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REVIEW ARTICLE

MATHEMATICAL MODELING OF OSCILLATORY CHEMICAL REACTIONS IN CLOSED VESSELS

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ABSTRACT

Oscillating reactions are among the most fascinating of chemical reactions .The system is considered here with two chemical species, the reactant and autocatalyst. The non steady-state concentration profiles of the reactant and autocatalyst in this model are obtained using He's Homotopy pertuburation method for all values parameters. Furthermore, in this work the numerical simulation of the problem is also reported using SCILAB/MATLAB program. A satisfactory agreement with numerical results is noted.

INTRODUCTION

There is a great deal of interest in chemical reactions which exhibit oscillatory solutions**.** These oscillations occur due to feedback in the system either chemical feedback such as autocatalysis or temperature feedback due to non-isothermal reaction. Thanga Pandi and Rajendran (2011) have obtained the non steady- state concentration profiles of the reactant, product and autocatalyst in Gray-Scott model using He's Homotopy pertuburation method for small values parameters. Shanthi *et al*. (2012) discussed the Gray-Scott scheme, which represents cubic-autocatalysis with linear catalyst decay. See Gray (1988) and Gray and Scott (1990) for reviews and descriptions of much of this work. Recently Marchant (2002) obtained the steady-state solutions for the cubic-autocatalytic reaction with linear decay in a reaction- diffusion cell using semi-analytical method. However, to the best of our knowledge there was no analytical result corresponding to the steady-state concentration of reactant and autocatalyst for all positive values of parameters have reported. The purpose of this communication is to derive the approximate analytical expressions for the concentrations of the reactant and autocatalyst for non steady state, using homotopy perturbation method.

Kinetic model and rate equations

We consider a complete reaction kinetics based upon the cubic autocatalator, with the intermediate species A produced via a simple first-order decay process from a precursor or reactant P. Thus, the scheme is represented as follows (Merkin (1986)):

$$P \rightarrow A \qquad rate = k_0 p \qquad \text{.....(1)}$$

$$A + 2B \rightarrow 3B \qquad rate = k_1 ab^2 \qquad \text{.....(2)}$$

$$B \rightarrow C \qquad rate = k_2 b \qquad \text{.....(3)}$$

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Later, the uncatalysed process parallel to step (1), namely

$$A \rightarrow B \qquad rate = k_3 a \qquad \text{……..(4)}$$

will also be included, along with the reverse of these two reactions. The governing differential rate equations for the reaction scheme (1) to (3) can be represented as follows:

$$dp/d\bar{t} = -k_0 p \qquad \text{……..(5)}$$

$$da/d\bar{t} = k_0 p - k_1 a b^2 \qquad \text{……...(6)}$$

$$db/d\bar{t} = k_1 a b^2 - k_2 b \qquad \text{……(7)}$$

The initial concentration of species A is $a(t=0) = a_0$. Thus, by introducing the following dimensionless variables

$$\alpha = a/a_0, \quad \beta = b/a_0, \quad \pi = p/a_0, \tau = k_1 a_0^2 \bar{t}, \varepsilon' = k_0/k_1 a_0^2 \text{ and } \tau_2 = k_1 a_0^2/k_2 \qquad \text{…….(8)}$$

the differential equations (5) to (7) becomes in dimensionless form as follows:

$$d\pi/d\tau = -\varepsilon'\pi \qquad \text{…….(9)}$$

$$d\alpha/d\tau = \varepsilon'\pi - \alpha\beta^2 \qquad \text{…….(10)}$$

$$d\beta/d\tau = \alpha\beta^2 - \beta/\tau_2 \qquad \text{…….(11)}$$

Since the equation (9) is first order linear differential equation, we can obtain the concentration of the precursor as

$$\pi = \pi_0 \exp(-\varepsilon'\tau) \qquad \text{…….(12)}$$

where $\pi_0 = p_0/a_0$ in the dimensionless form for the initial concentration of *P*. Substituting this exact result in the equation (9) leaves two rate equations describing the evolution of the concentrations of A and B (the intermediate species). The initial concentration of the precursor reactant may be commonly many orders of magnitude greater than that of a species such as *A*, thus π_0 may be very large compared with unity. i.e. k_0 will be small compared with $k_1 a_0^2$, or equivalently, ε' will be small compared with unity. Now the equations (10) and (11) becomes

$$d\alpha/d\tau = ke^{-\varepsilon'\tau} - \alpha\beta^2 \qquad \text{…….(13)}$$

$$d\beta/d\tau = \alpha\beta^2 - m\beta \qquad \text{…….(14)}$$

where $\kappa = \varepsilon'\pi_o$ and $m = 1/\tau_2$ …….(15)

The initial conditions becomes

$$\alpha(\tau=0) = 1, \ \beta(\tau=0) = \beta_0, \qquad \text{…….(16)}$$

where $\beta_0 = b_0/a_0$ is some constant.

