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SOLUTION OF MIXED BOUNDARY VALUE PROBLEMS BY INTEGRAL EQUATIONS AND METHODS OF WEIGHTED RESIDUALS WITH APPLICATION TO HEAT CONDUCTION AND DIFFUSION-REACTION SYSTEMS*

P. L. MILLS† AND M. P. DUDUKOVIĆ‡

Abstract. Integral equation methods are considered as an alternate approach for the solution of mixed boundary value problems in heat conduction or diffusion with chemical reaction that can be described by dual or triple series equations. The approach is based upon reduction of the dual or triple series to a single or a set of Fredholm integral equations of the first kind whose kernel $K(x, y)$ and forcing function $f(x)$ are presented in terms of an appropriate infinite series which can be derived from the separation-of-variables solution. The solution of the integral equation for the unknown function $g(y)$ is obtained by a modified quadrature method which accounts for the presence of a logarithmic singularity in the kernel for $x = y$ which is a common occurrence in problems of this type. The approach is illustrated by solving selected example problems involving either diffusion and reaction or heat conduction. Comparisons are made to exact solutions where available and also to other approximate solutions based upon the method of weighted residuals (MWR). The results of various numerical experiments suggest that the integral equation approach yields results of the same or superior accuracy and with an order-of-magnitude less effort than those based upon MWR.

Key words. integral equations, method of weighted residuals, heat conduction, diffusion, reaction, dual series

1. Introduction. Certain types of heat or mass transfer processes which are often associated with chemical or mechanical engineering analysis can result in mixed boundary value problems. These problems arise whenever the differential boundary operator contains a discontinuity due to the specification of two or more different types of boundary conditions along a common boundary. Some particular applications which suggest the variety of systems that have recently been examined include: (i) diffusion and reaction in porous catalyst pellets which have both partial internal and external wetted surfaces [16], [18], [32], [35], [37], [42], [51], (ii) diffusion of reactants to a solid surface which contains a nonuniform distribution of catalyst [30], (iii) determination of effective diffusion coefficients for spherical catalyst pellets by interpretation of either dynamic [10] or steady-state [34] tracer gas response measurements, (iv) analysis of conduction heat transfer between solids which have partial contact along a common boundary [11], [43]–[45], and (v) fluid mechanical modeling of a particular type of Stokes flow phenomena [50]. Problems of a similar nature are also common in other branches of science and engineering such as fracture mechanics [31], [41], [55] and electrostatics [7], to name a few. Additional references regarding the origin and application of various other mixed boundary value problems are given by Kelman [27].

Despite the widespread application and growing interest in mixed boundary value problems (hereafter represented as mixed BVP) that has developed over the past eighteen years since the appearance of Sneddon's monograph [48], no unified method of solution to the various subclasses of problems, such as dual or triple orthogonal series and dual or triple integral equations, seems to exist. In the case of mixed BVP

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which are described by dual or triple orthogonal series, for example, closed-form expressions for the series coefficients exist only for a few special forms of the series kernel and modifier (cf. Kelman and Koper [24] for the definition of this terminology). These special cases apply generally to problems defined on semi-infinite domains and are not applicable to many of the more recent physical application problems cited above from the chemical or mechanical engineering literature which have been posed for mainly finite domains. In the few cases where the physical problem is described by dual orthogonal series which have a closed-form solution, special algorithms such as those developed by Kelman and co-workers [25], [26], [28] must be applied to obtain numerical results.

The need to obtain numerical results for various mixed BVP which occur in chemical and mechanical engineering applications has resulted in a number of alternate approaches. These include the application of more or less standard finite difference methods [16], [30], [32], [42], [45], the artificial interface method [9], [20]–[22], and the least squares method [24], [35]. The least squares method has also been treated as a subset of the various methods of weighted residuals for mixed BVP described by dual or triple orthogonal series [38], [39]. The extensive numerical testing of the least squares method by Kelman and Koper [24] on various dual orthogonal series as well as the convergence theorems established by Feinerman and Kelman [12] suggest that this method is perhaps the most reliable one used to date. A comparison of the results produced by the various weighted residual methods (Galerkin, collocation and least squares) for a number of dual and triple orthogonal series arising in certain chemical engineering applications supports this conclusion [38], [39].

A key result of the extensive testing cited above for the least squares method on diffusion-reaction problems is that rather large linear systems of equations ($N \geq 100$ in some cases) must be solved before an accurate approximation to the dual or triple orthogonal series coefficients is obtained. Although very good approximations for certain problems has been demonstrated for small values of N , for example, $N \leq 10$ (cf. Kelman [24], [27]), more precise results for a quantity, such as a flux evaluated at the boundary for small values of the series discontinuity c on the interval $0 \leq x \leq 1$, often require larger values of N .

Our recent growing interest in development of an alternate approach for solving some particular dual orthogonal series for diffusion-reaction systems as a means of verifying the results obtained by the least squares method has been one motivation for the present study. This approach is based upon reduction of the dual orthogonal series to a single integral equation of the first kind whose solution may be obtained numerically by application of a quadrature technique. The success of the method relies upon summation of the particular kernel series which arises in each application to a suitable closed-form. It is especially worth noting that these kernel series are not the type encountered in classic dual orthogonal series. This is because they contain certain key dimensionless groups which appear in the kernel series as a result of reaction source terms or the specification of mixed Dirichlet, Neumann, or Robin type boundary conditions. Evidence is given, based upon the results of several example problems each of which has a physical significance, that this integral equation approach is capable of yielding more accurate results with less computational effort when compared to the method of weighted residuals. The merits of using this approach for the solution of triple orthogonal series is also given. The possibilities for implementation of the methods on smaller computing devices such as hand programmable calculators or microprocessors which has already been performed previously for the least squares method [24] are suggested.

