Cellular automata modeling of continuous stirred tank reactors

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Abstract−The classical dynamical systems model of continuous stirred tank reactors (CSTR) in which a first order chemical reaction takes place is reformulated in terms of the stochastic cellular automata procedure developed in the works of Seyborg [2] and Neuforth [3], which is extended by including the feed flow of chemical reactants. We show that this cellular automata model is able to simulate the dilution rate and the mixing process in the CSTR, as well as the details of the heat removal due to the jacket. The cellular automata approach is expected to be of considerable applicability at any industrial scale and especially for any type of microchemical system.

Key words: Stochastic Cellular Automata, Continuous Stirred Tank Reactors, Probabilistic Minimal Noise Rule, Kinetic Rates

INTRODUCTION

A chemical reactor could be any vessel containing chemical reactions. In general, a reactor is designed to maximize the yield of some particular products while requiring the least amount of money to purchase and operate. Normal operating expenses include energy input, energy removal, raw material costs, labor, etc. Energy changes can occur in the form of heating or cooling, or agitation. The latter is quite important because an appropriate mixing has a large influence on the yield. Therefore, the design and operation of mixing devices often determines the profitability of the whole plant.

In particular, in the widely developed continuous stirred tank reactors (CSTR) one or more fluid reagents are introduced into a tank equipped with an impeller while the reactor effluent is removed [1]. The impeller stirs the reagents to ensure proper mixing. Classical CSTR dynamical models, based on coupled deterministic ordinary differential equations (ODEs), are the usual approach to chemical systems at the macroscopic scale. They have been demonstrated to have considerable usefulness, but it should be mentioned that they are based on the common assumption that spatial inhomogeneities may be neglected. Thus, ODEs are a mean field approach and the analytical solutions of the ordinary differential equations (ODEs) provide an accurate model only in this case.

When the system is not homogeneous, application of the above assumption often yields a model that does not accurately represent the system. This is the case, for example, of CSTRs with a highly viscous medium where spatial heterogeneities exist in species concentrations, temperature, etc. Of course, the application of partial differential equations (PDEs) to model spatial inhomogeneities such as diffusion and hydrodynamic turbulence may produce accurate models. However, their solution requires advanced numerical techniques such as finite element methods. Moreover, the numerical techniques for solving the PDEs could be computationally expensive and do not account for localized stochastic phenomena. In particu-

lar, chemical systems are discrete from the microstructure until the molecular level and statistical fluctuations in concentration and temperature occur at these local scales.

Cellular automata are an attractive alternative to PDEs to model complex systems with inhomogeneities of this type. A cellular automata lattice is comprised of discrete cells whose states are functions of the previous state of the cell and its neighbors. Rules are used to update each cell by scanning the value of the cells in the neighborhood. Seyborg [2] and Neuforth [3] have shown that stochastic cellular automata models can be successfully applied to simulate first order chemical reactions. In their papers, they worked on a squared lattice of cells, each of them having a chemical reactant. The reactions are performed by considering a probability of change, from reactant A to product B, proportional to the kinetics constant that defines the chemical equation. However, this type of calculation does not apply directly to the CSTR case, where a chemical feed flow is present.

In this paper, we extend the stochastic cellular automata (CA) model to CSTRs by simulating the feed flow flux by means of a random selection of a subset of cells to which the flux conditions with respect to chemical concentration and temperature are imposed. We would like to remark that mixing in a stirred tank is complicated and not well described despite the extensive usage of dimensionless numbers and models based on ODEs [4]. Therefore, more accurate models are essential for developing and testing control strategies or even to explore new reactor geometries.

The organization of the paper is the following. Section 2 presents briefly the standard ODE-based CSTR model. Section 3 describes the CA method that we implemented for jacketed CSTRs. The simulations are displayed and briefly discussed in Section 4. The paper ends up with several concluding remarks.

THE ODE-BASED MODEL OF JACKETED CSTR

As already mentioned, we consider an ideal jacketed CSTR where the following exothermic and irreversible first-order reaction is taking place:

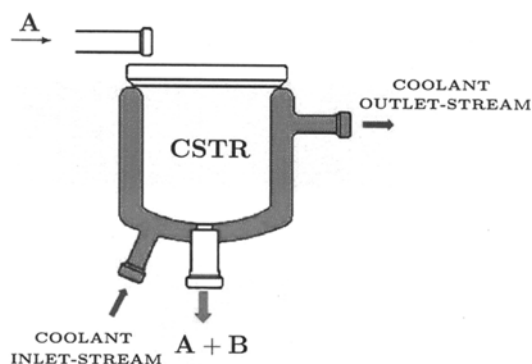†To whom correspondence should be addressed.
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Fig. 1. Schematic representation of the jacketed CSTR.