Analytical solutions of concentrations of the species using HPM

Recently many authors have applied the HPM (Ghori, Ahmed, and .Siddiqui (2007),Ozis and Yildirim, (2007), Cai., Wu and Li (2006) and Ariel (2010)) to various problems and demonstrated the efficiency of the HPM for handling non-linear structures. This method is a combination of homotopy in topology and classic perturbation techniques. He (1999) have used the HPM to solve many linear and nonlinear problems. Recently Rajendran and co-workers (2010) obtained the analytical solution for various non-linear problems in enzyme-substrate reaction mechanisms. The HPM is unique in its applicability, accuracy and efficiency. The HPM uses the imbedding parameter *p* as a small parameter and only a few iterations are needed to search for an asymptotic solution. By solving the Eqs. (13) and (14) using homotopy perturbation method (Refer Appendix-A), we can obtain the concentration of species as follows:

$$\pi(\tau) = \pi_0 e^{-\varepsilon'\tau} \qquad \text{.....(17)}$$

$$\alpha(\tau) = \alpha_{ini} + \frac{k}{\varepsilon'} - \frac{k e^{-\varepsilon'\tau}}{\varepsilon'} + \beta_{ini}^2 \left(\frac{\alpha_{ini} e^{-2m\tau}}{2m} + \frac{k e^{-2m\tau}}{2m\varepsilon'} - \frac{k e^{-\left(\varepsilon'+2m\right)\tau}}{\varepsilon'\left(\varepsilon'+2m\right)} \right) - \beta_{ini}^2 \left(\frac{\alpha_{ini}}{2m} + \frac{k}{2m\varepsilon'} - \frac{k}{\varepsilon'\left(\varepsilon'+2m\right)} \right) \qquad \text{.....(18)}$$

$$\beta(\tau) = \frac{\alpha_0\beta_{ini}^2}{m} + \left(\beta_{ini} - \frac{\alpha_0\beta_{ini}^2}{m} \right) e^{-m\tau} + l_0\left(1 - e^{-m\tau}\right) + l_1\left(e^{-m\tau} - e^{-2m\tau}\right) + l_2\left(\tau e^{-m\tau}\right) + l_3\left(e^{-m\tau} - e^{-\left(\varepsilon'+2m\tau\right)}\right) + l_4\left(e^{-m\tau} - e^{-m\tau}\right) \qquad \text{.....(19)}$$

where

$$k = \varepsilon'\pi_0, \quad l_0 = \frac{1}{m^3}\left(\alpha_{ini} + \pi_0\right)\left(\alpha_0^2\,\beta_{ini}^4\right) \quad , \quad l_1 = \frac{1}{m}\left(\alpha_{ini} + \pi_0\right)\left(\beta_{ini} - \frac{\alpha_0\beta_{ini}^2}{m} \right)^2 \quad l_2 = 2\frac{\alpha_0\beta_{ini}^2}{m}\left(\beta_{ini} - \frac{\alpha_0\beta_{ini}^2}{m} \right)\left(\alpha_{ini} + \pi_0\right) \quad ,$$

$$l_3 = \frac{-\pi_0\left(\beta_{ini} - \frac{\alpha_0\beta_{ini}^2}{m} \right)^2}{\varepsilon' + m} \quad , \quad l_4 = \frac{-\pi_0\left(\alpha_0^2\beta_{ini}^4\right)}{m^2\left(m - \varepsilon'\right)} \qquad (20)$$

Equations (18) and (19) represent the new simple analytical expression of concentration of species for all values of parameters.

Numerical simulation

The non-linear differential equations (13) and (14) are also solved by numerical methods using SCILAB/MATLAB software. Its numerical solution is compared with homotopy perturbation method in Figs. (1) - (2) and Tables 1 and 2. It gives a satisfactory agreement for all values of parameters $k, m, \varepsilon' \text{ and } \pi_0$. The SCILAB/MATLAB program is also given in Appendix-B.

