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MODELLING AND SIMULATION OF A LAMINAR FLOW TUBULAR REACTOR (LFTR)

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ABSTRACT:

Residence Time Distribution (RTD) is a popular technique to determine the extent of non-ideality in reactors but can be used only after design and fabrication. There are no methods available to incorporate non-ideality at the design stage. However E curve is available for constant density laminar flow which can be used to predict conversion. The proposed model equation has analytical solutions only for some special cases such as elementary first-order and second order reactions and that too for special cases of reactants in stoichiometirc ratio. Therefore in the present work attempt has been made to formulate, test and validate a numerical model which will incorporate non-ideality in the reactor performance before the design is made. The results of this numerical model are compared with those of simulations carried out on commercial Computational Fluid Dynamics (CFD) package namely STAR CCM+ ver. 5.04.008. Numerical model results agree well with the simulation results. The small difference in the results of numerical method and simulation were analyzed and the reasons for the same were identified. The difference between the conversion obtained by the numerical model and simulation was found to increase, reach a maximum and then decrease as the Damkohler Number (N_{Da}) was increased. It was also found that for a reaction having kinetics $-r_A = kC_A^2$, a maxima occurred at $N_{Da} = 1.63$ irrespective of initial concentration of reactants. Further it was observed that the plot of conversions predicted by numerical model versus conversion in simulation was universal in the sense that all such curves overlapped for different values of rate constant and initial concentration of reactants for a given reaction type.

KEYWORDS: Computational Fluid Dynamics (CFD), Residence Time Distribution (RTD), Conversion, Laminar Flow, Modelling

[I] INTRODUCTION

A new emerging tool in chemical process engineering is the combination of computational fluid dynamics and numerical mathematics backed by the immense growth in computing power [3]. Historically a strong empirical base had been devised to approximately solve extremely complicated problems encountered in

chemical reaction engineering such as the design of process equipment including chemical reactors involving non ideal flow (Levenspiel. 1962).

Levenspiel asserts that with the precise knowledge of the velocity distribution inside the reactor, we can accurately predict its performance without going into the details of how it can be done. Strong coupling effects between transport and reaction rate processes and non linear dependence of the kinetic and transport coefficients on the state variables have led to difficulties in describing mixing systems. Even with detailed solutions available, numerical results do not directly provide required results such as average exit conversion of a reactant or yield of intermediate product [2]. In the past, chemical engineers have circumvented this problem by developing low dimensional models of reactors by making a certain prior assumptions as the length and time scales of reaction, diffusion and convection and applying the conservation equations only at the macroscopic level. Accurate low dimensional models in terms of average (and measurable) variables are desired for the purpose of design, control and optimization of chemical processes [2].

Chakraborty and Balakotaiah illustrated the use of Liapunov–Schmidt Technique [2] based on Classical Bifurcation theory for the purpose of spatial averaging, eliminating spatial degrees of freedom and derive accurate low-dimensional models for a class of convection diffusion reaction (CDR) equations. However a complete quantification of the mixing process by solving the CDR equations from the reactor scale (global) to the molecular diffusion and reaction scale (local) may sometimes be impractical for design based applications. With the intensive computation required for individual cases and generalization improbable to achieve, microbalance models are not of great practical importance to the engineer, who is mainly interested in the average value of concentration [2,3].

Since E curve is theoretically available for Laminar Flow, it can be exploited for design of LFTRs that incorporate non-ideality. The main focus of the present work is to develop a numerical model to determine the performance of a laminar flow reactor and compare the conversion with that of CFD simulations.

