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Studies on the Method of Orthogonal Collocation VII: An Efficient Collocation Method for Diffusion-convection Problems with Chemical Reaction

Moustafa A. Soliman and Khalid Alhumaizi
Chemical Engineering Department, College of Engineering, King Saud University, P.O. Box 800, Riyadh 11421, Saudi Arabia
E-mail: Humaizi@ksu.edu.sa

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Abstract. In this work we present a new numerical scheme based on orthogonal collocation method to solve a reacting flow problem. The method is based on recasting the problem to become a larger set of first order differential equations and we show by numerical computation that the application of the method of orthogonal collocation to this new set of equations leads to a more efficient numerical scheme. Numerical simulation is carried out for steady state case.

1. Introduction

A variety of numerical methods have been presented for the numerical solution of partial differential equations which involve steep spatial gradients of species moving with time (Finlayson, 1980). The problems of transient heat conduction or material diffusion in simple and complicated geometries, convection-diffusion-reaction problems in chemical reactors are examples of such problem. Finlayson (1980) presents some of the numerical methods used for solving the transient diffusion problems including finite difference, finite element and orthogonal collocation. The main task of all different techniques is to reduce the ODEs to a lower order set. This is needed in order to reduce excessive demand for computational time required by the solutions of systems with high dimensionality. The reduced system can be a good approximation to the original problem only if the parameters such as grid spacing, node distribution, etc. of the reduction technique are adequately selected.

Within the framework of the method of weighted residuals, the method of orthogonal collocation has been used to solve unsteady transport problems. The orthogonal collocation method is used to transform the partial differential equations into a set of algebraic equations that are solved by an external non-linear solver. The collocation method approximates the solution to the differential equations by a linear combination of basis functions determined by requiring that the ODE be satisfied at each of a discrete set of mesh points, and that the boundary conditions be satisfied. The orthogonal collocation method uses series expansion based on orthogonal basis functions where the coefficients are determined by the minimization of some criteria (Villadsen and Michelsen, 1978; Soliman, 2004). The collocation methods are elegant in their simplicity and efficient in their application. They have several important advantages over the other discretization methods. It provides a high order of convergence, gives a continuous approximate solutions, and easily handles general boundary conditions while still being simple to program. Several attempts have been made to draw guidelines of implementing the collocation method to chemical engineering problems (Lefervre *et al*., 2000).

Differential material balances over tubular chemical reactors are described by mathematical

model of the diffusion-convection types. This work is concerned with the numerical simulation of a reacting flow system described by a reaction-diffusion-convection model:

$$\frac{1}{Pe}\frac{d^2u}{dx^2}-\frac{du}{dx}=R(u), \qquad Pe=D/v \tag{1}$$

where v is the convection velocity, D is the diffusion coefficient, L is the reactor length, x is the dimensionless distance and R is the reaction source term. Two different mixed boundary condition cases are considered. In this work, we recast the problem to become a larger set of first order differential equations and we show by numerical computation that the application of the method of orthogonal collocation to this new set of equations leads to a more efficient numerical scheme. Numerical simulation is carried out for steady state case.

Collocation method formulation

The describing equation for tubular reactor with axial dispersion can be written as:

$$\frac{1}{Pe}\frac{d^2u}{dx^2}-\frac{du}{dx}=R(u) \tag{1}$$

These equations are usually solved by the method of orthogonal collocation with boundary points at x=0, and at x=1. The collocation points are usually chosen as the zeros of Legendre polynomials. In the following, we present the formulation for the new collocation method for two types of mixed boundary conditions.

(a) Neumann-Robbins conditions

The derivative of the dependent variable is given as a function of the dependent variable itself at one of the boundary conditions while it is constant at the other boundary condition.

$$\left.\frac{1}{Pe}\frac{du}{dx}\right|_{x=0}=u(0)-1 \tag{2}$$

and

$$\left.\frac{du}{dx}\right|_{x=1}=0 \tag{3}$$

Legendre polynomials are the most suitable polynomial for this purpose. The direct application of the orthogonal collocation method leads to:

$$\frac{1}{Pe}\sum_{j=1}^{N+2}B_{ij}u_j-\sum_{j=1}^{N+2}A_{ij}u_j=R(u_i) \qquad i=2,3,.....,N+1 \tag{4}$$

$$\frac{1}{Pe}\sum_{j=1}^{N+2}A_{1j}u_j-u_1=1 \tag{5}$$

$$\sum_{j=1}^{N+2}A_{N+2\,j}u_j=0 \tag{6}$$

It has been indicated by Michelsen (1994) that for large *Pe*, boundary condition (3) affects the concentration only close to the exit (point x=1) of the reactor. Thus, for large *Pe* we can eliminate Eq. (3). We can obtain the exit concentration by integrating Eq. (1) subject to Eqs. (2) and (3) to get:

$$u(1)=1-\int_0^1 R(u)dx \tag{7}$$

We would like to present here an approach which would replace Eq. (3) by Eq. (7) and which is suitable for any *Pe*.