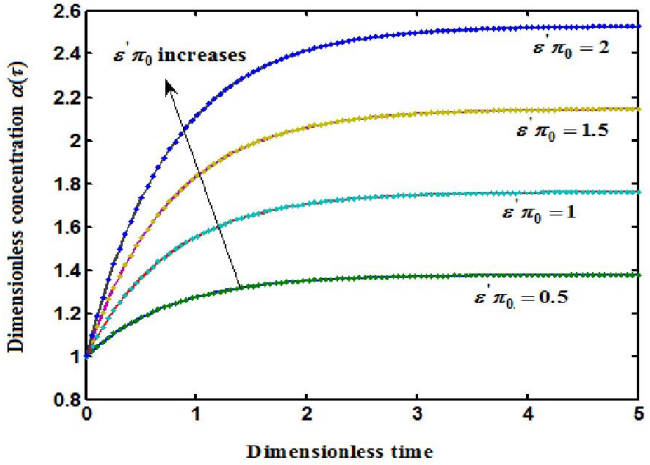

Fig. 1. Comparison of concentration of α for $m = 1, \varepsilon' = 1.3$ and various values of $\varepsilon'\pi_0$ Doted lines represent the analytical solution and solid lines the numerical solution

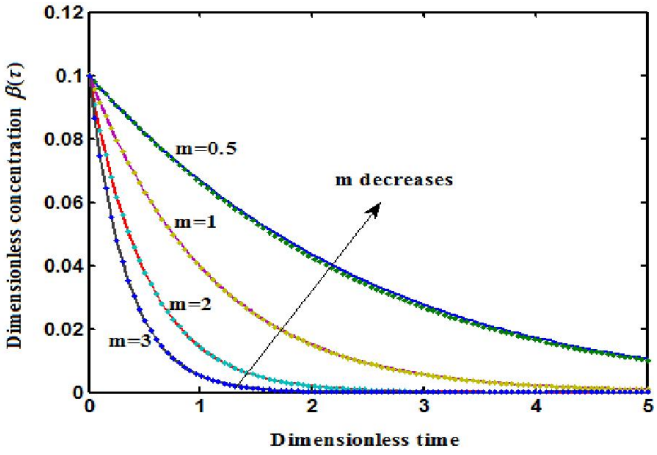

Fig. 2. Comparison of concentration of β for $\varepsilon'\pi_0 = 0.5, \varepsilon' = 1.3$ and various values of m. Doted lines represent the analytical solution and solid lines the numerical solution

