused by a chemical reaction so that $l_R = l_T$. Results of the computations are shown in Table 1. Under those conditions for which oscillatory instabilities do not occur, the results agree with those shown in Figure 2. It is interesting to note that no oscillatory instability was found for exothermic reactions. For endothermic reactions oscillatory instability was found only for the case $N_{Le} = 11$. The tendency toward oscillatory instability at high values of the Lewis number is consistent with the trends reported for a nonreacting system by Veronis (*2*). Since Lewis numbers are of order unity for gases and are considerably less than unity for most liquids, the results indicate that oscillatory instability is not expected for the range of parameters of physical interest as long as one deals with a destabilizing concentration profile such as we considered here.

One of the undesirable features of any application of the Galerkin method is uncertainty over the adequacy of the trial function and consequent accuracy of the final results. The present case is no exception. By setting the value of $\gamma = 0.05$ the Galerkin solution slowly and monotonically approached a limiting value of R_C from above for all eight combinations studied. The results in Table 1 are for $M = 20$ and are probably slightly higher than the correct answer. We base this prediction upon the results for the two special cases A and B cited earlier. Values of A1 and A2 for the eight examples lie between those of Case A ($A1 = -A2 = 657.5$) and Case B ($A1 = 384.7$, $A2 = 0$), or slightly to the right of Case A on the $A1 - A2$ plane of Figure 2. Recalling that the error for Case A is zero it seems reasonable to infer that the 1.5% error obtained with Case B represents an extreme case, and that the approximations used do not introduce appreciable errors.

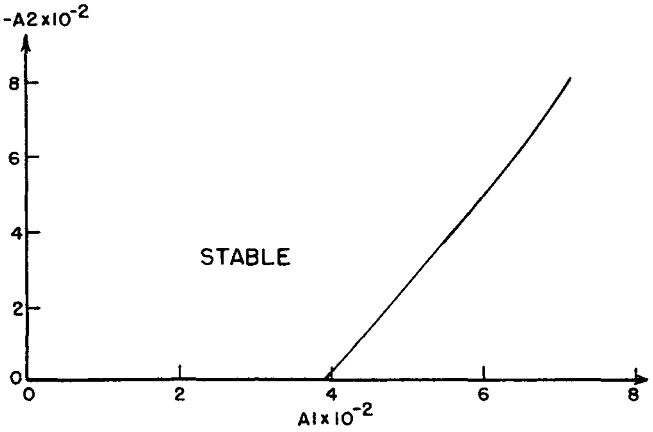

Fig. 2. Critical conditions for stability. Free velocity boundary condition, $N_H \neq 0$ exchange of stabilities assumed.

TABLE 1. SEARCH FOR OSCILLATORY INSTABILITY

N_{Le}	N_{Pr}	N_{Sc}	$l_R N_H$	a	R_C	Oscillation
0.107	1.071	10.0	1.222	2.35	700.0	no
0.251	0.226	0.900	−1.332	2.00	596.5	no
0.296	0.266	0.900	−0.608	2.15	649.1	no
0.455	0.410	0.900	−1.073	1.95	695.8	no
0.587	0.528	0.900	−0.543	2.15	699.4	no
1.279	0.640	0.500	0.591	2.15	399.5	no
7.061	6.355	0.900	0.125	1.95	400.0	no
11.0	9.9	0.900	−0.025	2.20	730.9	yes, $\sigma_i = 0.25$

CONCLUSION

We have shown that for free-free boundary conditions on the velocity a modified Galerkin method can be developed to study oscillatory instability for the problem of a catalytic reaction which occurs at the boundary of a fluid in a gravitational field. Application of the method to several specific values of Prandtl and Schmidt numbers has indicated that oscillatory instabilities are not expected for physically realizable situations when the concentration Rayleigh number is destabilizing. This conclusion is valid for both endothermic and exothermic reactions. Since other work on related problems has shown that alteration of velocity boundary conditions does not have a profound effect, it is reasonable to expect that this same result should be valid in a qualitative sense when velocity constraints at the boundary are changed to the more usual fixed conditions.

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NOTATION

a = wave number of disturbance in the xy plane, made dimensionless with d
$A_i(\tau)$ = undetermined time-dependent velocity perturbation coefficient
A1 = $l_T R_T + sl_C R_C$
A2 = $l_C(l_R R_C{}^{1/2} R_T{}^{1/2} N_H - sR_C)$
$B_i(\tau)$ = undetermined time-dependent temperature perturbation coefficient
C = concentration, mass/volume
$C_i(\tau)$ = undetermined time-dependent concentration perturbation coefficient
C_p = heat capacity per unit mass
D = d/dz
$\mathcal{D}$ = mass diffusivity