First, we recast Eq. (1) into two first order boundary value problems:

$$\frac{1}{Pe}\frac{du}{dx}-u=v, \qquad u(1)+v(1)=0 \tag{8}$$

$$\frac{dv}{dx}=R(u), \qquad v(0)=v_o=-1 \tag{9}$$

To apply the method of orthogonal collocation, we notice that Eq. (8) has a boundary point at x=1, whereas Eq. (9) has a boundary point at x=0. Let $\underset{\sim}{A}$ be the matrix of first derivatives for the boundary point at x=1, and $\underset{\sim}{A}'$ be the matrix of first derivative for a boundary point at x=0. Thus, we have:

$$\frac{1}{Pe}\sum_{j=1}^{N+1}\underset{\sim}{A}_{ij}u_j-u_i=v_i \qquad i=1,2,.....,N \tag{10}$$

where

$u_{N+1}=u(1)$

$$\sum_{j=1}^{N} \underset{\sim}{A}'_{ij+1} v_j + \underset{\sim}{A}'_{i1} v_0 = R(u_i) \qquad i = 1, 2, ..., N \quad (11)$$

Notice also that:

$$\underset{\sim}{A}'_{ij+1} = -\underset{\sim}{A}_{N+1-i, N+1-j} \quad (12)$$

Using Eq. (12) in Eq. (11) and substituting Eq. (10) into Eq. (11), we obtain:

$$\frac{1}{Pe}\left[-\sum_{j=1}^{N+1} AA_{i,j} u_j \right] + \sum_{j=1}^{N} A_{n+1-i, N+1-j}\, u_j - A_{N+1-i, N+1} v_o = R(u_i) \quad (13)$$

where

$$AA_{ij} = \sum_{k=1}^{N} A_{N+1-i, N+1-k} A_{kj} \qquad \begin{matrix} i = 1, 2, ..., N \\ j = 1, 2, ..., N+1 \end{matrix} \quad (14)$$

Also, we can discretize Eq. (7) to:

$$u_{N+1} = u(1) = 1 - \sum_{i=1}^{N+1} w_i R_i \quad (15)$$

where w_i are the weights of the integral. Equations (13-15) are N+ 1 non-linear equation in N+ 1 unknown.

(b) Dirichlet-Neumann condition

For this case the value of the dependent variable is given at on the boundary condition, *x=0*.

$$u(0) = 1 \quad (16)$$

Then we can recast the problem in the following form:

$$\frac{du}{dx} = v \qquad u_o = u(0) = 1 \quad (17)$$

$$\frac{1}{Pe}\frac{dv}{dx} - v = R(u) \qquad v_{N+1} = v(1) = 0 \quad (18)$$

Now Eq. (17) has a boundary condition at x=0, while Eq. (18) has a boundary condition at x=1 so the application of the collocation method leads to:

$$\sum_{j=0}^{N} \underset{\sim}{A}'_{ij+1} u_j = v_i \qquad i = 1, 2, ..., N \quad (19)$$

or

$$-\sum_{j=0}^{N} \underset{\sim}{A}_{N+1-i, N+1-j} u_j = v_i \qquad i = 1, 2, ..., N \quad (20)$$

and

$$\frac{1}{Pe}\sum_{j=1}^{N} \underset{\sim}{A}_{ij} v_j - v_i = R(u_i) \qquad i = 1, 2, ..., N \quad (21)$$

Combining Eqs. (20) and (21), we obtain:

$$\frac{1}{Pe}\left[-\sum_{j=0}^{N} AA_{N+1-i, N+1-j}\, u_j \right] + \sum_{j=0}^{N} A_{n+1-i, N+1-j}\, u_j = R(u_i) \quad (22)$$

Thus, we need to solve Eq. (22) subject to the boundary condition Eq. (17).