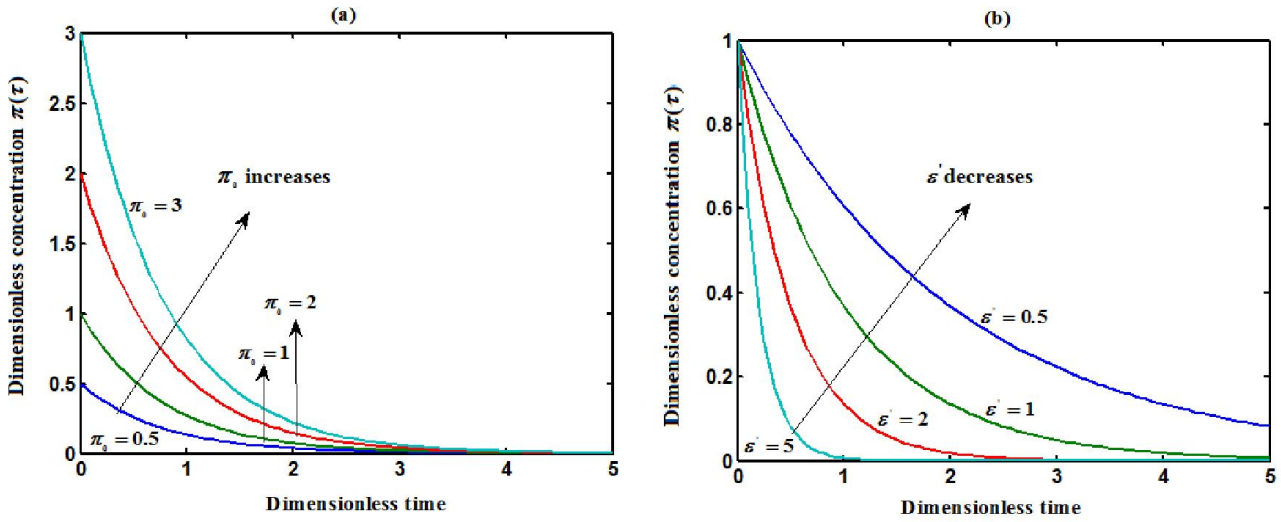

Fig.3 Profiles of the normalized concentrations π versus dimensionless time τ calculated using equation (18) for various values of parameters. (a) $\varepsilon' = 1.3$ and various values of π_0

(b) $\pi_0 = 1$ and various values of ε'

