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Approximate solution for the nonlinear model of diffusion and reaction in porous catalysts by means of the homotopy analysis method

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Abstract

In this paper, the homotopy analysis method (HAM), one of the most effective method, is applied to obtain the approximate solution of the nonlinear model of diffusion and reaction in catalyst pellets for the case of *n*th-order reactions. The approximate analytical solution obtained by HAM logically contains the solution obtained with Adomian decomposition method. The homotopy analysis method contains the auxiliary parameter $\hbar$, which provides us with a simple way to adjust and control the convergence region of solution series. By suitable choice of the auxiliary parameter $\hbar$, we can obtain reasonable solution for large Thiele modulus.

Keywords: Catalyst pellet; Diffusion and reaction; Thiele modulus; Homotopy analysis method; Soliton solution; Series solution

1. Introduction

Most of engineering problems, especially some diffusion and reaction equations are nonlinear, and in most cases it is difficult to solve them, especially analytically. Perturbation method is one of the well-known methods to solve nonlinear problems, it is based on the existence of small/large parameters, the so-called perturbation quantity [10,22]. Many nonlinear problems do not contain such kind of perturbation quantity, and we can use non-perturbation methods, such as the artificial small parameter method [21], the δ-expansion method [13], the Adomian's decomposition method [6]. However, both of the perturbation and non-perturbation methods cannot provide us with a simple way to adjust and control the convergence region and rate of given approximate series.

Also, the derivation of the Adomian decomposition method using the homotopy analysis method (HAM) is shown by Allan [7]. Adomian's decomposition method is a powerful analytic technique for strongly nonlinear problems. Adomian's decomposition method is valid for ordinary and partial differential equations, no matter whether they contain small/large parameters, and thus is rather general. Moreover, the Adomian approximation series converge quickly. However, Adomian's decomposition method has some restrictions. Approximates solutions given by Adomian's decomposition method often contain polynomials. In general, convergence regions of power series are small, thus acceleration techniques are often needed to enlarge convergence regions. This is mainly due to the fact that power series is often not an efficient set of base functions to approximate a nonlinear problem, but unfortunately Adomian's decomposition method does not provide us with freedom to use different base functions. Like the artificial small parameter method and the δ-expansion method, Adomian's decomposition method itself also does not provide us with a convenient way to adjust convergence region and rate of approximation solutions.

One of the old nonlinear problem in chemical engineering is the model of coupled diffusion and reaction in porous catalyst pellets. Thiele [27] obtained the analytical solution for first order reaction in 1939, see Refs. [8,29] for more details. However, most of their conclusions were based on first reaction order. Some authors, like Satterfild [24], have considered the *n*th-order reactions, but without obtaining the approximate solutions. Also, there are many numerical algorithms for solving the nonlinear models of fluid flow, heat transfer and chemical reactor. But the difficulty is that, most of them are dependent to initial guess and the reaction rate and indeed they cannot give analytical expression as solution.

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Currently, homotopy analysis method is employed for the analytic solution, which introduced first by Liao [15]. This method which has been successfully applied to many nonlinear problems in engineering and science, the steady-state laminar viscous flows past a sphere (governed by nonlinear Navier–Stokes equations) [14], the magnetohydrodynamic flows of non-Newtonian fluids over a stretching sheet [16], boundary-layer flows over an impermeable stretched plate [17], unsteady boundary-layer flows over a stretching flat plate [18], exponentially decaying boundary layers [19], nonlinear model of combined convective and radiative cooling of a spherical body [20], the unsteady magnetohydrodynamic flows of non-Newtonian fluids [31], the unsteady mixed convection flow on a heated vertical flat plate in a porous medium [9]. All of these successful applications verified the validity, effectiveness and flexibility of the HAM. Also, many types of nonlinear problems were solved with HAM by others [1–5,11,12,23,26,28,30].

Recently, the nonlinear model of diffusion and reaction in porous catalysts was considered by Sun et al. [25], by using the Adomian decomposition method. This method is special case of HAM [1,15], which we show later. By HAM, we can obtain the reasonable solution even for large Thiele modulus.