2. Formulation of the problem and integral equations. The example problems to be treated later in the paper are based generally upon a transport equation which has the following form:

$$(2.1) \quad \nabla^2 u - c^2 u = 0 \quad \text{in } D$$

where D is a region enclosed by the smooth boundary and

$$(2.2) \quad \partial D = \sum_{i=1}^m \partial D_i$$

is used to represent the boundary along which the mixed or discontinuous boundary conditions occur. The parameter m may assume values of either $m=2$ or $m=3$, corresponding to dual or triple orthogonal series respectively. The parameter c may be assigned zero, positive, or imaginary values. Laplace's equation is obtained for $c=0$ which is commonly used to describe steady-state diffusion of heat or mass in isotropic media. The case where c is imaginary yields the Helmholtz equation where $|c|$ represents the wave number. The case¹ where $c>0$ is the one which is commonly encountered in the theory of diffusion and reaction and is used to describe the concentration of a species which disappears by a first-order reaction at any location in D of a catalyst. The quantity u typically is used to denote an appropriately defined dimensionless quantity such as a dimensionless temperature $u = T/T_b$ in the case of heat conduction, or a dimensionless concentration $u = C/C_b$ in the case of mass transfer or reaction. The subscript b denotes bulk conditions such as those conditions encountered in the absence of any finite heat or mass transport resistance which otherwise might exist on the boundary ∂D .

The mixed or discontinuous boundary conditions which are commonly encountered in applications involving equation (2.1) are assumed in this work to consist of a combination of Dirichlet, Neumann, or Robin boundary conditions which can be represented in general as:

$$(2.3) \quad \lambda_i u + \nu_i \frac{\partial u}{\partial n} = \omega_i \quad \text{on } \partial D_i$$

The parameters λ_i , ν_i and ω_i are related to certain physical quantities and are defined on $(0, \infty)$ with each having a different value, some possibly zero, on each ∂D_i . Details of the meaning of these various parameters and their physical significance is omitted for brevity but can be obtained by the interested reader in monographs such as those of Özisik [40] for heat conduction, or Aris [2] for diffusion-reaction, to name a few. Specific examples of (2.1) and the associated mixed boundary conditions expressed by (2.3) for the case of dual orthogonal series are given by Kelman [20], Collins [6], and Mills and Duduković [37] corresponding to $c=0$, $c=i\omega$, and $c>0$ as described above.

The approach used by Kelman and Koper [24] in applying the least squares method to dual orthogonal series, and the present authors for both dual [38] or triple [36], [39] orthogonal series when using one of the methods of weighted residuals, is to utilize the form of the solution which satisfies the partial differential equation (2.1) and all but the mixed boundary conditions (2.3). Such a form can often be found by analytical methods such as separation-of-variables and can be represented by an infinite

¹ The appropriate designation of (2.1) for this case has been the subject of some debate [29]. Our preference is to denote it as a form of the generalized Emden-Fowler equation as given in Mehta and Aris [33, p. 612], where the choice of $f(u) = u$ is made.

series written generally as

$$(2.4) \quad u(\xi, \eta) = \sum_{n=0}^{\infty} a_n X_n(\eta) \psi_n(\xi)$$

where $X_n(\eta)$ and $\psi_n(\xi)$ are the eigenfunctions associated with the solution u in dimensionless (usually) coordinates (η, ξ) . The a_n are the dual or triple orthogonal series coefficients and are the unknowns to be determined. Application of the mixed boundary conditions given by equation (2.3) to equation (2.4) leads to the following set of either dual ($m=2$) or triple ($m=3$) orthogonal series which must be solved for the a_n :

$$(2.5) \quad \sum_{n=0}^{\infty} a_n \mu_{nj} \psi_n(\xi) = f_j(\xi), \quad \xi_{j-1} < \xi < \xi_j, \quad j=1, m.$$

In terms of the nomenclature suggested in [24], $\psi_n(\xi)$ is the series kernels, $f_j(\xi)$ are the prescribed functions on ∂D , while the μ_{nj} are the series modifiers. For the case outlined above, these are obtained by evaluation of $X_n(\eta)$ and its derivatives as indicated by (2.3) for the appropriate values of η along the boundary ∂D . Appropriate dimensionalization of ξ ensures that $\xi_0=0$ and $\xi_m=1$ so that the series kernel $\psi_n(\xi)$ is orthogonal over $(0, 1)$ with respect to some weighting function $w(\xi)$, but not over a subinterval (ξ_{j-1}, ξ_j) , in general.

Formation of the residual ε from (2.5) followed by defining the appropriate inner product allows the various methods of weighted residuals (Galerkin, collocation, and least squares) to be applied. The development of the final set of working equations for these methods is given elsewhere [24], [38], to which the interested reader is referred for details. All of the methods lead to the following infinite system of algebraic equations whose unknowns are the series coefficient a_j :

$$(2.6) \quad \sum_{j=0}^{\infty} a_j A_{ij} = F_i, \quad i=0, 1, \dots, \infty.$$

Expressions for the inner products and matrix elements A_{ij} and F_i for each method are summarized in Table 1 for reference. Closed-form expressions for the A_{ij} and F_i can be usually obtained by performing a piecewise integration over (ξ_0, ξ_m) . These have been tabulated for a variety of trigonometric series kernels, such as $\cos n\pi x$ and $\sin n\pi x$ and Legendre polynomials $P_n(\cos \theta)$ [27], [38], [39]. Computer software packages which apply the least squares method [13] or, more generally, one of the methods of weighted residuals (Galerkin, collocation, or least squares), to a user specified series kernel and series modifier [38], [39] are also available. This latter software package was used by the present authors to obtain some of the example problem results which are given later in the paper.