[II] EXISTING ANALYTICAL METHODS

Conversion in any existing reactor may be determined using RTD information and applying appropriate model. Best predictions are obtained when the RTD obtained from real reactors and those of the models exactly match. This requires theoretical RTD information of various models. This is given by Levenspiel for some models. If there is no matching of the RTD information, then new model is to be proposed, RTD information obtained and compared. This process has to be repeated for different models until matching RTD is obtained. Once this matching is achieved, the conversion in the model is expected to match the conversion in real reactor. This work gets simplified for LFTRs as theoretical RTD information can be accurately determined. Segregated flow model is appropriate if the reaction mixture is macro fluidic [4]. Theoretical RTD information for LFTR is in the form of E-Curve which is determined using

$$E(t) = \begin{cases} 0 & \text{for } t < \frac{\bar{t}}{2} \\ \frac{\bar{t}^2}{2t^3} & \text{for } t \geq \frac{\bar{t}}{2} \end{cases} \qquad (1)$$

Segregated flow model assumes chunks of fluid flowing as if each chunk is a batch reactor. This means there are several small batch reactors flowing through the reactor and coming out at different times. Collectively the conversion of all such batch reactors is determined using

$$\frac{\bar{C}_A}{C_{A0}} = \int_0^\infty \left(\frac{C_A}{C_{A0}}\right)_{ele} E \, dt \qquad (2)$$

Table 1 gives a summary of analytical solutions for simple cases.

Table-1

Order of reaction	Rate Equation	$\bar{C}_A/C_{A_0}$	Author
Zero	$-r_A = k$	$\left(1-\frac{k\bar{t}}{2C_{A_0}}\right)^2$	Bosworth(1948)
One	$-r_A = kC_A$	$y^2 ei(y) + (1-y)e^{-y}, y = \frac{k\bar{t}}{2}$	Cleland and Wilhelm(1956)
Two	$-r_A = kC_A^2$	$1 - kC_{A_0}\bar{t}\left[1 - \frac{kC_{A_0}\bar{t}}{2}\ln\left(1+\frac{2}{kC_{A_0}\bar{t}}\right)\right]$	Denbigh(1951)

Table: 1. Analytical solution for equation (2) [1]

Even for simple reactions of the type

$$A + B \rightarrow \text{products} \qquad (3)$$

with initial concentrations of reactants not in stoichiometirc ratio, analytical solution of equation (2) is not possible for all kinetics. This is due to two counts, one obtaining $(C_A/C_{A0})_{ele}$ as an explicit function of time and two, the determination of the integral in equation (2). There is a need to address this issue. There are no methods available in the literature cited by the authors. The numerical model developed in this paper deals with the determination of conversion using equation (2).

[III] NUMERICAL MODEL

As discussed above each element (ele) of fluid behaves as a batch reactor in transit, each of these elements have its own conversion associated with itself which can be expressed as:

$$(X_A)_{ele} = 1 - \left(\frac{C_A}{C_{A0}}\right)_{ele} \qquad (4)$$

The numerical model was applied to several cases of reaction and kinetics

Case 1:

$A + B \rightarrow \text{products}, \ (M \neq 1), \ -r_A = kC_A C_B$

Performance equation of batch reactor for this case is given by

$$\ln\left[\frac{M - X_A}{M(1 - X_A)}\right] = kC_{A0}(M-1)t \qquad (5)$$

The equation (5) may so rearranged to get

$$X_A = 1 - \frac{M-1}{M \cdot \exp[kC_{A0}(M-1)t] - 1} \qquad (6)$$

From which $(C_A/C_{A0})_{ele}$ can be known for different values of t using equation (4) expressed as

$$\left(\frac{C_A}{C_{A0}}\right)_{ele} = \frac{M-1}{M \cdot \exp[kC_{A0}(M-1)t] - 1} \qquad (7)$$

Thus the numerical values of $(C_A/C_{A0})_{ele}$ will be available for any value of t. Combining these values and E(t) given by equation (1), integral in equation (2) may be determined. This gives the performance of the reactor.