2. The model of diffusion and reaction

The predict of diffusion and reaction rates in porous catalysts in an important problem in chemical engineering, indeed when the reaction rate depends on concentration in a nonlinear case. In this heterogeneous system, the system is constructed as simple diffusion using an effective diffusion coefficient. Suppose that for diffusion, all the microscopic details of the porous medium are lumped together into the effective diffusion coefficient D_e for reactant. With this approximation a mass balance on a volume of the porous medium gives

$$\frac{\partial c'}{\partial t} = \nabla D_e \nabla c' - r(c'), \tag{1}$$

where t is the time, c' the chemical reactant concentration, and $r(c')$ the rate of reaction per unit volume. Now, suppose that the diffusion occurs at a steady state in a porous slab, which is infinite in two directions, giving a large plane sheet with diffusion through the thickness of the sheet. We simplify the Eq. (1) to one dimensional case when D_e is constant, to give

$$\frac{d^2c'}{dx'^2} - \frac{r(c')}{D_e} = 0, \tag{2}$$

where x' is the diffusion distance.

Now, we consider here one side of the slab as impermeable and the concentration is held fixed at the other side [25]. The two boundary conditions, where l is catalyst pore length, are given by

$$x'(0) = 0, \quad -D_e\frac{dc'}{dx'} = 0, \tag{3}$$

and

$$x'(1) = l, \quad c' = c'_s. \tag{4}$$

As Ref. [25], we consider the reaction A → B, with the rate depending on the nth power of concentration of A, depend by kc'^n, where the reaction constant k is a function of temperature. Now, we want to predict the overall reaction rate, or the mass transfer in and out of the catalyst pellet. By change of variable $x = x'/l$ and $u = c'/c'_s$, we have the following dimensionless equation derived from Eqs. (2)–(4) as

$$\frac{d^2u}{dx^2} - \varphi^2 u^n = 0, \tag{5}$$

with boundary conditions

$$\left.\frac{du}{dx}\right|_{x=0} = 0, \quad u(1) = 1, \tag{6}$$

where the Thiele modulus, $\varphi^2 = k(0)l^2u^{n-1}(0)/D_e$. It is pointed that, the Eqs. (1), (2) and (5) are valid under isothermal conditions and when there are no convection boundary effects and no coupling between chemical reaction and diffusion. The relative importance of the diffusion and reaction phenomena is measured by the Thiele modulus.

3. Basic idea of HAM

In this paper, we apply the homotopy analysis method [15,18–20] to the discussed problem. To show the basic idea, let us consider the following differential equation

$$\mathcal{N}[w(x)] = 0,$$

where $\mathcal{N}$ is a nonlinear operator, x denotes independent variable, $w(x)$ is an unknown function, respectively. For simplicity, we ignore all boundary or initial conditions, which can be treated in the similar way. By means of generalizing the traditional homotopy method, Liao [15] constructs the so-called zero-order deformation equation

$$(1-p)\mathcal{L}[\phi(x;p) - w_0(x)] = p\hbar H(x)\mathcal{N}[\phi(x;p)], \tag{7}$$

where $p \in [0, 1]$ is the embedding parameter, $\hbar \neq 0$ is a nonzero auxiliary parameter, $H(x) \neq 0$ is an auxiliary function, $\mathcal{L}$ is an auxiliary linear operator, $w_0(x)$ is an initial guess of $w(x)$, $\phi(x; p)$ is a unknown function, respectively. It is important, that one has great freedom to choose auxiliary things in HAM. Obviously, when $p = 0$ and $p = 1$, it holds

$$\phi(x;0) = w_0(x), \quad \phi(x;1) = w(x),$$

respectively. Thus, as p increases from 0 to 1, the solution $\phi(x; p)$ varies from the initial guess $w_0(x)$ to the solution $w(x)$. Expanding $\phi(x; p)$ in Taylor series with respect to p, one has

$$\phi(x;p) = w_0(x) + \sum_{m=1}^{+\infty} w_m(x)p^m, \tag{8}$$

where

$$w_m(x) = \frac{1}{m!}\left.\frac{\partial^m\phi(x;p)}{\partial p^m}\right|_{p=0}. \tag{9}$$

If the auxiliary linear operator, the initial guess, the auxiliary parameter $\hbar$, and the auxiliary function are so properly chosen, the series (8) converges at $p = 1$, one has

$$w(x) = w_0(x) + \sum_{m=1}^{+\infty} w_m(x),$$

which must be one of solutions of original nonlinear equation, as proved by Liao [15].