d = distance between horizontal bounding planes
$\underline{\underline{E}}$ = matrix form for coefficients on left-hand side of Equations (A1) to (A3)
$\underline{F}$ = matrix form for unknowns $A_i(\tau)$, $B_i(\tau)$, $C_i(\tau)$
$\underline{\underline{G}}$ = matrix form for coefficients on right side of Equations (A1) to (A3)
g = acceleration due to gravity
$\underline{\underline{H}}$ = $\underline{\underline{E}}^{-1}\underline{\underline{G}}$
k = thermal conductivity
l_C = ± 1 as the concentration of A decreases or increases, respectively, with z
l_R = ± 1 for an exothermic or endothermic reaction, respectively
l_T = ± 1 as the temperature decreases or increases, respectively, with z
M = limit on summation for trial functions
N_H = $\dfrac{Q\alpha_T}{\alpha_C \rho_0 C_p}$
N_{Le} = $\mathfrak{D}/\kappa$
N_{Pr} = ν/κ
N_{Sc} = $\nu/\mathfrak{D}$
Q = absolute value of heat of reaction per unit mass of reactant
R_C = $\dfrac{\alpha_C \beta_C g d^4}{\nu \mathfrak{D}}$
R_T = $\dfrac{\alpha_T \beta_T g d^4}{\kappa \nu}$
s = ± 1 as the density of the system decreases or increases, respectively, with increasing concentration C
T = temperature
t = time
u, v, w = x, y, and z components, respectively, of velocity perturbation
w_i = trial function for z component of velocity perturbation
x, y = rectangular Cartesian coordinates in a horizontal plane
z = vertical coordinate, dimensional or dimensionless. increasing in an upward direction

Greek Letters

α_C = $|\partial \ln \rho/\partial C|$
α_T = $|\partial \ln \rho/\partial T|$
β_C = $|d\overline{C}/dz|$
β_T = $|d\overline{T}/dz|$
γ = arbitrary parameter in Equation (10d), set equal to 0.05
θ = temperature perturbation
θ_i, $\hat{\theta}_i$ = trial functions for temperature perturbation
κ = thermal diffusivity = $k/(\rho_0 C_p)$
μ = viscosity
ν = kinematic viscosity = μ/ρ_0
ξ = concentration perturbation
ξ_i = trial function for concentration perturbation
ρ = density
σ = growth rate for disturbance with real and imaginary parts σ_r and σ_i, respectively. Made dimensionless with ν/d^2
τ = $t\nu/d^2$

Superscript and Subscripts

— = stationary state solution
d = evaluated at $z = d$ ($z = 1$)
0 = evaluated at $z = 0$

LITERATURE CITED

1. Wankat, P. C., and W. R. Schowalter, *AIChE J.*, **17**, 1346 (1971).
2. Veronis, G., *J. Fluid. Mech.*, **34**, 315 (1968).
3. Baines, P. G., and A. E. Gill, *ibid.*, **37**, 289 (1969).
4. Chandrasekhar, S., "Hydrodynamic and Hydromagnetic Stability," Oxford Univ. Press, London (1961).
5. Nield, D. A., *J. Fluid Mech.*, **29**, 545 (1967).
6. Wankat, P. C., Ph.D. thesis, Princeton Univ., New Jersey (1970).
7. Kantorovich, L. V., and V. I. Krylov, "Approximate Methods of Higher Analysis," (trans. C. D. Berger), Interscience, New York (1958).
8. Finlayson, B. A., and L. E. Scriven, *Appl. Mech. Rev.*, **19**, 735 (1966).
9. Finlayson, B. A., *J. Fluid Mech.*, **33**, 201 (1968).

APPENDIX

Component Equations for Matrix Equation (8)

Define the inner product

$$<u, v> = \int_0^1 uv dz$$

Then application of the expansions Equations (7) to the differential Equations (4) leads to the following 3M equations where $j = 1, \ldots, M$:

$$\sum_{i=1}^{M} <(D^2 - a^2)w_i, w_j> \frac{dA_i}{d\tau}$$

$$= \sum_{i=1}^{M} <(D^2 - a^2)^2 w_i, w_j> A_i$$

$$- R_T^{1/2} a \sum_{i=1}^{M} <\theta_i, w_j> B_i - sR_C^{1/2} a \sum_{i=1}^{M} <\xi_i, w_j> C_i$$

$$- R_T^{1/2} a \sum_{i=1}^{M} <\hat{\theta}_i, w_j> C_i \qquad \text{(A1)}$$

$$N_{Pr} \sum_{i=1}^{M} [<\theta_i, \theta_j> + <\theta_i, \hat{\theta}_j>] \frac{dB_i}{d\tau}$$

$$+ N_{Pr} \sum_{i=1}^{M} [<\hat{\theta}_i, \theta_j> + <\hat{\theta}_i, \hat{\theta}_j>] \frac{dC_i}{d\tau}$$

$$= l_T R_T^{1/2} a \sum_{i=1}^{M} [<w_i, \theta_j> + <w_i, \hat{\theta}_j>] A_i$$

$$+ \sum_{i=1}^{M} [<(D^2 - a^2)\theta_i, \theta_j> + <(D^2 - a^2)\theta_i, \hat{\theta}_j>] B_i$$

$$+ \sum_{i=1}^{M} [<(D^2 - a^2)\hat{\theta}_i, \theta_j> + <(D^2 - a^2)\hat{\theta}_i, \hat{\theta}_j>] C_i \qquad \text{(A2)}$$

$$N_{Sc} \sum_{i=1}^{M} <\xi_i, \xi_j> \frac{dC_i}{d\tau} = \sum_{i=1}^{M} <(D^2 - a^2)\xi_i, \xi_j> C_i$$

$$+ l_C R_C^{1/2} a \sum_{i=1}^{M} <w_i, \xi_j> A_i \qquad \text{(A3)}$$

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