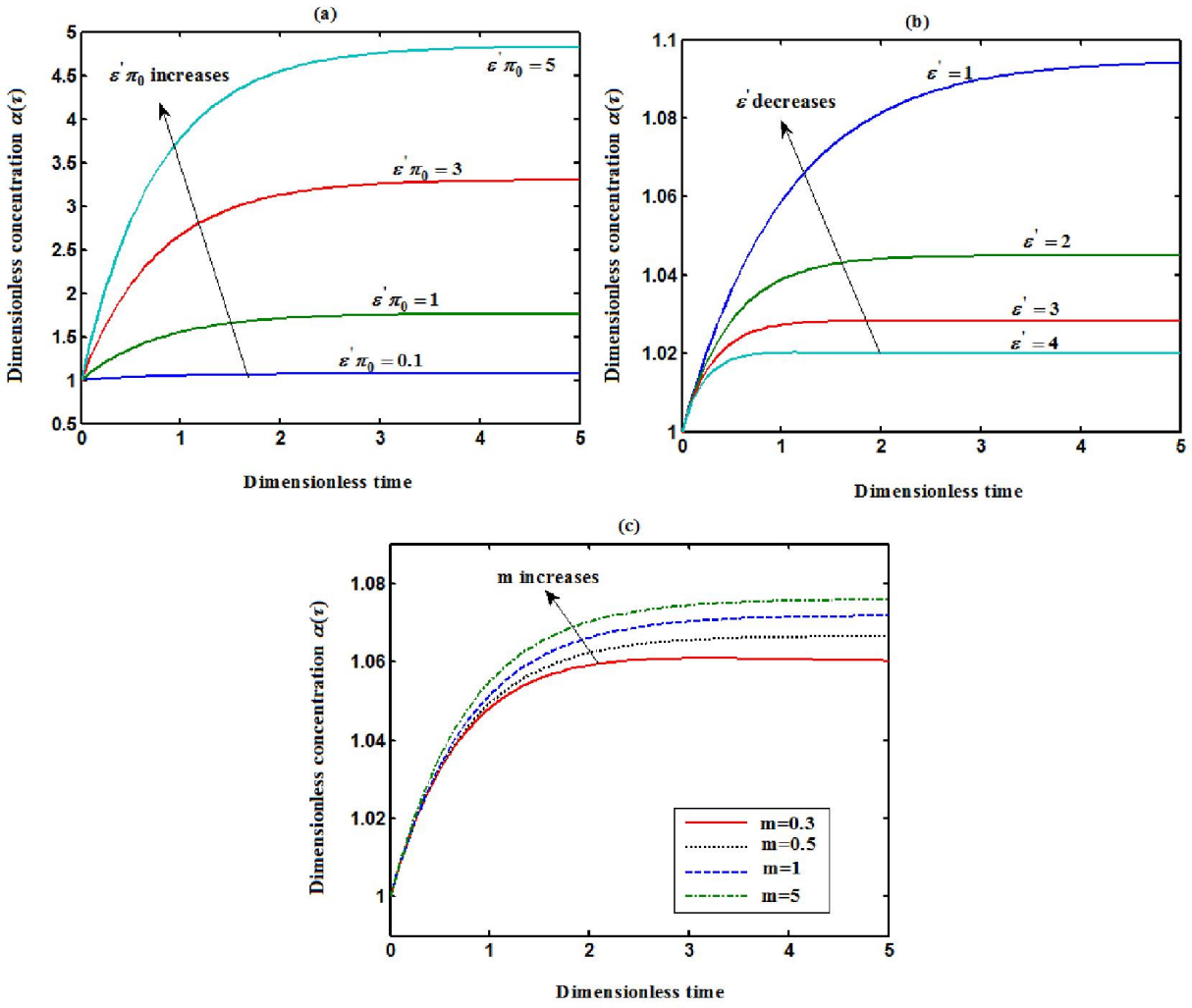

Fig.4 (a) Profiles of the normalized concentrations α versus dimensionless time τ calculated using equation (19) for various values of parameter. (a) $m = 1, \varepsilon' = 1.3$ and various values of $\varepsilon'\pi_0$. (b) $m = 1, \varepsilon'\pi_0 = 0.1$ and various values of ε'.