According to the definition (9), the governing equation can be deduced from the zero-order deformation equation (7). Define the vector

$$\vec{w}_k = \{w_0(x), w_1(x), \cdots, w_k(x)\}.$$

Differentiating Eq. (7) m times with respect to the embedding parameter p and then setting $p = 0$ and finally dividing them by $m!$, we have the so-called mth-order deformation equation

$$\mathcal{L}[w_m(x) - \chi_m w_{m-1}(x)] = \hbar H(x) R_m(\vec{w}_{m-1}), \tag{10}$$

where

$$R_m(\vec{w}_{m-1}) = \frac{1}{(m-1)!} \left. \frac{\partial^{m-1} \mathcal{N}[\phi(x; p)]}{\partial p^{m-1}} \right|_{p=0}, \tag{11}$$

and

$$\chi_m = \begin{cases} 0, & m \leq 1, \\ 1, & m > 1. \end{cases}$$

It should be emphasized that $w_m(x)$ for $m \geq 1$ is governed by the linear Eq. (10) with the linear boundary conditions that come from original problem, which can be easily solved by symbolic computation software such as Maple and Mathematica.

4. The solution with HAM

4.1. First approach

From (5) and (6), it is obvious that $u(x)$ can be expressed by the power series

$$u(x) = c_0 + \sum_{m=1}^{+\infty} c_m x^{2m},$$

where c_m is coefficient to be determined. In first approach, we assume $u(x) = \gamma w(x)$, where $\gamma = c_0 = u(0)$. Hence, the original Eqs. (5) and (6) become

$$w''(x) - \varphi^2 \gamma^{n-1} w^n(x) = 0, \tag{12}$$

with the boundary conditions

$$w(0) = 1, \quad w'(0) = 0, \tag{13}$$

and

$$\gamma w(1) = 1, \tag{14}$$

where $u(0) \neq 0$ and the prime denotes the differentiation with respect to x.

According to the Eq. (12) and the boundary conditions (13), the solution can be expressed by

$$w(x) = 1 + \sum_{m=1}^{+\infty} d_m x^{2m}, \tag{15}$$

where $d_m (m = 1, 2, \ldots)$ are coefficients to be determined.

Due to the boundary conditions (13) and under the *Rule of Solution Expression* denoted by (15), it is natural to choose $w_0(x) = 1$ as the initial approximation of $w(x)$. Under the Rule of Solution Expression, it is obvious to choose the auxiliary linear operator

$$\mathcal{L}[\phi(x; p)] = \frac{\partial^2}{\partial x^2} \phi(x; p),$$

with the property

$$\mathcal{L}[C_1 + C_2 x] = 0,$$

where C_1 and C_2 are constants.

From (12), we define a nonlinear operator

$$\mathcal{N}[\phi(x; p)] = \frac{\partial^2 \phi}{\partial x^2} - \varphi^2 \gamma^{n-1} \phi^n(x; p), \tag{16}$$

and then construct such a homotopy

$$\mathcal{H}[\phi(x; p)] = (1 - p)\mathcal{L}[\phi(x; p) - w_0(x)] - p\hbar H(x) \mathcal{N}[\phi(x; p)],$$

where $\hbar$ is a nonzero auxiliary parameter. Setting $\mathcal{H}[\phi(x; p)] = 0$, we have the *zero-order deformation equation*

$$(1 - p)\mathcal{L}[\phi(x; p) - w_0(x)] = p\hbar H(x) \mathcal{N}[\phi(x; p)], \tag{17}$$

subject to the boundary conditions

$$\phi(0; p) = 1, \qquad \left. \frac{\partial \phi(x; p)}{\partial x} \right|_{x=0} = 0, \tag{18}$$

where $p \in [0, 1]$ is an embedding parameter. When the parameter p increases from 0 to 1, the solution $\phi(x; p)$ varies from $w_0(x)$ to $w(x)$. Differentiating Eqs. (17) and (18) m times with respect to p then setting $p = 0$ and finally dividing them by $m!$, we gain the *mth-order deformation equation*

$$\mathcal{L}[w_m(x) - \chi_m w_{m-1}(x)] = \hbar H(x) R_m(\vec{w}_{m-1}(x)), \tag{19}$$

with boundary conditions

$$w_m(0) = w'_m(0) = 0. \tag{20}$$

Due to the Rules of Solution Expression (15), the auxiliary function $H(x)$ can be either in the form $H(x) = x^{\alpha}$, where α is coefficient to be determined by the so-called *Rule of Coefficient Ergodicity*, i.e. all coefficients in (15) can be modified as the order of approximation tends to infinity. Under the Rule of Coefficient Ergodicity, our calculation indicate that, for all equations under consideration, $\alpha = 0$ for the Rule of Solution Expression (15).