Case 2:

$A + 2B \rightarrow \text{products}, (M \neq 2), \ -r_A = kC_A C_B$

Applying the procedure described in case 1 for this reaction and kinetics, we get performance equation as

$$\ln\left[\frac{M-2X_A}{M(1-X_A)}\right] = kC_{A0}(M-2)t \quad (8)$$

On simplifying above equation, we get the following expression.

$$\left(\frac{C_A}{C_{A0}}\right)_{ele} = \frac{M-2}{M\cdot\exp[kC_{A0}(M-2)t]-2} \quad (9)$$

Case 3: $A + B \rightarrow products,\ (M = 1),\ -r_A = kC_A C_B$

The rate equation simplifies to $-r_A = kC_A^2$. Batch reactor performance equation may be used to get $(C_A/C_{A0})_{ele}$ as an explicit function of t which when substituted in equation (2) results into

$$\frac{\bar{C}_A}{C_{A0}} = 1 - kC_{A0}\bar{t}\left[1-\frac{kC_{A0}\bar{t}}{2}\ln\left(1+\frac{2}{kC_{A0}\bar{t}}\right)\right] \quad (10)$$

[IV] CFD SIMULATIONS

The validity of the conversion obtained from the numerical model was affirmed by carrying out simulations in a CFD package STAR CCM+ ver. 5.04.008. A tubular reactor was modeled using this software with diameter=1m and length=5 m. A steady state simulation was carried out under laminar flow conditions with constant density system in an isothermal continuum of Newtonian fluid. Initially the reaction system chosen was

$$NaOH + CH_3COOC_2H_5 \rightarrow CH_3COONa + C_2H_5OH$$

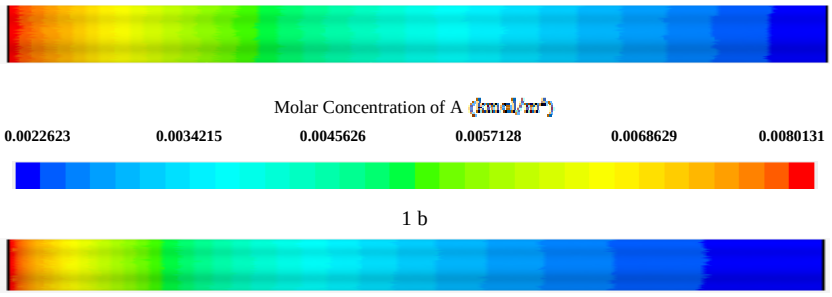
.

This was of the reaction type $A + B \rightarrow products$ with kinetics $-r_A = kC_A C_B$. This reaction could not be generalized because all the chemical and physical properties were taken from inbuilt material database and rate constant could not be varied. Hence to generalize this reaction type, a material database was created with reacting species as A and B with specified product. This was done so that rate constant, k could be varied for various simulation runs and conversion could be obtained for the same. A dilute solution was maintained at the inlet by specifying mass fraction of all the reacting species along with water. The average velocity at the inlet was chosen such that the flow is laminar. From the concentration profile obtained at the outlet, the average concentration of A, $\bar{C}_A$ was calculated in order to obtain the simulated conversion as defined by

$$X_{AS} = 1 - \frac{\bar{C}_A}{C_{A0}} \quad (11)$$

For all the three mentioned cases i.e., case 1, case 2 and case 3, the conversion obtained from the simulation (X_{AS}) and the numerical model (X_{AN}) were tabulated and compared.