(c) $\varepsilon' = 1.3, \varepsilon'\pi_0 = 0.1$ and various values of m

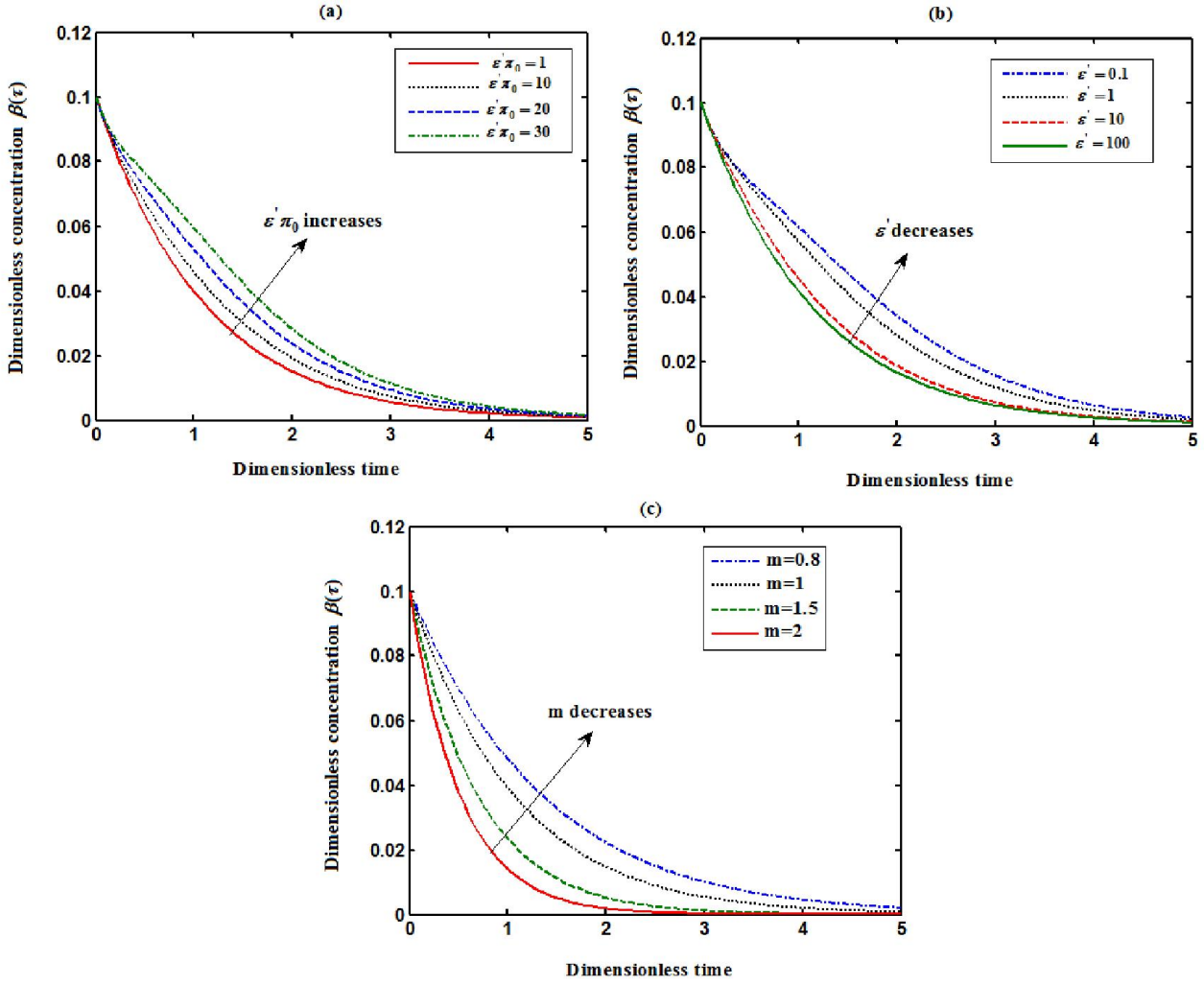

Fig.5 Profiles of the normalized concentrations intermediate species *B* ((β) for (a) $m=1,\ \varepsilon^{'}=1.3$ and various values of $\varepsilon^{'}\pi_0$. (b) $m=0.95,\ \varepsilon^{'}\pi_0=20$ and various values of $\varepsilon^{'}$. (c) $\varepsilon^{'}=0.3,\ \varepsilon^{'}\pi_0=0.1$ and various values of m. The curves are plotted using equations (20)

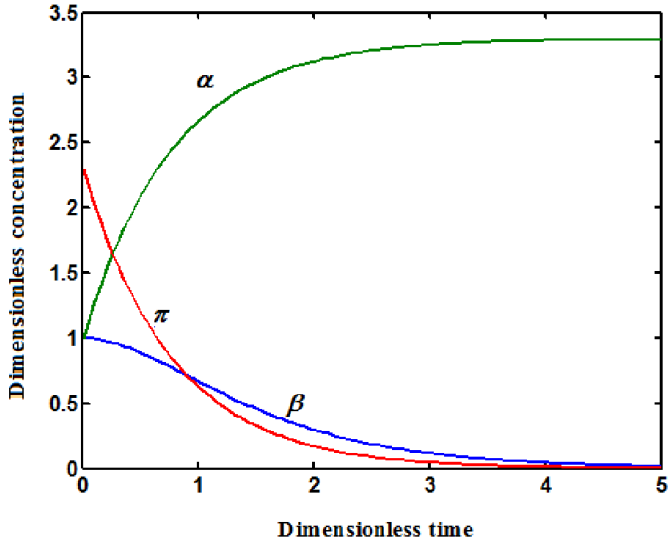

Fig. 6. Plot of concentration profiles of π,α and β .The values of the parameters are $\varepsilon^{'}\pi_0=1,\ \varepsilon^{'}=1.3,\ m=1,\ \pi_0=2.3$

List of symbols

symbol	meaning	units
a	Concentration of intermediate A	mol dm^{-3}
b	Concentration of intermediate B	mol dm^{-3}
k_0	Rate constant for decay of precursor	s^{-1}
k_1	Rate constant for autocatalytic step	$dm^6 mol^{-2} s^{-1}$
k_2	Rate constant for autocatalyst decay	s^{-1}
k_3	Rate constant for uncatalysed step	s^{-1}
p	Concentration of precursor P	mol dm^{-3}
$\bar{t}$	time	s
α	A/a_0, Dimensionless concentration of A	none
β	B/a_0, Dimensionless concentration of B	none
ε'	$k_0 / k_1 a_0^2$,Dimensionless rate constant for decay of precursor	none
k	$\varepsilon' \pi_0$,Bifurcation parameter	none
π	p / a_0 , Dimensionless concentration of P (precursor/reactant)	none
τ	$k_1 a_0^2 \bar{t}$, Dimensionless time	none
τ_2	$k_1 a_0^2 / k_2$, Dimensionless catalyst lifeline	none

Table 1: Comparison of normalized non-steady-state concentration of α with simulation results for various values of $\varepsilon' \pi_0$ and for some fixed values of $\varepsilon' = 1.3 and m = 1$

τ	Concentration α								
	when $\varepsilon' \pi_0$ = 1			when $\varepsilon' \pi_0$ = 1.5			when $\varepsilon' \pi_0$ = 2		
	Eq.(18)	Simulation	% of error deviation	Eq.(18)	Simulation	% of error deviation	Eq.(18)	Simulation	% of error deviation
0	0.9999	1.0000	0.01	1.0000	1.0000	0.00	1.0000	1.0000	0.00
1	1.5541	1.5537	0.02	1.8334	1.8329	0.02	2.1126	2.1120	0.02
2	1.7057	1.7051	0.03	2.0610	2.0603	0.03	2.4163	2.4153	0.04
3	1.7471	1.7464	0.04	2.1232	2.1224	0.03	2.4993	2.4979	0.05
4	1.7584	1.7574	0.05	2.1402	2.1393	0.04	2.5219	2.5203	0.06
5	1.7615	1.7598	0.09	2.1448	2.1432	0.07	2.5281	2.5260	0.08
	Average error % 0.04			Average error % 0.03			Average error % 0.04		