Obviously, the solution of Eq. (19) is

$$w_m(x) = \chi_m w_{m-1}(x) + \hbar \int_0^x \int_0^{\eta} R_m[\vec{w}_{m-1}(\xi)] \mathrm{d}\xi \, \mathrm{d}\eta + C_1 + C_2 x, \tag{21}$$

where the integral constants C_1 and C_2 are determined by the boundary conditions (20).

In this way, we get $w_m(x)$ for $m = 1, 2, 3, \ldots$, successively and at the Mth-order approximation, we have the analytic solution of Eq. (12):

$$w(x) \approx W_M(x) = \sum_{m=0}^{M} w_m(x), \tag{22}$$

and we can determine γ from boundary condition (14), i.e.

$$\gamma w(1) \approx \gamma W_M(1) = 1.$$

4.2. Second approach

Now, we define a nonlinear operator

$$\mathcal{N}[\phi(x; p), \Gamma(p)] = \frac{\partial^2 \phi}{\partial x^2} - \varphi^2 \Gamma^{n-1}(p)\phi^n(x; p), \tag{23}$$

and then construct such a homotopy

$$\mathcal{H}[\phi(x; p), \Gamma(p)] = (1 - p)\mathcal{L}[\phi(x; p) - w_0(x)] - p\hbar H(x)\mathcal{N}[\phi(x; p), \Gamma(p)],$$

where $\hbar$ is a nonzero auxiliary parameter. Setting $\mathcal{H}[\phi(x; p), \Gamma(p)] = 0$, we have the *zero-order deformation equation*

$$(1 - p)\mathcal{L}[\phi(x; p) - w_0(x)] = p\hbar H(x)\mathcal{N}[\phi(x; p), \Gamma(p)], \tag{24}$$

subject to the boundary conditions (18). When $p = 0$ and 1, the above equation has the solution

$$\phi(0; p) = w_0(x),$$

and

$$\phi(x; 1) = w(x), \quad \Gamma(1) = \gamma.$$

When the parameter p increases from 0 to 1, the solution $\phi(x; p)$ varies from $w_0(x)$ to $w(x)$, so does the $\Gamma(p)$ from γ_0 to γ. If this continuous variation is smooth enough, the Maclaurin's series with respect to p can be constructed for $\phi(x; p)$ and $\Gamma(p)$, and further, if these two series are convergent at $p = 1$, we have

$$w(x) = w_0(x) + \sum_{m=1}^{+\infty} w_m(x), \qquad \gamma = \gamma_0 + \sum_{m=1}^{+\infty} \gamma_m,$$

where

$$w_m(x) = \frac{1}{m!} \left.\frac{\partial^m \phi(x; p)}{\partial p^m}\right|_{p=0}, \qquad \gamma_m = \frac{1}{m!} \left.\frac{\partial^m \Gamma(p)}{\partial p^m}\right|_{p=0}.$$

Differentiating Eq. (24) and its boundary conditions (18) m times with respect to p then setting $p = 0$ and finally dividing them by $m!$, we gain the *mth-order deformation equation*

$$\mathcal{L}[w_m(x) - \chi_m w_{m-1}(x)] = \hbar H(x) R_m[\vec{w}_{m-1}(x), \vec{\gamma}_{m-1}], \tag{25}$$

with boundary conditions (20), where

$$\vec{\gamma}_k = \{\gamma_0, \gamma_1, \cdots, \gamma_k\}.$$

Due to the Rules of Solution Expression, as before we get $H(x) = 1$. Note that as before, we still have the freedom to choose the value of the auxiliary parameter $\hbar$. Also, the solution of Eq. (25) is

$$w_m(x) = \chi_m w_{m-1}(x) + \hbar \int_0^x \int_0^\eta R_m[\vec{w}_{m-1}(\xi), \vec{\gamma}_{m-1}]\mathrm{d}\xi\,\mathrm{d}\eta + C_1 + C_2 x, \tag{26}$$