TABLE 1
Summary of matrix expression for MWR.

Method	Inner product	A_{ij}	F_i
Least squares	$\frac{\partial}{\partial a_n} \langle \varepsilon, \varepsilon \rangle = 0$	$\int_{\xi_0}^{\xi_m} w(\xi) \mu_{ni} \mu_{nj} \psi_i(\xi) \psi_j(\xi) d\xi$	$\int_{\xi_0}^{\xi_m} w(\xi) \mu_{ni} f_i(\xi) \psi_i(\xi) d\xi$
Galerkin	$\langle \varepsilon, w(\xi) \psi_j(\xi) \rangle$	$\int_{\xi_0}^{\xi_m} w(\xi) \mu_{nj} \psi_i(\xi) \psi_j(\xi) d\xi$	$\int_{\xi_0}^{\xi_m} w(\xi) f_i(\xi) \psi_i(\xi) d\xi$
Collocation	$\langle \varepsilon, w(\xi) \delta(\xi - \xi_i) \rangle$	$\mu_{ni} \psi_j(\xi_i)$	$f_i(\xi_i)$

An alternate approach, which is the one of prime interest in this paper, is to convert the dual or triple series expressed by (2.5) to a single or set of integral equations, respectively. The development given below corresponds specifically to dual series equations with treatment of triple series being similar. Returning to equation (2.5), it is assumed that one of the series corresponding to a particular value of j can be extended into the remaining interval denoted by index k by introduction of an unknown function $g_k(\xi)$ (to be determined) as illustrated below:

$$(2.7) \quad \sum_{n=0}^{\infty} a_n \mu_{nj} \psi_n(\xi) = \begin{cases} f_j(\xi), & \xi_{j-1} < \xi < \xi_j, \\ g_k(\xi), & \xi_{k-1} < \xi < \xi_k, \quad k = 1, 2, k \neq j. \end{cases}$$

Formation of the inner product $\langle \psi_n(\xi), \psi_n(\xi) \rangle$ over (ξ_0, ξ_2) yields the following Fredholm integral equation for the unknown function $g_k(\xi)$:

$$(2.8) \quad \int_{\xi_{k-1}}^{\xi_k} K_{kj}(\xi, \xi') g_k(\xi') d\xi' = f_k(\xi) - h_{kj}(\xi).$$

The appropriate integral equation kernel $K_{kj}(\xi, \xi')$ and forcing function $h_{kj}(\xi)$ are expressed as infinite series composed of the series kernel $\psi_n(\xi)$ and series modifiers μ_{nj} and μ_{nk} as shown below:

$$(2.9) \quad K_{kj}(\xi, \xi') = \sum_{n=0}^{\infty} \frac{\mu_{nk} \psi_n(\xi) \psi_n(\xi')}{\mu_{nj} \int_{\xi_0}^{\xi_2} \psi_n^2(\xi') d\xi'},$$

$$(2.10) \quad h_{kj}(\xi) = \int_{\xi_{j-1}}^{\xi_j} f_j(\xi') K_{kj}(\xi, \xi') d\xi'.$$

The dual orthogonal series coefficients a_n can be obtained from the following relation provided that the integral equation expressed by (2.8) can be solved to yield $g_k(\xi)$:

$$(2.11) \quad a_n = \frac{1}{\mu_{nj} \int_{\xi_0}^{\xi_2} \psi_n^2(\xi) d\xi} \left[\int_{\xi_{j-1}}^{\xi_j} f_j(\xi') \psi_n(\xi') d\xi' + \int_{\xi_{k-1}}^{\xi_k} g_k(\xi') \psi_n(\xi') d\xi' \right].$$

3. Limitations of integral equation methods. The basic notions of solving dual or triple orthogonal series by continuation of one of the series into the remaining portion of the interval by introduction of an unknown function is outlined by Sneddon [48]. This approach often relies upon an ingenious selection for the form of the continuation function $g_k(\xi)$ so that the resulting kernel series can be summed to a closed form and the integral equation has an exact inverse. Our experience in applying the methods outlined in Sneddon's monograph [48] to certain mixed boundary value problems for finite, two-dimensional domains having discontinuous boundary conditions has shown that the resulting integral equations are not of the classical type. For these cases, closed-form inverses are not readily obtained. However, for problems where a separation-of-variables solution having the form of (2.4) exists, the procedure outlined in § 2 can be applied to yield the Fredholm integral equation given by (2.8). If the kernel series given by (2.9) can be summed in closed form, or summed numerically to a specified accuracy by an appropriate series summation acceleration method [46], [47], [57], then the integral equation can usually be inverted numerically to obtain discrete solution values for $g_k(\xi)$. Cases where the kernel series cannot be numerically evaluated to a specified accuracy represent an apparent limitation of the method in its present form. For these cases, alternate methods such as finite differences, finite elements, or the method of weighted residuals, as mentioned earlier, must be used.

Finite element methods, such as those described by Finlayson [15], become necessary for problems described on irregular geometry or for problems having non-linear source terms [16], [32]. These situations, among others, represents another limitation of integral equation methods. A comparison between the finite element method and integral equation method for a linear operator problem of practical interest in heterogeneous catalysis [30] has shown that the integral equation is more efficient.