A→B

The CSTR modeling equations in dimensionless form are the following [5]

$$\frac{dX_1}{d\tau} = -\phi X_1 k(X_2) + q(X_{1f} - X_1) \qquad (1)$$

$$\frac{dX_2}{d\tau} = \beta \phi X_1 k(X_2) - (q+\delta)X_2 + \delta X_3 + qX_{2f} \qquad (2)$$

$$\frac{dX_3}{d\tau} = \frac{q_c}{\delta_1}(X_{3f} - X_3) + \frac{\delta}{\delta_1 \delta_2}(X_2 - X_3), \qquad (3)$$

where X_1, X_2, and X_3 are the dimensionless concentration of reactant A, temperature, and cooling jacket temperature, respectively. We note that it is possible to use the dimensionless coolant flow rate, q_c, to manipulate X_2.

The relationships between the dimensionless parameters and variables and the physical variables are the following:

$$k(X_2) = \exp\left(\frac{X_2}{1 + X_2\gamma^{-1}}\right), \quad \gamma = \frac{E}{RT_{f0}}, \quad X_3 = \frac{T_c - T_{f0}}{T_{f0}}\gamma, \quad X_2 = \frac{T - T_{f0}}{T_{f0}}\gamma,$$

$$\beta = \frac{(-\Delta H)C_f}{\rho C_p t_{f0}}, \quad \delta = \frac{UA}{\rho C_p Q_0}, \quad \phi = \frac{V}{Q_0}k_0 e^{-\gamma}, \quad X_1 = \frac{C}{C_{f0}}, \quad X_{1f} = \frac{C_f}{C_{f0}},$$

$$X_{2f} = \frac{T_f - T_{f0}}{T_{f0}}\gamma, \quad \delta_1 = \frac{V_c}{V}, \quad \tau = \frac{Q_0}{V}t, \quad X_{3f} = \frac{T_{cf} - T_{f0}}{T_{f0}}\gamma, \quad \delta_2 = \frac{\rho_c C_{pc}}{\rho C_p},$$

where the meaning and the nominal values of these parameters are given in Table 1. In the rest of the paper we shall use the solution of this ODE model as the theoretical case with which the CA simulations will be compared.

STOCHASTIC CA MODEL FOR JACKETED CSTR

The process simulated in this work is the exothermic reaction that converts a chemical A into a product B in a jacketed CSTR. Our model is composed of three squared lattices of cells, all of the same size. The first lattice that we call the chemical lattice is for chemicals A and B and provides the chemical distribution in the tank reactor. This distribution is given as follows: in each cell there is only one unit of reactant A or one unit of product B (not necessarily representing a single molecule), under the condition that all cells are occupied. The second lattice is the tank temperature lattice. It contains the temperatures T_{ij} in real-numbered values. The chemical lattice and the temperature lattice are of the same dimension and their cells are in one-to-one correspondence.

The third lattice represents the coolant system. This lattice is of the same dimension as the previous ones and therefore each temperature tank cell is in "contact" with a coolant jacket cell.

Table 1. Parameters of the model

Symbol	Meaning	Value (arb. units)
C	Reactor composition	0.001
C_f	Feed composition	1.0
q	Dimensionless reactor feed flow rate	1.0
q_c	Dimensionless coolant flow rate	1.65
q_{cs}	Steady-state value of Q	1.0
T	Reactor temperature	1.0
T_c	Heat transfer coefficient	1.0
UA	Heat transfer coeff. times the heat transfer area	1.0
V	Reactor volume	1.0
V_c	Cooling jacket volume	1.0
X_{1f}	Dimensionless feed concentration	1.0
X_{2f}	Dimensionless feed temperature	0.0
X_{3f}	Dimensionless coolant feed temperature	1.0
β	Dimensionless heat of reaction	8.0
δ	Dimensionless heat transfer coefficient	0.3
δ_1	Dimensionless volume ratio	0.1
δ_2	Dimensionless density times the heat capacity ratio	0.5
φ	Hill's threshold parameter	0.072
γ	Dimensionless activation energy	20.0
$\rho_c C_{pc}$	Density multiplied by the heat capacity of the coolant	1.0
τ	Dimensionless time	--

In our model the first process in each time step is the irreversible conversion of chemical A into product B. The conversion rate is determined by $\varphi k(X_2)$ as in [5], where X_2 is the average temperature of the tank temperature lattice. This first order kinetics "constant" is multiplied by the time step in order to get the proportion of the reactant A that is expected to be converted into product B in each evolution step.

If the time step is small enough, this number could be also considered as the probability that a molecule of chemical A is converted in product B. Such proportion is compared with a randomly generated number, one different random number for each cell in the lattice containing reactant A. If the random number is less than the proportion, the reactant A is changed for product B in the cell. Since the reaction is exothermic, the temperature value in the temperature array is increased by β (according to Eq. (2)) in the corresponding cell.