where the integral constants C_1 and C_2 are determined by the boundary conditions (20). As before, we choose $w_0(x) = 1$ and hence, we get $w_m(x)$ for $m = 1, 2, 3, \ldots$, successively and at the Mth-order approximation, therefore we have the analytic solution of Eq. (12):

$$w(x) \approx W_M(x) = \sum_{m=0}^{M} w_m(x), \qquad \gamma \approx \Gamma_M = \sum_{m=0}^{M} \gamma_m, \tag{27}$$

and we can determine γ from boundary condition (14), i.e.

$$\gamma w(1) \approx \Gamma_M W_M(1) = 1,$$

hence, $\gamma_0 = 1$ and $\gamma_1, \gamma_2, \ldots$ are obtained successively.

5. Results

5.1. First approach

According to Section 4.1, we should ensure that the solution series (15) converges. Note that this series contains the auxiliary parameter $\hbar$, which influence its convergence region and rate. We should therefore focus on the choice of $\hbar$. When $\hbar = -1$ the expression (22) is as the same series solution obtained by Adomian decomposition method, which obtained by Sun et al. [25]. Thus, the homotopy analysis solution logically contains the Adomian decomposition solution [15].

As pointed by Liao [15], the auxiliary parameter $\hbar$ can be employed to adjust the convergence region of the series (22) in homotopy analysis solution. In general, by means of the so-called $\hbar$-curve, it is straightforward to choose an appropriate range for $\hbar$ which ensure the convergence of the solution series. Fig. 1 shows the $u(0) = \gamma$ under different $\hbar$, in case of $n = 1$ and $\varphi = 10$. For showing the efficiency of HAM, we choose

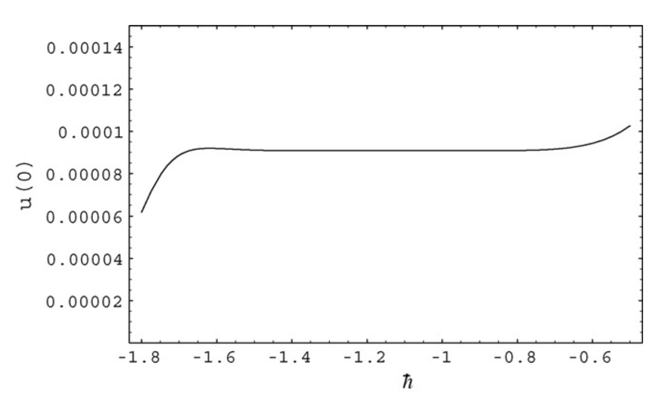

Fig. 1. $u(0)$ with different $\hbar$, in case of $n = 1$ and $\varphi = 10$ at the 15th order of approximation.

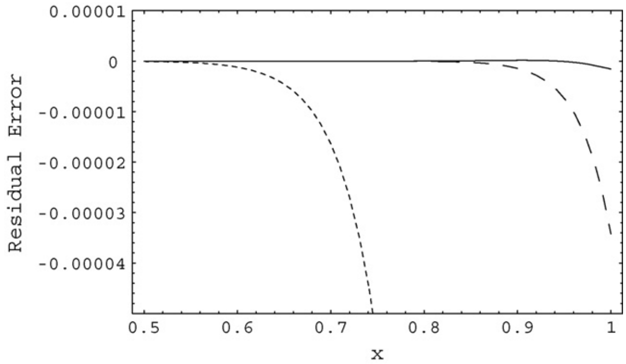

Fig. 2. Error of 15th-order HAM solution in case of $n = 1$ and $\varphi = 10$, solid line: $\hbar = -1.1$; dotted line: $\hbar = -0.9$; dashed line: $\hbar = -1$.