The case when we have boundary conditions of given values for u(0) and u(1) was treated in references (Soliman, 1992; 2004) by spline collocation method. In the spline method, the domain is divided into two zones with each zone carrying one boundary condition. The criteria to determine the points of the division of the two zones and junction conditions were developed.

Discussion and Conclusions

The two-point collocation method in our new scheme should be compared with the one collocation point in the classical scheme because both of them represent a polynomial of second order. On the other hand, the classical collocation requires the solution of one non-linear equation and two linear equations and thus can be combined in one single non-linear equation.

The new scheme requires the solution of two non-linear equations. The third equation gives the exit concentration explicitly in terms of two interior

concentrations. Before presenting the results of implementation of the collocation schemes on our problem, we will discuss the main differences between the new method and the classical one. For one point standard collocation scheme the matrix A and B will be based on both boundary points and they will be given at the collocation point x=0.5 by:

A=[-1 0 1], B=[4 -8 4]

whereas for new formulation A and AA will be based on one boundary point only. For one collocation point x=0.5, these matrices are given by:

A=[-2 2], AA=[4 -4]

The discretized form derived by the standard method (Eq. (4)) becomes:

$$\frac{1}{Pe}(4u_o - 8u_1 + 4u_2) - (u_2 - u_o) = R(u_1)$$

One can notice in this formulation that in the convection term u_1 does not appear and this causes the profile for u to oscillate because the corresponding coefficient matrix will not be diagonally dominant. This problem has been treated by investigators for convection dominant problems using upwinding approach (Finlayson, 1992). This approach means that the upstream coefficient (in our case u_o) is larger than the downstream one (u_2). We see that Eq. (13) of the new method can be written as

$$\frac{1}{Pe}(-4u_1 + 4u_2) - 2(u_1 - 1) = R(u_1)$$

For the new approach, the coefficient of the downstream term u_2 in the convection term is zero. For large n the coefficients of the downstream terms will be smaller than the coefficients of the upstream terms, and the corresponding coefficient matrix will be diagonally dominant. Thus, the new scheme has the upwinding scheme feature which gives smooth profiles.

We can look at the new formulation in another way based on perturbation techniques (Nayfeh, 1971). Equations (9) and (17) can be considered as the regular perturbation part of the problem, whereas Eqs. (8) and (18) can be considered as the singular perturbation part of the problem. In other words, we are combining the regular solution with the singular solution. This is another reason for the power of the new scheme. In the standard formulation, Eq. (3) is imposed accurately. For large *Pe* and from the perturbation theory, we know that it only affects a very small boundary layer which enlarges as *Pe* is reduced, so trying to satisfy Eq. (3) which only affects small boundary layer causes the profile oscillation. In the new formulation and for n=1, Eq. (3) is not satisfied at all. For large n, Eq. (3) is only satisfied approximately

In the following, we apply the proposed technique on Eq. (1) for three different reaction rate expressions. The number of collocation points in the figures will be denoted with N for the classical method and NN for the new method. In the following cases, we assume that convection dominates the flow and large *Pe* can be used. *Pe* is fixed in all cases to be 1000. First we consider the Neumann-Robbins boundary conditions case with different source terms. Figure 1 shows the results for the first-order reaction source term defined by:

$$R(u) = ku$$

where k is the rate constant and its value is fixed to be 2. As we can notice from the simulation runs shown in Fig. 1 that when using two collocation points in the new scheme for this case, we find that the solution is very close to the exact solution compared with the traditional method with N=1. If we use more points, one can notice that there is no much difference in the results because the profile has a curvature near the end of the reactor that is very close to the exit no flux condition.

The results for the proposed and classical schemes for non-linear source term defined as a second-order reaction rate expression are shown in Fig. 2. The source term is defined as:

$$R(u) = ku^2$$

Again the new scheme performance is much better for the lower number of collocation points. When using four collocation points or higher, we obtain similar results for both schemes.

Figure 3 shows the simulation results for the zero order reaction scheme, where the profile is almost linear.