The second simulated process is the tank temperature diffusion that can be considered as an energy diffusion. It can be performed by means of finite differences, but in order to obtain an almost ODE-independent model we have implemented a moving average method, where the value of the temperature in a cell at the next time step is the average temperature of its neighborhood. This procedure gives similar results to those of finite differences, as shown by Weimar for reaction-diffusion systems simulated by cellular automata [6]. We used a square neighborhood formed by $(2R+1)^2$ cells, where R is the number of steps that we have to walk from the center of the cell in order to reach the most far horizontal (vertical) cell in the neighborhood.

The third simulated process is the tank feed flow. We have simulated the feed flow rate q in a stochastic way. In order to get an approximation to the proportion of the tank that must be replaced by the incoming flow, q is multiplied by the time step and by the total number of cells in the lattice. This gives us a real number x. Then, following Weimar [6], we used a probabilistic minimal noise rule, $i.e.$, we defined the probability p=x−[x] (where [x] is the maximal integer number ≤x) in order to decide if [x] or [x] +1 cells will be replaced by the flow. We chose [x] with probability 1−p and [x] +1 with probability p. This method conserves the proportion x in a statistical way. Subsequently, a cell is selected in a random way, by means of two random numbers which are used to select a row and a column, in such a way that all cells have the same probability of being selected. If the cell has been selected in the same time step, a new selection is made. This is repeated until we have reached the number of cells that must be replaced. Finally, the selected cells are changed in the temperature lattice by the feed flow temperature X_{2f}, and in the reactive lattice is put a unit of reactant A with probability X_{1f}, that represents the concentration of chemical A in the feed flow. In the simulations presented in this work we used $X_{1f}=1$. This method of flow simulation could be improved in several ways, in order to simulate different tank geometries or for showing the flow direction. However, we want only to show that the CA method could fit the CSTR behavior in a very good approximation, with the advantage of spatial analysis.

The fourth simulated process is the energy interchange between the tank and the jacket. This has been done by directly calculating the energy/temperature interchange between each tank temperature cell and its corresponding jacket temperature cell. This interchange
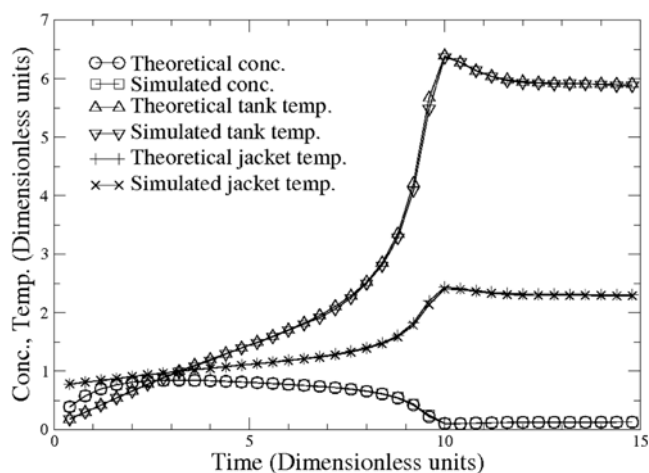

Fig. 2. Comparison between the curves from the differential equations and the curves obtained from simulations with the CA approach. Initial values are: $X_1=0.1$, $X_2=0.1$, $X_3=0.1$. For the six curves: 37 points separated by a dimensionless time of 0.4 were taken from 20000-point simulations with a dimensionless time step of 0.001.

is dictated by the difference between the two temperatures and it is weighted by δ, δ_1, and δ_2 as in Eqs. (1) and (2).

The fifth and the sixth simulated processes are the coolant flow and the coolant temperature diffusion, respectively. Both of them are performed in a similar way as for the concentration and temperature tank.

SIMULATIONS

In this section we first present a comparison between the curves obtained by differential equations and those obtained from the implemented cellular automata model (see Fig. 2). It is clear that cellular automata simulations resemble with excellent agreement the values for the concentration of chemical A, the tank temperature and the jacket temperature at all times. Using a time step of 0.001 it is shown that the curves coincide at the initial time, transient time and for stable state. We have found that we can maintain this remarkable fitness by properly adjusting the time step to a sufficiently small value.