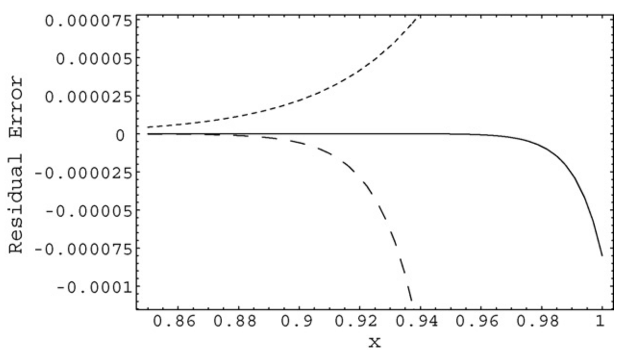

Fig. 3. Error of 35th-order HAM solution in case of $n = 2$ and $\varphi = 10$, solid line: $\hbar = -1.3$; dotted line: $\hbar = -1.4$; dashed line: $\hbar = -1$.

large value for φ in our examples. It is seen that convergent results can be obtained when $-1.4 \lesssim \hbar \lesssim -0.8$. Thus, we can choose an appropriate value for $\hbar$ in this range to get convergent solution. We can investigate the influence of $\hbar$ on the r esidual error defined as

$$\text{Residual error} \approx W''_M(x) - \varphi^2\gamma^{n-1}W^n_M(x)$$

in Fig. 2. We can see that the best value of $\hbar$ in this case is not -1. Figs. 3–7 show the results of HAM for different values of n and φ. Also in Fig. 4, the numerical solution obtained by *finite difference method* that implements the three-stage Lobatto

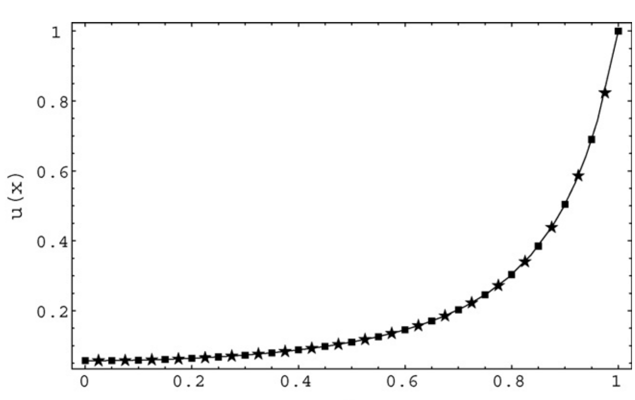

Fig. 4. Approximation of $u(x)$ by HAM in case of $n = 2$, $\varphi = 10$ and $\hbar = -1.3$, solid line: 35th-order approximation; symbols star: 20th-order approximation; symbols box: finite difference method.

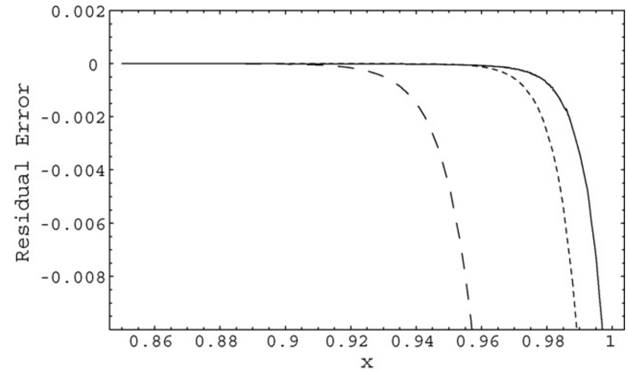

Fig. 5. Error of 50th-order HAM solution in case of $n = 2$ and $\varphi = 20$, solid line: $\hbar = -1.4$; dotted line: $\hbar = -1.3$; dashed line: $\hbar = -1$.

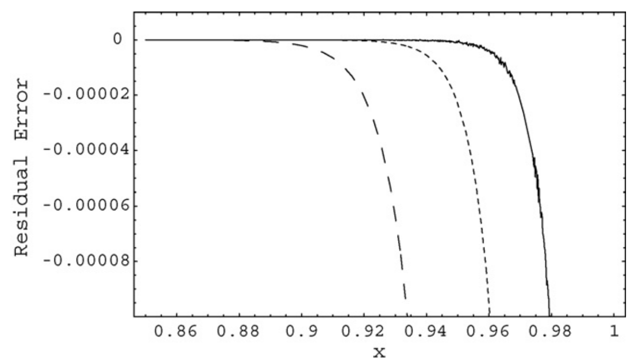

Fig. 6. Error of 50th-order HAM solution in case of $n = 4$ and $\varphi = 10$, solid line: $\hbar = -1.4$; dotted line: $\hbar = -1.2$; dashed line: $\hbar = -1$.

formula. This is a collocation formula and the collocation polynomial provides a C_1-continuous solution that is fourth order accurate uniformly in [0, 1]. Mesh selection and error control in this method are based on the residual of the continuous solution.