Most of the applications cited previously, including the example problems introduced in a later section, represent two-dimensional engineering type problems that have practical applications and a physical significance. These problems, including others not cited here for brevity, often involve regular geometry and linear operators having separation-of-variables solutions so that the integral equation method as outlined here can be applied. Extension to three dimensional problems is possible also but has not been studied in detail.

4. Solution of the integral equation. The integral equation given by equation (2.8) has been solved in the current work by a quadrature method and found to give reliable results. Limitations of this method and potential problems associated with solving Fredholm integral equations of the first kind are well known [3], [19] and have been the subject of a recent symposium [49] so that they will not be mentioned here. Various quadrature rules, such as trapezoidal, Simpson's, and Gaussian quadrature, have been incorporated into our integral equation computer software for comparison of performance. Numerical studies reported in the first example given in § 5 show that the error between the exact and numerical values for the dual series coefficients decreases when more accurate quadrature rules are applied. This generally agrees with results given by Baker [3].

A fairly common feature is that the kernel expression often contains a logarithmic singularity at $x = y$. Either quadrature rules designed for integrals with a logarithmic singularity can be used as described in Davis and Rabinowitz [8], or modified quadrature [3], [19] can be used. For the latter method, the following system of linear equations are obtained whose unknowns are the integral equation solution values $g_k(\xi_m)$ at the quadrature abscissas ξ_m :

$$(4.1) \quad \sum_{m=1}^N B_{mn} g_k = e_n, \quad n = 1, 2, \dots, N$$

where

$$(4.2a) \quad B_{mn} = \begin{cases} A(\xi_n) - \sum_{\substack{n=1 \\ n \neq m}}^N w_m K_{kj}(\xi_n, \xi_m), & n = m, \\ w_m K_{kj}(\xi_n, \xi_m), & n \neq m, \end{cases}$$

$$(4.2b) \quad e_n = f_k(\xi_n) - h_{kj}(\xi_n),$$

$$(4.2c) \quad A(\xi_n) = \int_{\xi_{k-1}}^{\xi_k} K_{kj}(\xi_n, \xi') d\xi'.$$

The quadrature indicated by (4.2c) represents an integrated form of the kernel. This has been performed in closed form for the example problems treated in the next section and must be treated on a problem-to-problem basis. Additional aspects related to the properties that the kernel $K_{kj}(\xi, \xi')$ and solution $g_k(\xi)$ should possess for modified quadrature to apply are discussed in the above cited references and are omitted here for brevity.

5. Numerical examples. In this section and § 6, two example problems are now considered that are typical of mixed boundary value problems in chemical engineering that lead to dual orthogonal series. The basic objective is to illustrate the application of both the method of weighted residuals and the integral equation method to these problems and determine some measure of their performance by comparison of numerical results for a few key parameters of interest.

Example 1. Heat conduction in a partially insulated plate. The first problem considers heat conduction in a semi-infinite plate. The faces at $x^* = l$ and at $y^* \rightarrow \infty$ are maintained at a prescribed temperature $T = T_\infty$. The lower edge at $y^* = 0$ is maintained at a prescribed surface temperature $T = T_s$ for $0 \leq x^* < c^*$ and is insulated for $c^* < x^* \leq l$. The line of symmetry is at $x^* = 0$. If heat generation is absent and the plate is isotropic, then setting $c = 0$ in (2.1) gives the Laplace or potential equation from which the steady-state temperature profile in the plate can be obtained.

The governing equation and associated boundary conditions are given below in terms of the dimensionless temperature difference $\theta = (T - T_\infty)/(T_s - T_\infty)$, dimensionless coordinates $x = \pi y^*/l$, $y = \pi y^*/l$, and dimensionless discontinuity $c = \pi c^*/l$:

$$(5.1) \quad \frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial y^2} = 0,$$

$$(5.2) \quad x = 0, \quad \frac{\partial \theta}{\partial x} = 0, \quad y \geq 0,$$

$$(5.3) \quad x = \pi, \quad \theta = 0, \quad y \geq 0,$$

$$(5.4a) \quad y = 0 \begin{cases} \theta = 1, & 0 \leq x < c, \\ \frac{\partial \theta}{\partial y} = 0, & c < x < \pi, \end{cases}$$

$$(5.4b) \quad y = 0 \begin{cases} \theta = 1, & 0 \leq x < c, \\ \frac{\partial \theta}{\partial y} = 0, & c < x < \pi, \end{cases}$$

$$(5.5) \quad \lim_{y \rightarrow \infty} \{\theta\} = 0, \quad 0 \leq x \leq \pi.$$

A solution which satisfies (5.1) and the boundary conditions given by (5.2), (5.3) and (5.5) is:

$$(5.6) \quad \theta(x, y) = \sum_{n=0}^{\infty} b_n e^{-(n+1/2)y} \cos \left[\left(n + \frac{1}{2} \right) x \right].$$

Application of the mixed boundary conditions (5.4a) and (5.4b) and defining a new set of coefficients $a_n = b_n(n+1/2)$ leads to the following dual series equations for determination of the series coefficients a_n :

$$(5.7a) \quad \sum_{n=0}^{\infty} \frac{a_n}{n+1/2} \cos \left[\left(n + \frac{1}{2} \right) x \right] = 1, \quad 0 \leq x < c,$$

$$(5.7b) \quad \sum_{n=0}^{\infty} a_n \cos \left[\left(n + \frac{1}{2} \right) x \right] = 0, \quad c < x \leq \pi.$$

These dual series contain one of the simplest forms for the series modifier encountered in various applications and possess a closed-form solution for the series coefficients. This example provides, therefore, an important benchmark since a direct comparison can be made between results produced by the closed-form solution and both the least squares and integral equation methods.