1 a



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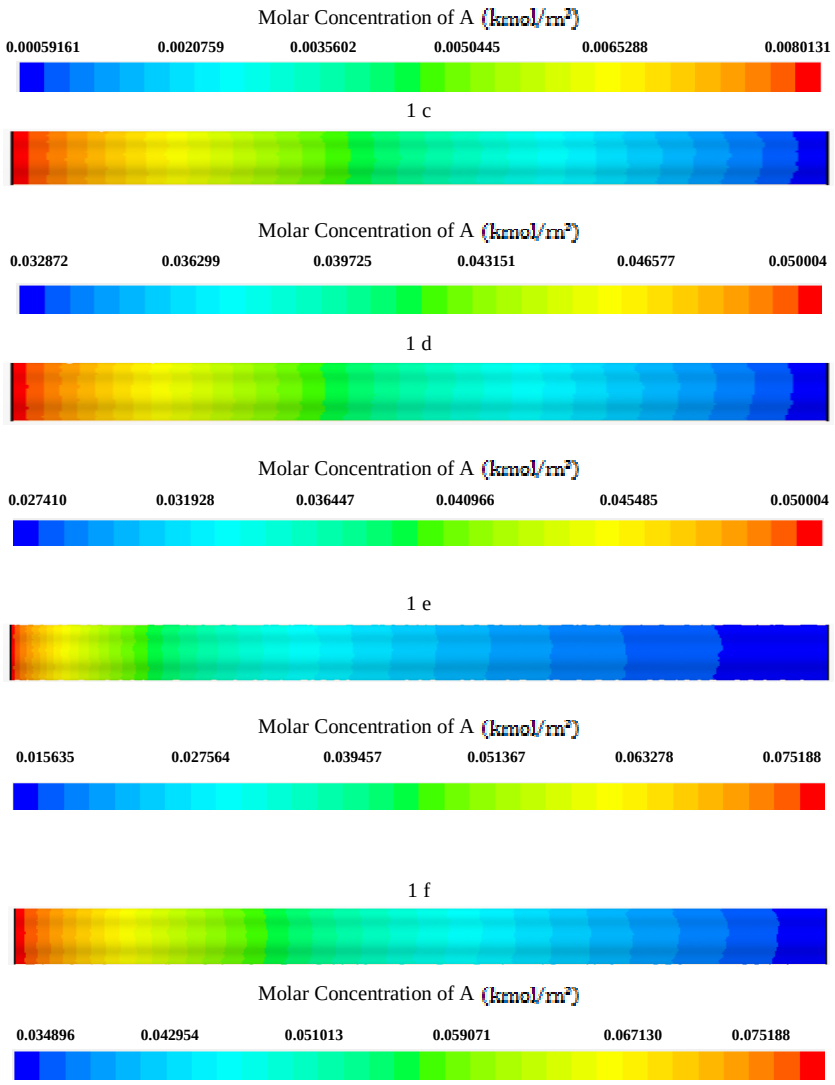


Fig: 1. Variation of molar Concentration of the limiting reactant across the tubular flow reactor for different reaction types. Details are enumerated in Table 2.

Table-2

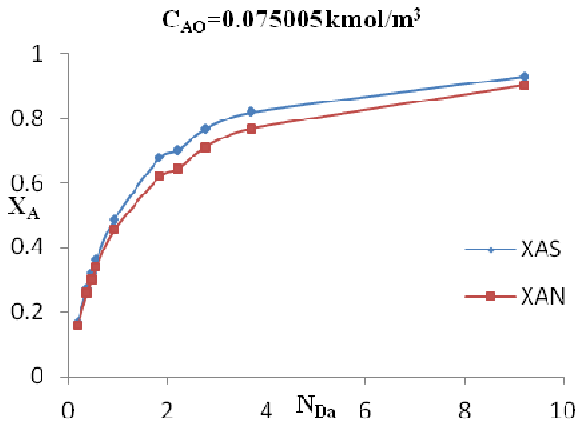
Stoichiometry	C_{A0} (kmol/m³)	M	k(m³/kmols)	Rate Equation	Figure No
A + 2B → Products	0.0080131	≠ 2	0.015	$-r_A = kC_AC_B$	1 a
			0.035		1 b
A + B → Products	0.050004	≠ 1	0.002	$-r_A = kC_AC_B$	1 c
			0.003		1 d
A + B → Products	0.075188	= 1	0.020	$-r_A = kC_A^1$	1 e
			0.006		1 f