Also, Table 1 shows the obtained value of $u(0)$ in these examples by HAM and finite difference method, which M shows the order of approximation and the reported $\hbar$ is for smaller residual error case. Another values of n and φ are illustrated in second approach.

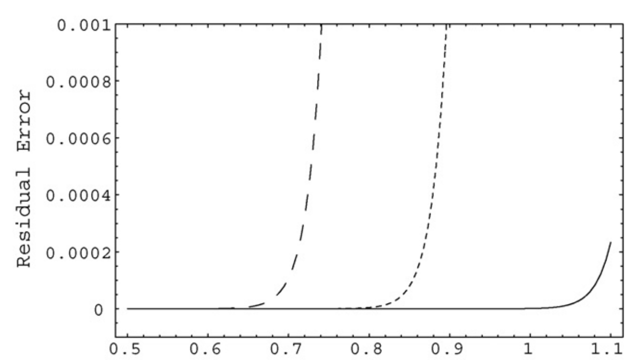

Fig. 7. Error of 20th-order HAM solution in case of $n = 0.5$ and $\varphi = 1.5$, solid line: $\hbar = -0.6$; dotted line: $\hbar = -0.8$; dashed line: $\hbar = -1$.

Table 1
Results for $u(0)$ at various values of n and φ

n	φ	M	$\hbar$	HAM	FDM
0.5	0.5	20	−1.0	0.881358	0.881359
0.5	1	20	−0.8	0.594446	0.594447
0.5	1.5	20	−0.6	0.294290	0.294290
2	1	20	−1.0	0.712256	0.712257
2	2	20	−1.3	0.443723	0.443723
2	4	20	−1.3	0.212590	0.212591
2	10	35	−1.3	0.0570842	0.0570843
2	20	50	−1.4	0.0175553	0.0175615
4	1	20	−1.2	0.779145	0.779148
4	10	50	−1.4	0.257003	0.257005

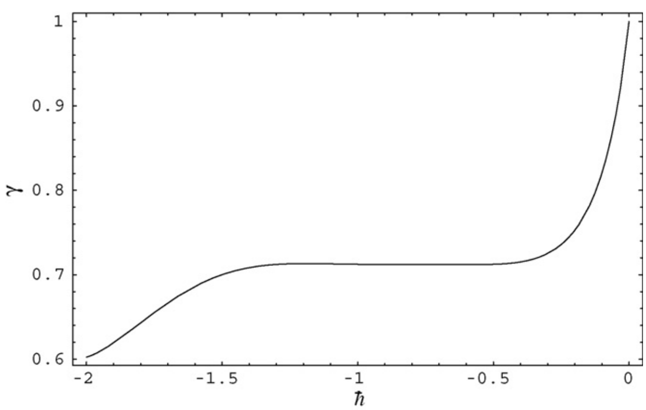

Fig. 8. γ with different $\hbar$, in case of $n = 2$ and $\varphi = 1$ at the 6th order of approximation.

5.2. Second approach

According to Section 4.2, we should ensure that the solution series (27) converge. In this case, the residual error is defined as

$$\text{Residual error} \approx W''_M(x) - \varphi^2 \Gamma_M^{n-1} W_M^n(x).$$

Fig. 8 shows the $u(0) = \gamma$ under different $\hbar$, in case of $n = 2$ and $\varphi = 1$. For showing the efficiency of HAM, we choose large value for φ in our examples.

It is seen that convergent results can be obtained when $-1.3 \lesssim \hbar \lesssim -0.5$. Figs. 9–12 show the results of HAM for different values of n and φ.

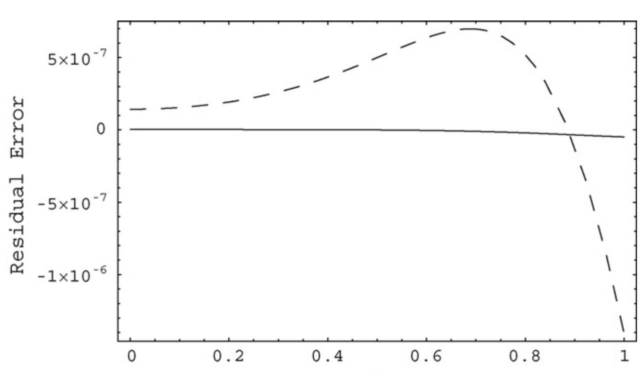

Fig. 9. Error of 15th-order HAM solution in case of $n = 2$ and $\varphi = 1$, solid line: $\hbar = -0.8$ with $u(0) = 0.712256$; dashed line: $\hbar = -1$.