The dual-series given by (5.7a) and (5.7b) can be reduced to a single integral equation by following the procedure outlined in § 2. The final result is

$$(5.8) \quad \int_0^c K(x, y) g(x) dx = \frac{\pi}{2},$$

where the kernel $K(x, y)$ is given by the following infinite series which can be readily summed using a result given by Gradshteyn and Ryzhik [17, eq. 1.442-2]:

$$(5.9) \quad \begin{aligned} K(x, y) &= \sum_{n=0}^{\infty} \frac{\cos[(n+1/2)x] \cos[(n+1/2)y]}{n+1/2} \\ &= \frac{1}{2} \ln \left| \frac{\cos(x/2) + \cos(y/2)}{\cos(x/2) - \cos(y/2)} \right|. \end{aligned}$$

The kernel becomes singular as $x \rightarrow y$, so that determination of $g(x)$ by modified quadrature requires evaluation of (4.2c). The final result is

$$(5.10) \quad A(y) = \int_0^c K(x, y) dx = 2 \left[f\left(\frac{c-y}{2}\right) + f\left(\frac{c+y}{2}\right) \right],$$

where the function f is given by [52, p. 149, eq. 16]:

$$(5.11) \quad \begin{aligned} f(z) &= \sum_{n=0}^{\infty} \frac{\sin[(2n+1)z]}{2n+1} \\ &= -\frac{1}{2} \int_0^z \ln \left(\tan \frac{y}{2} \right) dy \\ &= z \ln \frac{z}{2} - z + \sum_{k=1}^{\infty} (-1)^{k+1} \frac{2^{2k+1} - 1}{k(2k+1)!} B_{2k} z^{2k+1}, \quad |z| < \pi. \end{aligned}$$

The B_{2k} , in the usual notation, denotes the Bernoulli numbers of order $2k$.

Stretched coordinates $\omega = x/c$ and $\bar{\omega} = y/c$ are introduced in the above formulas for $K(x, y)$ and $A(y)$ so that the interval of integration conveniently spans $(0, 1)$. Once the $g(c\omega_k)$ are determined, the a_n are evaluated from the following expression for the series coefficients a_n using Filon quadrature [8], [14]:

$$(5.12) \quad a_n = \frac{2}{\pi} \int_0^c g(x) \cos \left[\left(n + \frac{1}{2} \right) x \right] dx.$$

A closed-form solution for the dual-series coefficients a_n has been obtained by Tranter using two alternate approaches [53], [54]. Setting $f(x) = 1$ and using the simpler approach [54] yields the following closed-form expression for the dual series coefficients a_n :

$$(5.13) \quad a_n = \frac{P_n(\cos c)}{K(\cos^2(c/2))}, \quad n = 0, 1, 2, \dots, \infty$$

where $P_n(\cos c)$ and $K(\cos^2 c/2)$ refer to the Legendre polynomials of order n and complete elliptic integral of the first kind, respectively.

Figure 1 shows a comparison between the dimensionless temperature profile calculated by both the exact solution, integral equation method, and the least squares method for $c = \pi/180$ and $y = \pi$ for $0 \leq x \leq \pi$. Little improvement in the least squares approximation is obtained as the number of terms N is increased from $N = 5$ to

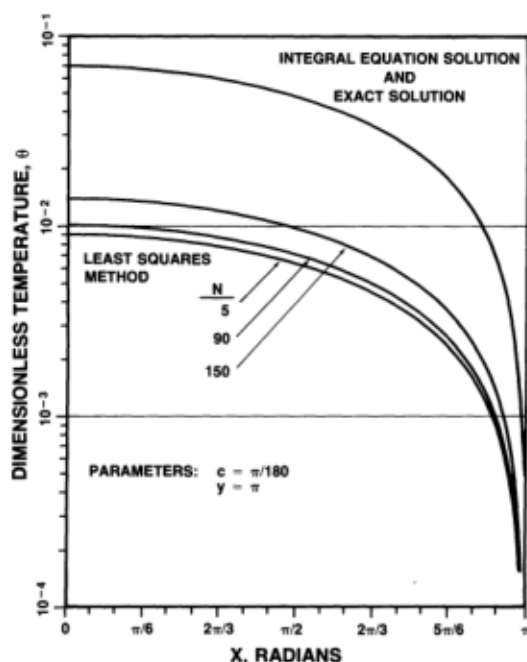


FIG. 1. Comparison between dimensionless temperature profiles calculated by the least squares method and exact solution for a fixed value of y and small c ($c = \pi/180$).