Table: 2. Indexing of Fig. 1

[V] RESULTS AND DISCUSSION

For the considered reaction cases, various simulations were run for different values of initial concentrations of reactant A and B such that A was always the limiting reactant (Fig: 1). The concentration of A at the exit was used to compute X_{AS}. The numerical conversion X_{AN} was obtained by numerical integration of equation (2) by plugging in

values of $(C_A/C_{A0})_{ele}$ specific for a given reaction type. A graph was plotted between simulated and numerical conversion i.e., X_{AS} and X_{AN}. A fairly good agreement was obtained between X_{AS} and X_{AN}. A comparative plot of the simulated and the numerical conversion as a function of the Damkohler Number is shown in Fig: 2. The difference in conversions could be due to the fact that the flow in the simulation model has flat velocity profile at the entrance and gradually becomes parabolic whereas the numerical model assumes parabolic profile everywhere. The difference between simulated conversion and numerical conversion was found to be less for low conversions, increased with increasing conversions, reached a maximum and approached zero for conversions close to unity. However it was found that for a given reaction type following a definite rate equation and stoichiometry, a single curve was obtained irrespective of the initial concentrations of A, when the simulated conversion is plotted against the numerical conversion (Fig: 3, Fig: 4 and Fig: 5). This curve can be used to obtain simulated conversion from numerical conversion for better and accurate design of laminar flow reactor. This is particularly useful as numerical conversion is accurate in its own way and simulated conversion takes into account even the underdeveloped flow field. For different values of C_{A0}, the difference between conversions as a function of *k* is shown in Fig: 6. An interesting observation was made for the reaction type $A + B \rightarrow Product$ with kinetics $-r_A = kC_A^2$. For all values of C_{A0}, *k* and space time, when a graph was plotted taking the difference of simulated and numerical conversions i.e., (X_{AS}-X_{AN}) versus Damkohler Number (N_{Da}), all the curves were found to merge to give a single curve independent of C_{A0} with a global maximum at N_{Da}=1.63 (Fig: 7).



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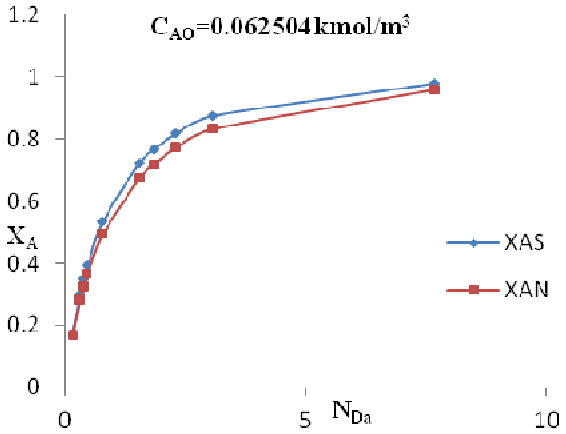