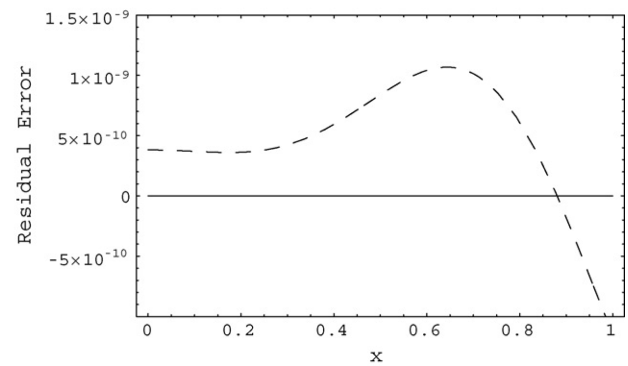

Fig. 10. Error of 15th-order HAM solution in case of $n = 2$ and $\varphi = 0.5$, solid line: $\hbar = -1.0$ with $u(0) = 0.895804$; dashed line: $\hbar = -1.2$.

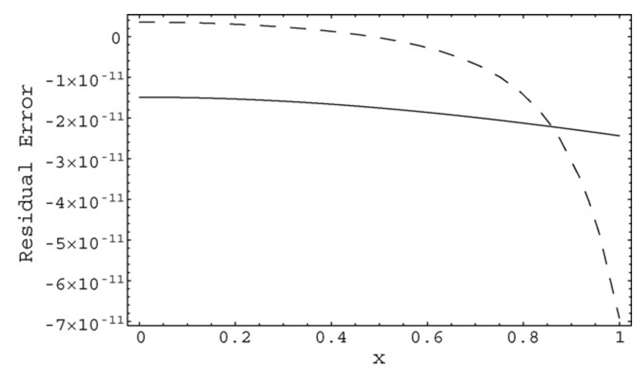

Fig. 11. Error of 15th-order HAM solution in case of $n = 0.5$ and $\varphi = 0.5$, solid line: $\hbar = -1.0$ with $u(0) = 0.881358$; dashed line: $\hbar = -1.2$.

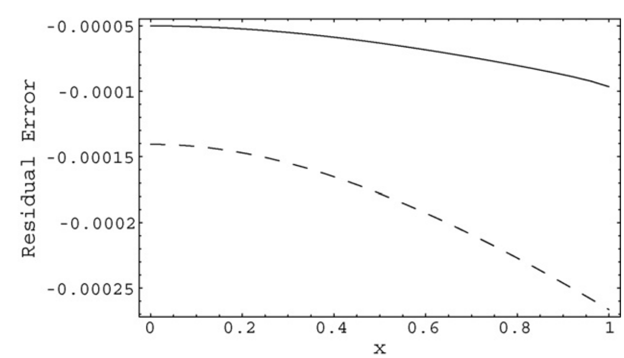

Fig. 12. Error of 15th-order HAM solution in case of $n = 0.5$ and $\varphi = 1$, solid line: $\hbar = -1.2$ with $u(0) = 0.594469$; dashed line: $\hbar = -1$.

6. Conclusions

In this paper, the homotopy analysis method [15] is applied to obtain the solutions for the nonlinear model of diffusion and reaction in porous catalysts. Also, it is shown that the obtained solution by HAM logically contains the solution obtained with Adomian decomposition method. Two different approaches are considered. We chose large value for φ to indicate the efficiency of HAM. The obtained results were also compared with the numerical solutions obtained by finite difference method.

The benefits of HAM with respect to Adomian's decomposition method are stated. It should be emphasized, HAM provides us with a convenient way to control the convergence of approximation series, which is a fundamental qualitative difference in analysis between HAM and other methods. So, this paper shows the flexibility and potential of the homotopy analysis method for complicated nonlinear problems in engineering.

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