Table 2. Comparison of normalized non-steady-state concentration of β with simulation results for various values of m and for some fixed values of $\varepsilon' = 1.3 and \varepsilon' \pi_0 = 0.5$

τ	Concentration β								
	when m = 0.5			when m = 1			when m = 2		
	Eq.(19)	Simulation	% of error deviation	Eq.(19)	Simulation	% of error deviation	Eq.(19)	Simulation	% of error deviation
0	0.1021	0.0999	2.20	0.1016	0.1000	1.60	0.1011	0.0999	1.20
1	0.0674	0.0667	1.04	0.0400	0.3965	1.01	0.0143	0.0142	0.70
2	0.0432	0.0434	0.46	0.0151	0.0150	0.66	0.0019	0.0019	0.00
3	0.0271	0.0276	1.81	0.0056	0.0056	0.00	0.0002	0.0002	0.00
4	0.0167	0.0172	2.90	0.0020	0.0020	0.00	0.0000	0.0000	0.00
5	0.0102	0.0106	3.77	0.0007	0.0007	0.00	0.0000	0.0000	0.00
	Average error % 2.03			Average error % 0.54			Average error % 0.31		

DISCUSSIONS

Figure 3-5 show the analytical expressions of dimensionless concentrations $\pi, \alpha \; and \; \beta$ of species for various values of dimensionless reaction parameters $k, m, \varepsilon' \; and \; \pi_0$. From these Figs 3(a-b), it is inferred that the value of the concentration of reactant always decreases from its initial value of the concentration for all values of parameters. Also the concentration increases when π_0 eases or ε' decreases. The concentration of π decreases become zero when dimensionless time ≥ 10. Figs 4 (a-c) and (c)

represents the plot of concentration of intermediate species α versus time for various values of parameters. From the figures it is observed that the concentration of intermediate species increases slowly from the initial concentration $(\alpha_0 = 1)$. Also From these figures ,it is inferred that the concentration increases when the value of the parameters of $\varepsilon'\pi_0$ and m increases or ε' decreases. Fig 5 (a) represents that the dimensionless concentration autocatalyst β versus time. From the Figures it is observed that concentration autocatalyst is increases when $\varepsilon'\pi_0$ increases or ε' and m decreases. It reaches the steady state when time $\tau \geq 3$.

Conclusion

In this work, the coupled system of time dependent non linear differential equations for oscillatory chemical autocatalytic reaction has been solved analytically using the HPM. Moreover, we have also presented an approximate analytical expression for the non-steady state concentration of profiles of reactant and product for all vales of time and other parameters. The extension of the procedure to well-stirred reactor (CSTR) models seems possible.

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Appendix (A). Solution of the nonlinear the equations using homotopy perturbation method

In this appendix, we indicate how Eqs. (13) and (14) may be solved using HPM . To illustrate the basic concepts of this method (HPM), we consider the following nonlinear differential equation $L(u) + N(u) - f(r) = 0$ where L is a linear operator, N is a nonlinear operator, and $f(r)$ is a given continuous function**.** We construct a Homotopy $\Omega \times [0,1] \rightarrow R$ which satisfies

$$(1-p)\left[\frac{d\alpha}{d\tau} - A e^{-\varepsilon'\tau}\right] + p\left[\frac{d\alpha}{d\tau} + \alpha\beta^2 - A e^{-\varepsilon'\tau}\right] = 0 \qquad \text{(A1)}$$

$$(1-p)\left[\frac{d\beta}{d\tau}+m\beta\right]+p\left[\frac{d\beta}{d\tau}+m\beta-\alpha\beta^2\right]=0 \qquad \text{(A2)}$$

The initial approximations are as follows:

$$\alpha(0)=1,\beta(0)=\beta_0 \qquad \text{(A3)}$$

The approximate solutions of (A1) and (A2) are given by

$$\alpha=\alpha_0+p\alpha_1+p^2\alpha_2+p^3\alpha_3+..... \qquad \text{(A4)}$$

$$\beta=\beta_0+p\beta_1+p^2\beta_2+p^3\beta_3+..... \qquad \text{(A5)}$$

Substituting Equations (A4) and (A5) comparing the coefficients of like powers of *P,* we obtain the following differential equations.