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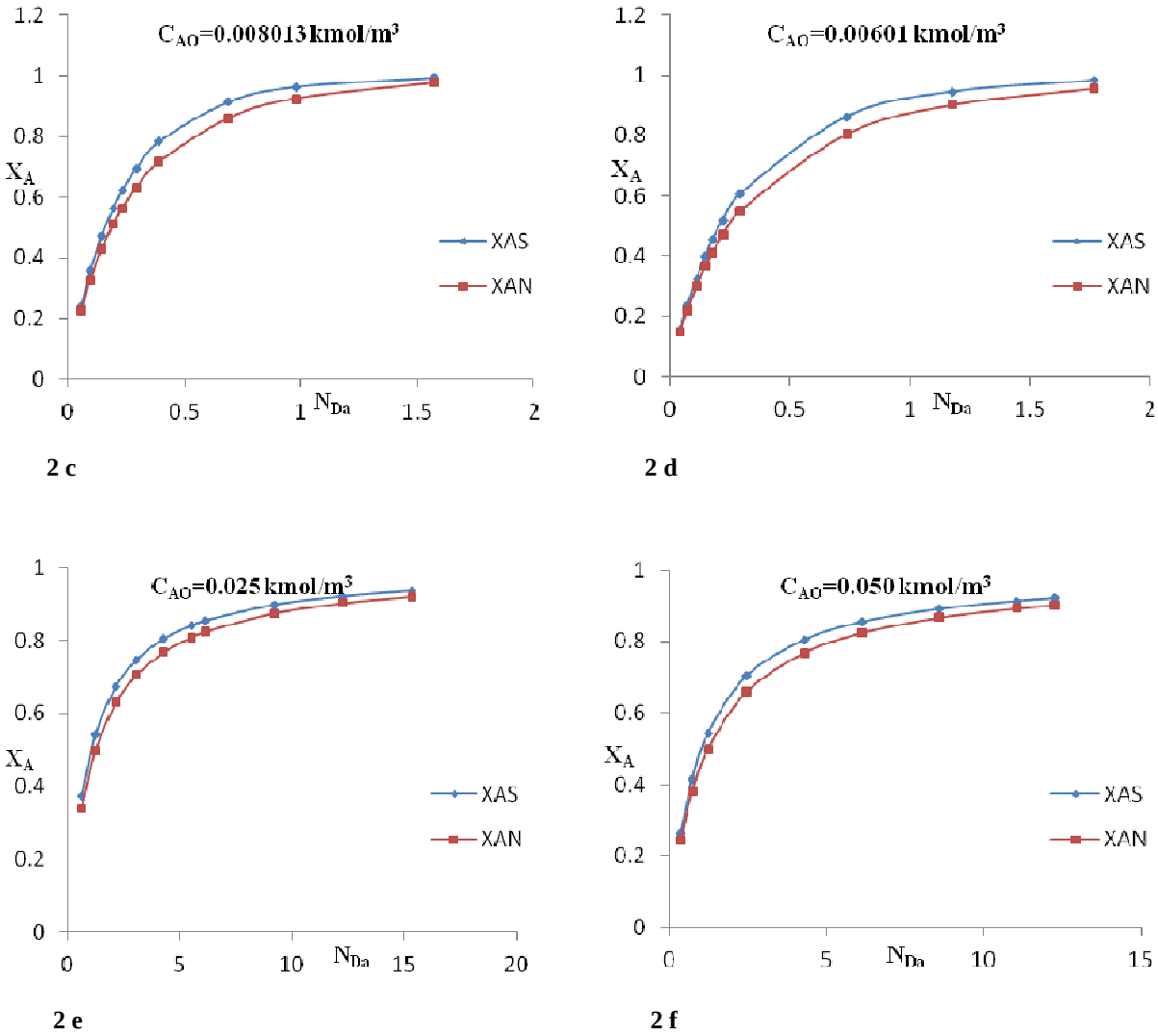

Fig: 2 Variation of conversion (simulated and numerical) with Damkohler Number for different reaction types. Details are enumerated in Table 3.

Table-3

Stoichiometry	C_{AO} (kmol/m³)	M	Rate Equation	Figure No
A + B → Products	0.050004	≠ 1	$-r_A = kC_AC_B$	2 a
				2 b
A + B → Products	0.075188	= 1	$-r_A = kC_A^2$	2 c
				2 d
A + 2B → Products	0.0080131	≠ 2	$-r_A = kC_AC_B$	2 e
				2 f

Table: 3. Indexing of Fig. 2

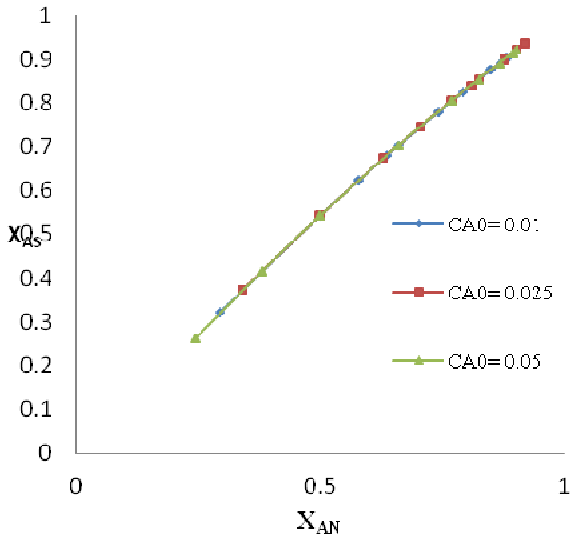

Fig: 3. Plot of numerical (X_{AN}) vs. simulation (X_{AS}) conversions for a reaction $A + B \rightarrow Products$ with $-r_A = kC_A^2$.