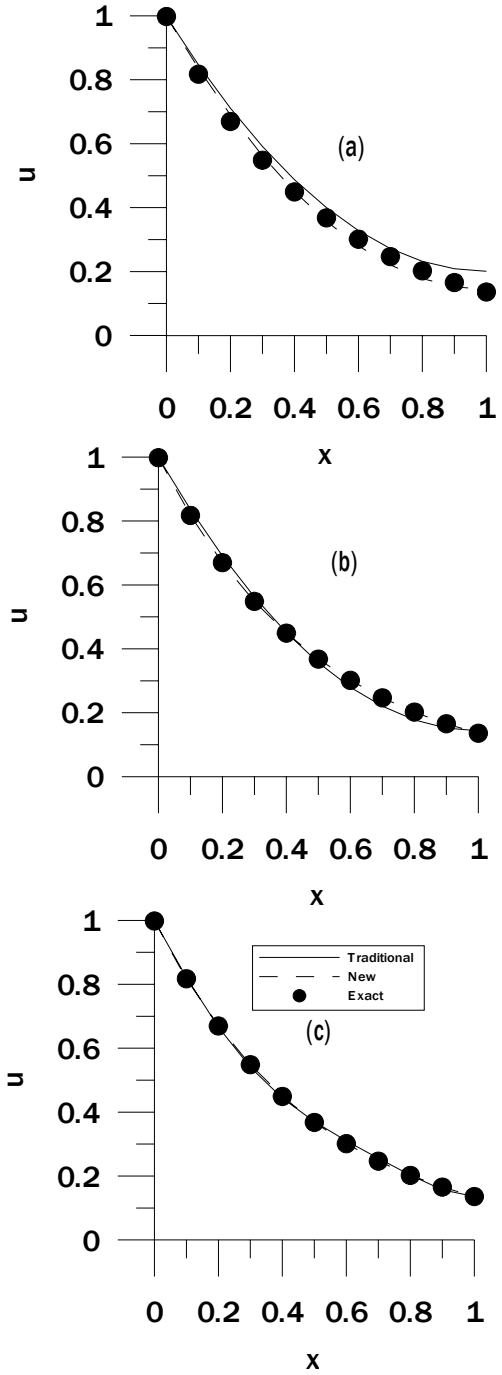

Fig. 1. Concentration profiles for the first-order reaction with k=2, Pe=1000 for (a)N=1, NN=2, (b)N=2, NN=4,(c)N=4, NN=8.

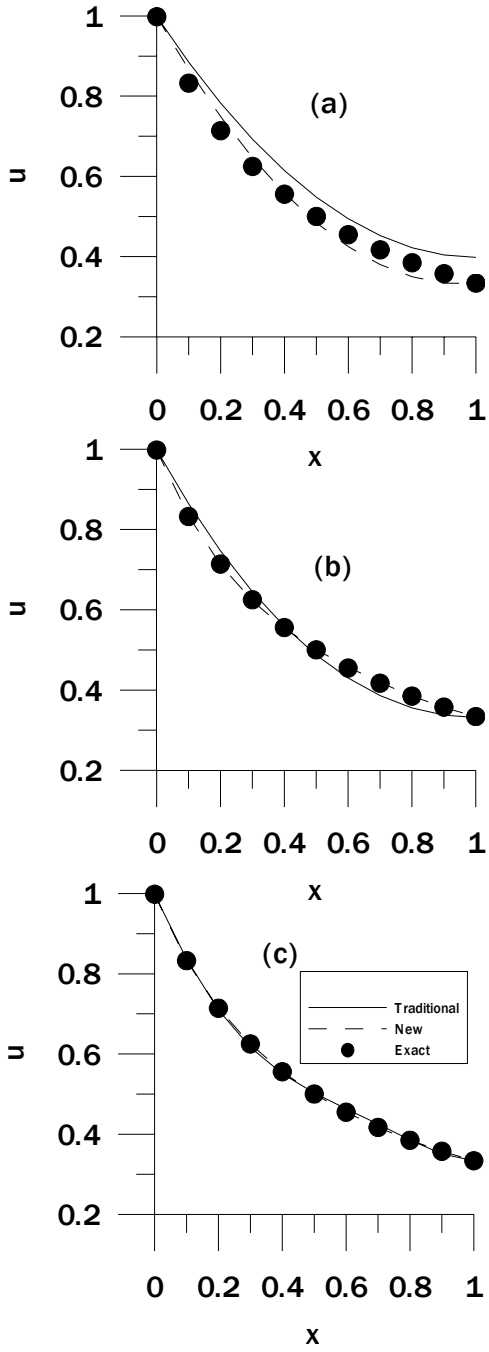

Fig. 2. Concentration profiles for the second-order reaction with k=2, Pe=1000 for (a)N=1, NN=2, (b)N=2, NN=4, (c)N=4, NN=8.