$N = 150$. Even at higher values of N , e.g., $N = 200$, the deviation is still substantial. The same comparison is made in Fig. 2 between the exact solution and the integral equation method outlined above for additional values of y . The agreement between results was within 3–4 significant digits so that the differences cannot be detected on

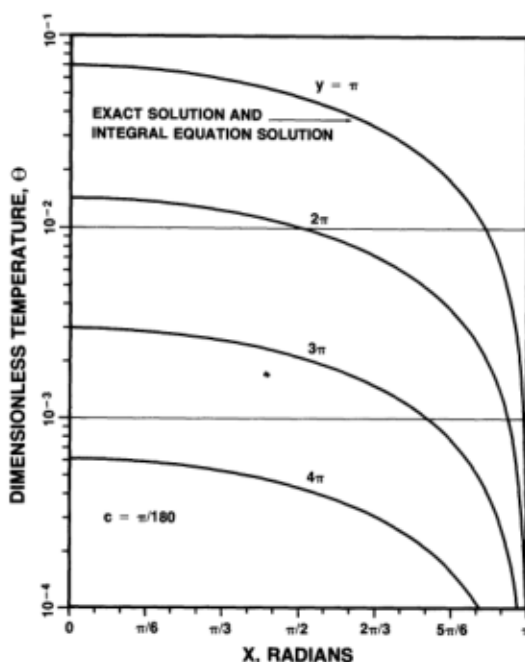


FIG. 2. Comparison between dimensionless temperature profiles calculated by the integral equation method and exact solution for various values of y and small c ($c = \pi/180$).

the figure. This represents a significant improvement when compared to the least squares results given on the previous figure. The approximate integral equation results were obtained using $N = 10$ Gaussian quadrature points so that the amount of computational effort, when compared to the least squares method at $N = 150$, was significantly less and can be readily performed on a microcomputer.

Table 2 gives a comparison between numerical values for the dual series coefficients calculated by the integral equation method, the least square method, and the exact solution for $c = \pi/180$. For comparison purposes, trapezoidal, Simpson's $\frac{1}{3}$, and Gaussian quadrature rules with $N = 9$ quadrature points were used for the integral equation method. The least square results, obtained using a matrix size of $N = 150$, still deviate significantly from the exact solution as indicated earlier in Fig. 1. The integral equation results are always in good agreement with the exact solution, irrespective of the quadrature rule used. Further reduction in the error between the exact solution and integral equation results is obtained for the more accurate quadrature rules. Results obtained using the Gaussian quadrature rule have the least error among those tested which agrees with those reported by Baker [3]. This indicates that it is not the accuracy of the quadrature rule, but the nature of the integral equation approach that is most significant.

TABLE 2
Dual-series coefficients a_n calculated by various methods for $c = \pi/180$.

n	Integral equation method ($N = 9$)			Least squares method ($N = 150$)	Exact solution
	trapezoidal	Simpson's $\frac{1}{3}$	Gaussian		
0	0.152369	0.163612	0.163058	0.330007×10^{-1}	0.16319
1	0.152346	0.163586	0.163034	0.201636×10^{-1}	0.16317
2	0.152301	0.163535	0.162985	0.175939×10^{-1}	0.16312
3	0.152232	0.163458	0.162911	0.164900×10^{-1}	0.16304
4	0.152140	0.163355	0.162813	0.158740×10^{-1}	0.16294
5	0.152026	0.163227	0.162690	0.154792×10^{-1}	0.16282
6	0.151888	0.163073	0.162543	0.152030×10^{-1}	0.16267
7	0.151728	0.162894	0.162371	0.149975×10^{-1}	0.16250
8	0.151545	0.162690	0.162175	0.148374×10^{-1}	0.16230
9	0.151339	0.162459	0.161954	0.147079×10^{-1}	0.16207

6. Example 2. Catalyst effectiveness factor in spherical ink-bottle pores. The second example is adopted from the investigation of Chu and Chon [5] which dealt with the geometry of catalyst pores and its effect on catalyst performance. The present case considers the problem of diffusion and surface reaction in a spherical ink-bottle pore whose surface is maintained at concentration C_s for $0 \leq \theta < \alpha$ but undergoes first-order surface reaction $r_s = k_s C_s$ for $\alpha < \theta \leq \pi$. The validity of assuming spherical geometry versus one having an irregular shape represents an idealization, but experimental data obtained by Broekhoff and De Boer [4] were satisfactorily interpreted using this geometry as a basis. More recent applications involving zeolite or related cage-type porous materials also support this assumption. With this supporting evidence, the governing equation and associated boundary conditions are:

$$(6.1) \quad \frac{\partial}{\partial \rho} \left(\rho^2 \frac{\partial u}{\partial \rho} \right) + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u}{\partial \theta} \right) = 0,$$

$$(6.2a) \quad \rho = 0, \quad \rho^2 \frac{\partial u}{\partial \rho} = 0,$$

$$(6.2b) \quad \left\{ \begin{array}{l} u = 1, \quad 0 \leq \theta < \alpha, \\ \frac{1}{\text{Da}} \frac{\partial u}{\partial \rho} + u = 0, \quad \alpha < \theta \leq \pi, \end{array} \right.$$

$$(6.2c) \quad \rho = 1$$

$$(6.2d) \quad \theta = 0, \pi, \quad \frac{\partial u}{\partial \theta} = 0, \quad 0 \leq \rho \leq 1.$$

The parameter Da refers to the Damköhler number [2] and is given by $\text{Da} = k_s R / D_e$ where k_s is the reaction rate constant, R is the pellet radius, and D_e is the effective diffusivity.