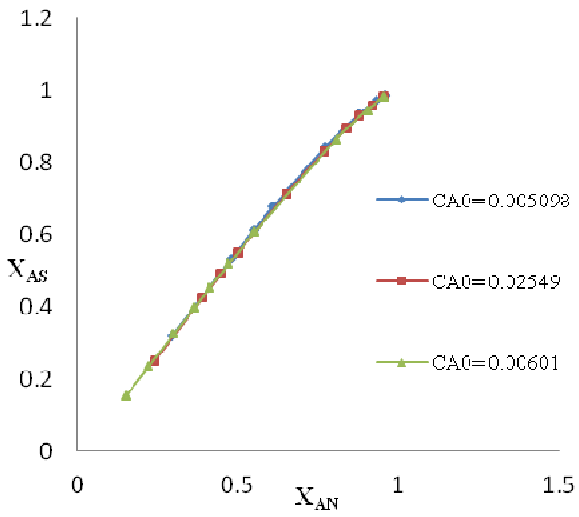

Fig: 4. Plot of numerical (X_{AN}) vs. simulation (X_{AS}) conversions for a reaction $A + 2B \rightarrow Products$ with $-r_A = kC_A C_B$ where $M \neq 2$

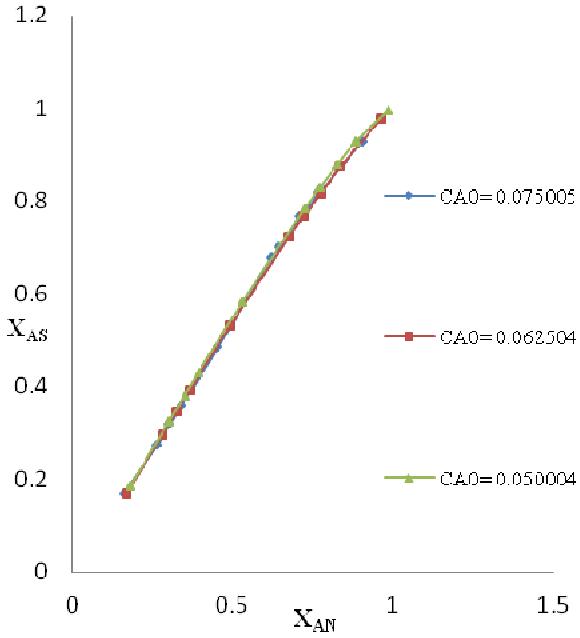

Fig: 5. Plot of numerical (X_{AN}) vs. simulation (X_{AS}) conversions for a reaction $A + B \rightarrow Products$ with $-r_A = kC_A C_B$ where $M \neq 1$

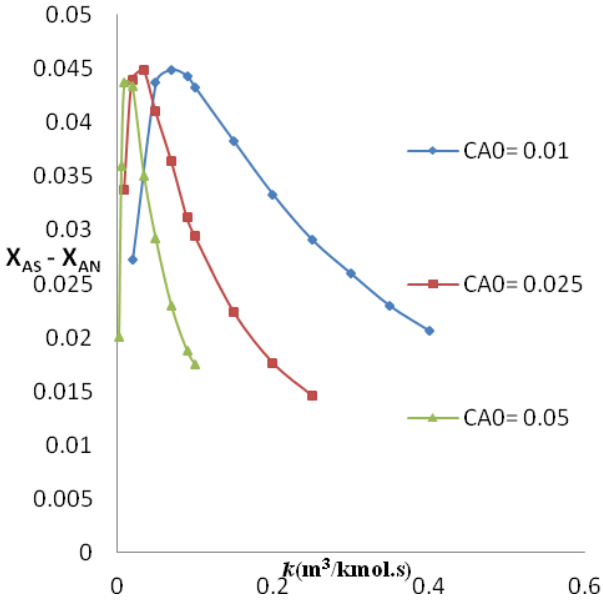

Fig: 6. Plot of Rate constant vs. the difference between simulation (X_{AS}) and numerical (X_{AN}) and conversions for a reaction $A + B \rightarrow Products$ with $-r_A = kC_A^2$.