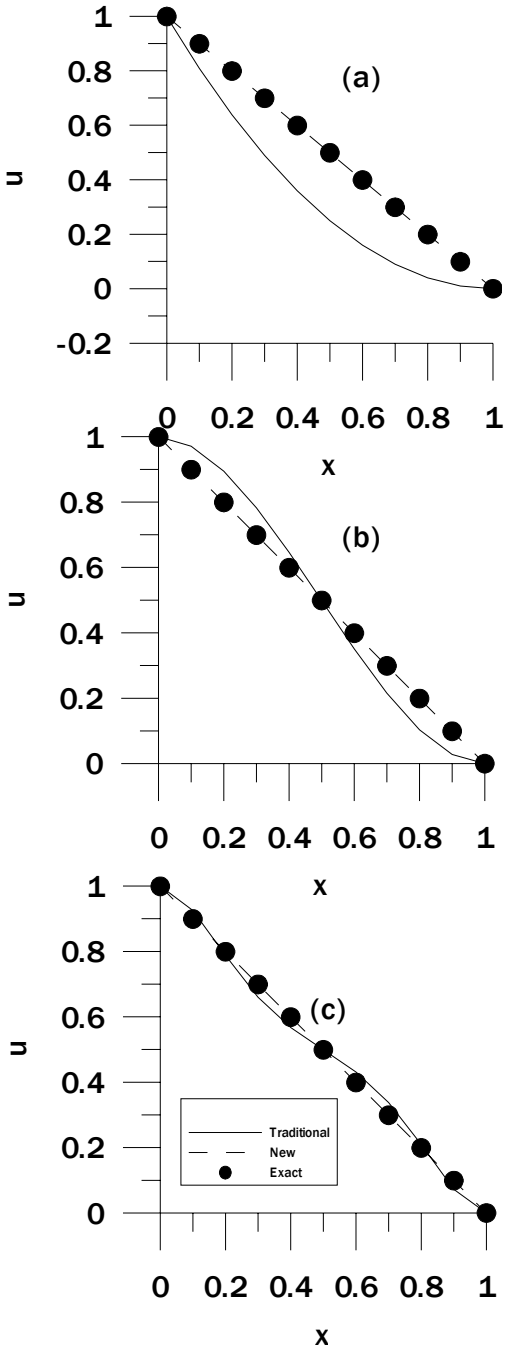

Fig. 3. Concentration profiles for the zero-order reaction with k=1, Pe=1000 for (a)N=1, NN=2, (b)N=2, NN=4, (c)N=4, NN=8.

$$R(u) = k$$

It is clear that the presented method outperforms the classical method. The one point new scheme is better than the four-point classical scheme which oscillates about the exact solution.

Figures 4-6 show the results for Dirichlet-Neumann boundary condition. The same parameters as cases 1-3 are used. It is found that highly accurate solutions are obtainable by simply increasing the number of internal collocation points. All results show that the new scheme is superior to the classical collocation method and can predict the exact solutions more accurately with lower number of collocation points.

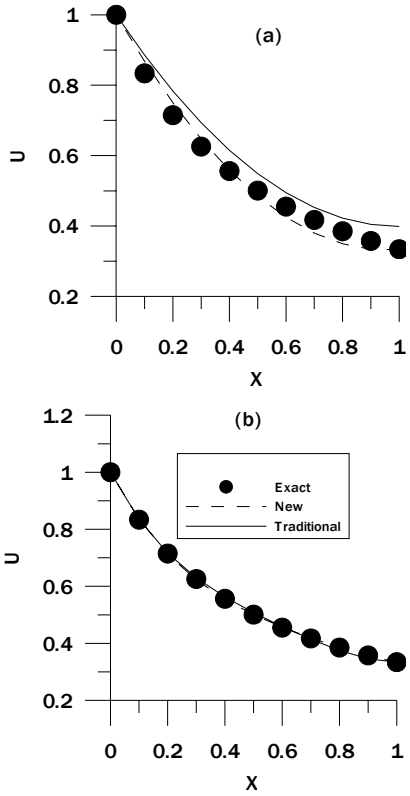

Fig. 4. Concentration profiles for the second order reaction with k=2, Pe=1000 for (a)N=1, NN=2, (b)N=2, NN=4.

Figure 7 shows the results for the zero-order reaction case with lower Pe (Pe=10). In this case, the inlet condition u(0) will be far from one. It can be seen clearly that the classical scheme with one collocation point is not able to predict u(0) accurately, while the new scheme gives accurate prediction.