Solving (6.1) subject to the boundary condition given by (6.2a) and (6.2d) gives the following solution for the dimensionless concentration $u = C / C_s$:

$$(6.3) \quad u(\rho, \theta) = \sum_{n=0}^{\infty} a_n \rho^n P_n(\cos \theta)$$

where $P_n(\cos \theta)$ are the Legendre polynomials. Application of (6.2b) and (6.2c) lead to the following dual Legendre series:

$$(6.4a) \quad \sum_{n=0}^{\infty} a_n P_n(\cos \theta) = 1, \quad 0 \leq \theta < \alpha,$$

$$(6.4b) \quad \sum_{n=0}^{\infty} a_n \left(1 + \frac{n}{\text{Da}} \right) P_n(\cos \theta) = 0, \quad \alpha < \theta \leq \pi.$$

These dual Legendre series can be converted to dual cosine series using Mehler's formula for the Legendre polynomials [17, eq. 8.913] followed by inversion of the resulting Abel integral equations. The final results are:

$$(6.5a) \quad \sum_{n=0}^{\infty} b_n \frac{n+1/2}{n+\text{Da}} \cos \left[\left(n + \frac{1}{2} \right) x \right] = \frac{1}{\text{Da}} \cos \left(\frac{x}{2} \right), \quad 0 \leq x < \alpha,$$

$$(6.5b) \quad \sum_{n=0}^{\infty} b_n \cos \left[\left(n + \frac{1}{2} \right) x \right], \quad \alpha < x \leq \pi,$$

where the a_n and b_n are related by the following expression:

$$(6.6) \quad b_n = a_n \frac{1 + n/\text{Da}}{n + 1/2}.$$

Integration of (6.5a) over $(0, x)$ and following the procedure outlined in § 2 leads to the following Fredholm integral equation of the first kind:

$$(6.7) \quad \int_0^\alpha K(x, y) g(y) dy = f(x)$$

where the kernel $K(x, y)$ and forcing function $f(x)$ are given by

$$(6.8a) \quad K(x, y) = S(\text{Da}, x - y) + S(\text{Da}, x + y)$$

$$(6.8b) \quad f(x) = \frac{2\pi}{\text{Da}} \sin \left(\frac{x}{2} \right).$$

The function S introduced in equation (6.8a) is given by the following Fourier series:

$$(6.9) \quad S(a, z) = \sum_{n=0}^{\infty} \frac{\sin[(n+1/2)z]}{n+a}, \quad a > 0.$$

Tolstov's method [52] can be followed to obtain a formula for $S(a, z)$ where the remainder is $O(n^{-3})$ which is suitable for numerical evaluation to a specified accuracy.

The quantity of interest is the catalyst effectiveness factor [2], [5] which is defined as the ratio of the actual rate of reaction occurring on the surface of the active pore area to the rate of reaction that would exist if the pore surface was maintained at the surface concentration C_s . The actual rate of reaction can be evaluated by integration of the flux or by integration of the concentration profile as shown below where the subscripts f and c are used to distinguish between the two expressions:

$$(6.10) \quad \begin{aligned} \eta_f &= -\frac{1}{Da(1+\cos \alpha)} \int_{\alpha}^{\pi} \frac{\partial u}{\partial \rho} \bigg|_{\rho=1} \sin \theta \, d\theta \\ &= -\frac{1}{Da(1+\cos \alpha)} \sum_{n=1}^{\infty} a_n n \int_{-1}^{\cos \alpha} P_n(x) \, dx \\ &= -\frac{1}{2(1+\cos \alpha)} \sum_{n=1}^{\infty} b_n \frac{2n+1}{n+Da} [P_{n+1}(\cos \alpha) - \cos \alpha P_n(\cos \alpha)], \end{aligned}$$

$$(6.11) \quad \begin{aligned} \eta_c &= \frac{1}{1+\cos \alpha} \int_{\alpha}^{\pi} u(1, \theta) \sin \theta \, d\theta \\ &= \frac{1}{1+\cos \alpha} \sum_{n=1}^{\infty} a_n \int_{-1}^{\cos \alpha} P_n(x) \, dx \\ &= \frac{b_0}{2} + \frac{Da}{2(1+\cos \alpha)} \sum_{n=1}^{\infty} b_n \frac{2n+1}{n(n+Da)} [P_{n+1}(\cos \alpha) - \cos \alpha P_n(\cos \alpha)]. \end{aligned}$$

The results reported below are based upon summation (6.11) using the first 100 terms since numerical experiments showed that the calculated effectiveness factors were unaffected within 4 significant digits when additional terms were used. Equation (6.11) is preferred over equation (6.10) since the series coefficients decrease by the additional factor of n^{-1} . The dual series coefficients b_n were calculated from the calculated values for $g(y)$ by application of Filon quadrature [14] to the following equation which follows from (6.5b):

$$(6.12) \quad b_n = \frac{2}{\pi} \int_0^{\alpha} g(y) \cos \left[\left(n + \frac{1}{2} \right) y \right] dy.$$

A comparison between effectiveness factor values calculated by both the integral equation method and the method of weighted residuals for $\alpha = \pi/180$ and $Da = 0.005$ is given in Fig. 3. For the integral equation method, the number of quadrature points was varied between 2 to 18 (ordinate values corresponding to the secondary x -axis), while the matrix size N was varied between 5 to 150 for the method of weighted residuals (ordinate values corresponding to the primary x -axis). Numerical values obtained at the smallest and largest matrix sizes for each method are also given on the figure for comparison. The Galerkin method yields results substantially below those for the other methods. The results for the least squares method decrease by only 0.001 in absolute magnitude as the matrix size N is increased from 5 to 150 and overestimate the results produced by the integral equation and collocation methods. The results