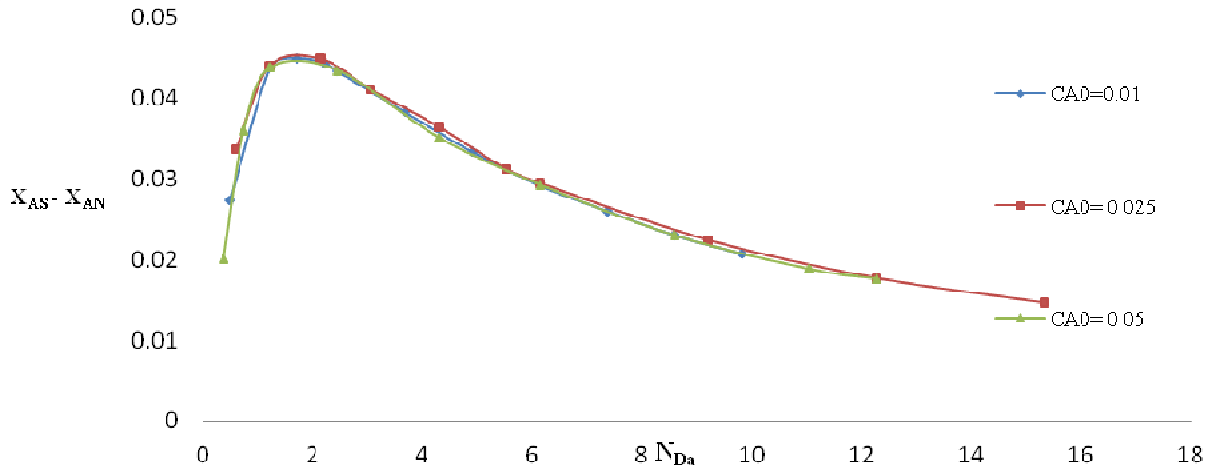

Fig: 7. Plot of Damkohler Number vs. the difference between simulation (X_{AS}) and numerical (X_{AN}) and conversions for a reaction $A + B \rightarrow Products$ with $-r_A = kC_A^2$.

[V] CONCLUSION

RTD modeling and its subsequent applications for the determination of reactor performance can be done only after the reactor is put into operation. For laminar flow reactors, the RTD can be done away with as this information is theoretically available. Using the theoretical RTD for the reactor facilitates prediction of rector performance before it is operational. This method is easier for simple kinetics but for higher order kinetics there are no analytical solutions available. The numerical model was developed, tested and validated. The comparison between numerical model predictions and simulation results agree fairly well. The reason for this small difference was identified. Difference in the conversions was found to be a function of the rate constant, initial concentration and space time. The difference in conversions was further found to be dependent on only Damkohler Number (N_{Da}). For all values of combination of initial concentrations and rate constants for which N_{Da} was same, the difference between conversions was found to be constant. The difference is found to increase, reach a maximum and then decrease with increasing N_{Da}. Maximum difference between conversions was found to occur at N_{Da} = 1.63 for all the cases tested, having kinetics $-r_A = kC_A^2$.

SYMBOLS AND NOTATION

M : Reactant ratio (C_{B0}/C_{A0}), dimensionless

k : Rate constant, $time^{-1}conc^{1-n}$(where n is the order of reaction)

C_A : Concentration of species A, $kmol/m^3$

C_{A0} : Initial concentration of species A, $kmol/m^3$

X_{AS} : Simulated conversion, dimensionless

X_{AN} : Numerical conversion, dimensionless

N_{Da} : Damkohler Number, dimensionless

t : Time, s

$\bar{t}$: Mean residence time, s

X_A : Conversion of species A, dimensionless

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