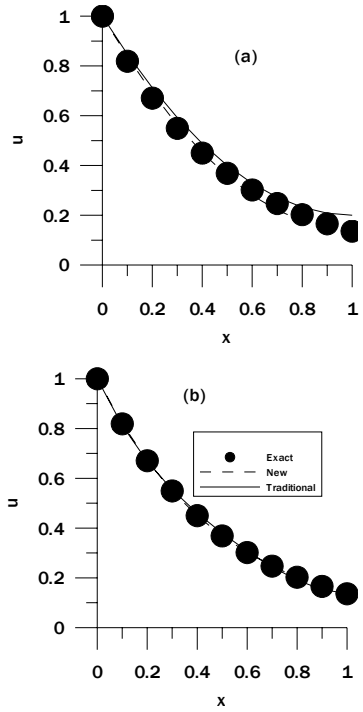

Fig. 5. Concentration profiles for first-order reaction with k=2, Pe=1000 for (a)N=1, NN=2, (b)N=2, NN=4.

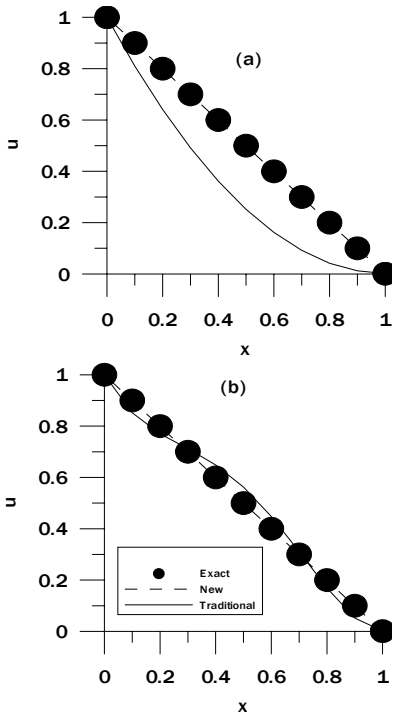

Fig. 6. Concentration profiles for zero-order reaction with k=2, Pe=1000 for (a)N=1, NN=2, (b)N=2, NN=4.

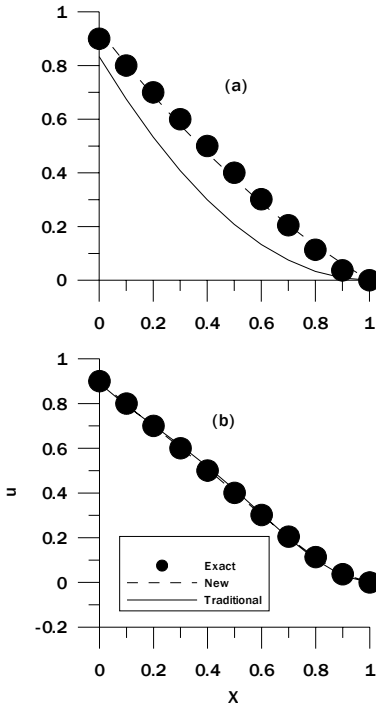

Fig. 7. Concentration profiles for zero-order reaction with k=1, Pe=10 for (a)N=1, NN=2, (b)N=2, NN=4.

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دراسات على طريقة التنظيم المتعامد ٧: طريقة تنظيم ذات كفاءة عالية لمسائل الانتشار والحمل في وجود تفاعل كيميائي

مصطفى علي سليمان، وخالد إبراهيم الحميزي

قسم الهندسة الكيميائية، كلية الهندسة، جامعة الملك سعود،
ص ب ٨٠٠، الرياض ١١٤٢١، المملكة العربية السعودية

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ملخص البحث. نقدم في هذا البحث طريقة عددية جديدة تعتمد على استخدام طريقة التنظيم المتعامد لحل مسائل السريان في وجود تفاعل كيميائي وتعتمد الطريقة على إعادة صياغة المسألة لتصبح مجموعة أكبر من المعادلات التفاضلية من الدرجة الأولى. وقد تبين من الحسابات العددية أن تطبيق طريقة التنظيم المتعامد على مجموعة المعادلات الجديدة في حالة الاستقرار تؤدي إلى طريقة عددية أكثر كفاءة من ناحية قضائها على الاهتزازات التي عادةً ما تنتج مع الطرق التقليدية.