$$p^0:\frac{d\alpha_0}{d\tau}-Ae^{-\varepsilon'\tau}=0 \qquad (A6)$$

$$p^1:\frac{d\alpha_1}{d\tau}+\alpha_0\beta_0^2=0 \qquad (A7)$$

$$p^0:\frac{d\beta_0}{d\tau}+m\beta_0-\alpha_0\beta_{ini}^2=0 \qquad (A8)$$

$$p^1:\frac{d\beta_1}{d\tau}+m\beta_1-\alpha_0\beta_0^2=0 \qquad (A9)$$

Upon solving the equations (A6)-(A9) and using the boundary conditions (A3), we get

$$\alpha_0=\alpha_{ini}+\frac{A}{\varepsilon'}-\frac{Ae^{-\varepsilon'\tau}}{\varepsilon'} \qquad \text{(A10)}$$

$$\alpha_1=\beta_{ini}^2\left(\frac{\alpha_{ini}e^{-2m\tau}}{2m}+\frac{Ae^{-2m\tau}}{2m\varepsilon'}-\frac{Ae^{-(\varepsilon'+2m)\tau}}{\varepsilon'(\varepsilon'+2m)}\right)-\beta_{ini}^2\left(\frac{\alpha_{ini}}{2m}+\frac{A}{2m\varepsilon'}-\frac{A}{\varepsilon'(\varepsilon'+2m)}\right) \qquad \text{(A11)}$$

$$\beta_0=\frac{\alpha_0\beta_{ini}^2}{m}+\left(\beta_{ini}-\frac{\alpha_0\beta_{ini}^2}{m}\right)e^{-m\tau} \qquad \text{(A12)}$$

$$\beta_1=l_0\left(1-e^{-m\tau}\right)+l_1\left(e^{-m\tau}-e^{-2m\tau}\right)+l_2\left(\tau e^{-m\tau}\right)+l_3\left(e^{-m\tau}-e^{-(\varepsilon'+2m\tau)}\right)+l_4\left(e^{-m\tau}-e^{-m\tau}\right) \qquad \text{(A13)}$$

where $l_0=\frac{1}{m^3}(\alpha_{ini}+\pi_0)(\alpha_0^2\beta_{ini}^4)$ $\quad l_1=\frac{1}{m}(\alpha_{ini}+\pi_0)\left(\beta_{ini}-\frac{\alpha_0\beta_{ini}^2}{m}\right)^2$

$$l_2=2\frac{\alpha_0\beta_{ini}^2}{m}\left(\beta_{ini}-\frac{\alpha_0\beta_{ini}^2}{m}\right)(\alpha_{ini}+\pi_0) \qquad l_3=\frac{-\pi_0\left(\beta_{ini}-\frac{\alpha_0\beta_{ini}^2}{m}\right)^2}{\varepsilon'+m} \qquad l_4=\frac{-\pi_0\left(\alpha_0^2\beta_{ini}^4\right)}{m^2(m-\varepsilon')}$$

Hence we obtain

$$\alpha \approx \alpha_0 + \alpha_1 \quad \text{(A14)}$$

$$\beta \approx \beta_0 + \beta_1 \quad \text{(A15)}$$

Substituting the equations (A10) to (A13) in the above equations we obtain the Eqs.(13) and (14) in the text.

Appendix B: Matlab/scilab program for the numerical solution of the non linear equations (13) and (14)

```
function main13;
options=odeset('RelTol',1e-6,'Stats','on');
%initial conditions
x0=[1;0.1];
tspan=[0,5];
tic
[t,x]=ode45(@TestFunction,tspan,x0,options);
toc
figure
hold on
plot(t,x(:,1),'red')
plot(t,x(:,2),'green')
legend('x1','x2')
ylable('x')
xlable('t')
return
function[dx_dt]=TestFunction(t,x)
dx_dt(1)=0.5*exp(-1.3*t)-x(1)*(x(2))^2;
dx_dt(2)=x(1)*x(2)^2-(3*x(2));
dx_dt=dx_dt';
return
```

****