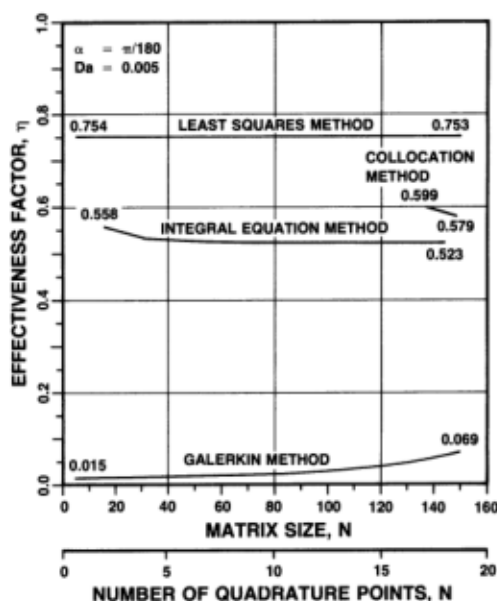


FIG. 3. Comparison between effectiveness factor values calculated by various methods as a function of the matrix size for an aperture angle $\alpha = \pi/180$.

produced by these latter two methods differ by only 0.056 which is small when compared to the others. For the collocation method, a matrix size $N = 137$ was necessary before a collocation point occurred in the range between 0 to $\pi/180$ corresponding to the range of the first dual series given by equation (6.5a). Considering that only a few collocation points occur over the range 0 to $\pi/180$ at $N = 150$, the ordinary collocation method appears to converge toward the integral equation results. Values for this latter method are relatively constant for $N \geq 4$ which represents a significant reduction in the linear system of equations when compared to $N = 150$. Chu and Chon [5] used the Galerkin method and a matrix size of $N = 48$ at this aperture angle which, in view of the above results, explains their unreasonably low values for the effectiveness factor in Fig. 1 of their original publication.

The results obtained by repeating the above exercise at a larger aperture angle ($\alpha = \pi/18$) are given in Fig. 4. The agreement between effectiveness factors calculated by the various methods represents a significant improvement when compared to the results given in Fig. 3. An interesting observation which applies to both Figs. 3 and 4 is that effectiveness factors calculated by the integral equations, least squares, and collocation methods decrease with increasing values of N , while those for the Galerkin method increase with increasing values of N . This seems to provide a means of assigning an upper and lower bounds on the exact value using the results for the integral equation method and the Galerkin method at the largest matrix size as the basis. For the case where $\alpha = \pi/18$, these bounds would be 0.9078–0.9150. The assessment of the effectiveness factor with this accuracy is more than sufficient for chemical engineering applications. The validity of the above bounds has not yet been rigorously established, however, and should be viewed as a semi-empirical result.

Application of the collocation method as described above uses collocation points θ_i in the interval $0 \leq \theta \leq \pi$ that are roots of the series kernel, namely, $P_{N-1}(\cos \theta_i) = 0$ for $i = 0, 1, \dots, N-1$ where N is the number of collocation points. This method for

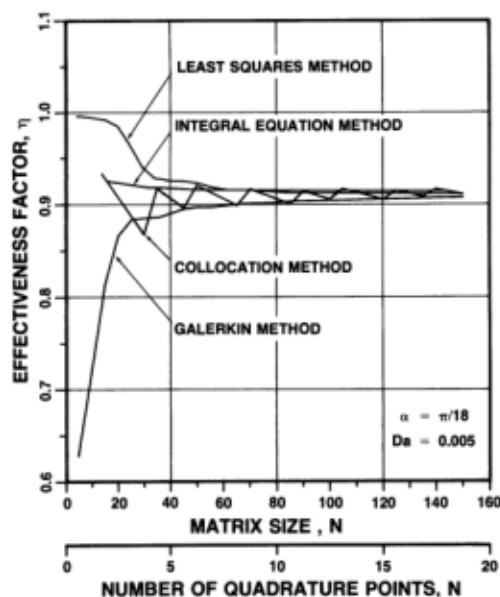


FIG. 4. Comparison between effectiveness factor values calculated by various methods as a function of the matrix size for an aperture angle $\alpha = \pi/18$.

choosing the collocation points becomes unsuitable when the discontinuity angle approaches one of the interval end points because the smaller portion of the entire interval will not contain a sufficient number of abscissas to yield a satisfactory approximation. The use of finite-element collocation [15] would divide the interval $(0, \pi)$ into NE subintervals, with each subinterval containing $N-2$ interior collocation points. In its simplest form for dual series, the interval $(0, \pi)$ could be divided into two subintervals, with the first one being defined over $(0, \alpha)$ and the second one over (α, π) . This approach should give superior results when compared to ordinary collocation over the entire interval, but the necessary development as applied to dual series equations lies outside the scope of the current work.

7. Conclusions. An alternate method for solution of dual and triple series equations has been proposed which is based upon reduction of the series to a Fredholm integral equation of the first kind. This equation is solved by a numerical quadrature method since closed-form inverses are not readily obtained or available for many problems involving practical applications.

The proposed method has been used to solve the example problems selected from applications in heat conduction and diffusion with reaction. An exact solution was available for the first example and was used for comparison to the least squares and integral equation method. It was shown that the proposed integral equation method produced results that were in good agreement with the exact solution, while those obtained by the least squares method had significant error. Similar behavior was found for a more difficult example where closed-form solutions cannot be obtained by current methods. For this example, results obtained by both the integral equation method and MWR were in better agreement for larger values of the boundary discontinuity which is typical for these and related problems examined so far.

The results of various numerical experiments suggest that the integral equation method requires the solution of a much smaller system of linear equations in comparison to MWR to achieve the same result. It was shown by the various examples that the

integral equation method yields infinite series for the kernel and forcing function whose translation to a closed-form must be treated on a problem-to-problem basis. Application of MWR to a given problem requires less effort by comparison, especially once generalized computer software is prepared.

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