When a kinetic constant based on the average temperature is used, it is implicitly assumed that the mixing in the CSTR is perfect, with any temperature inhomogeneities vanishing. One could ask what could be the change in the tank behavior if the mixing is almost perfect. It could be studied by using a model that considers the spatial distribution of temperature. We studied this effect by calculating the kinetic constant in a local manner by taking the temperature from each cell in the temperature lattice. The effect for a $1,000 \times 1,000$ cells array and a time step of 0.001 is shown in Fig. 3. It could be observed that the tank temperature curve for the perfect mixing and the one for the locally calculated kinetic constant are the same for almost all times. However, they separate during the transient period, leading to a reduction in the magnitude of the peak and a little delay in its appearance. The curve was calculated with a tank temperature diffusion process per time step with an R=1 neighborhood. If more diffusion steps are used per time step, the curves obtained tend to
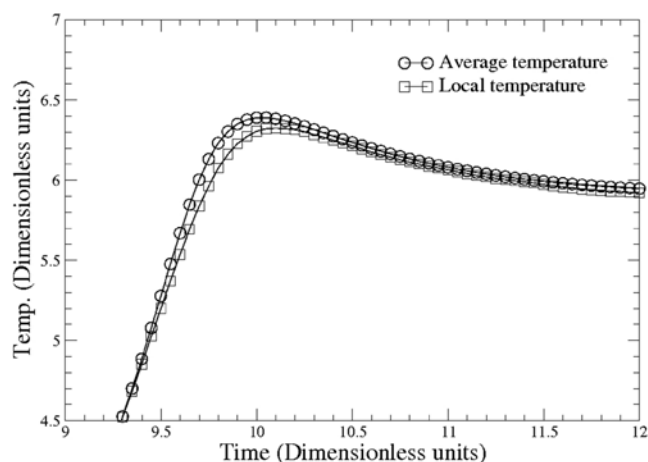
Fig. 3. Comparison between the tank temperature evolution curves for a kinetic constant based on average temperature and for kinetics constant calculated on the base of the local temperature. Initial values are: X_1=0.1, X_2=0.1, X_3=0.1. The array is of 1,000×1,000 cells and the dimensionless time step is of 0.001; one temperature diffusion step per time step.
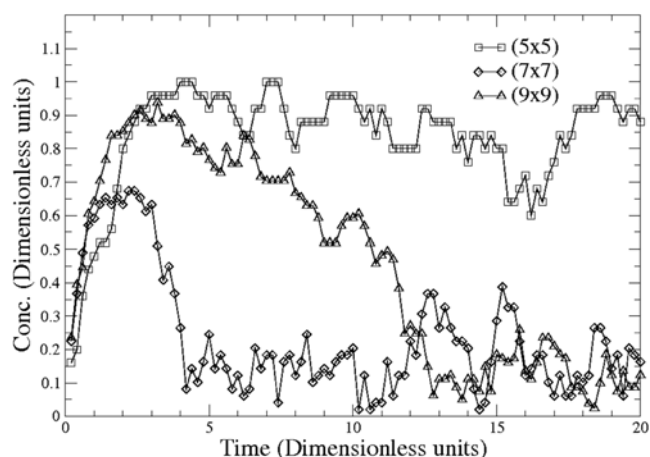

Fig. 4. Behaviors that could be found in systems with small number of elements (cells for the CAs and clusters of molecules in the real case). These behaviors are different from the expected one for systems with a large number of elements. The initial values are X_1=0.1, X_2=0.1, X_3=0.1. The employed time step is 0.001. One hundred points separated by a time lag of 0.2 were taken from 20000-point simulations with a time step of 0.001; one temperature diffusion step per time step.

the theoretical one as is expected. Besides, it could be also interpreted as the effect of a perfectly mixed tank, but with material where each component tends to maintain its energy.

The ODEs generally represent the characteristics of the global system, where it has a large enough number of elements such that the statistical fluctuations are of small amplitude.

However, when the system size and the quantity of elements are diminished, the statistical fluctuations could be of increasing importance. In this way, another advantage of the cellular automata method proposed here is its flexibility with respect to the reactor size, and its stochastic nature, that allows one to study how much the system could be affected by the initial conditions and by the stochastic features of the process.

We have performed several simulations applying the parameters presented above in lattices of very small dimensions, N×N with N≤20, in which case we can think of the CA stochastic model as a good description of a catalytic surface dividing two regions, one carrying the chemical A and the other as a temperature reservoir. Our model is a simple approach, useful as a first approximation, in the analysis and study of microreactors or even nanoreactors. We recall that the usage of microreactors for *in situ* and on-demand chemical production is gaining increasing importance as the field of microreaction engineering has already demonstrated a potential to impact a wide spectrum of chemical, biological, and process system applications [7]. There are already many successfully developed microreactors for chemical applications such as partial oxidation reactions [8], phosgene synthesis [9], multiphase processing [10], and (bio)chemical detection [11].

Fig. 4 displays the variability that could be found in CSTR systems at small scales. It is clear evidence that the statistical fluctuations are a primordial issue at this scale. In addition, one could notice that the dynamical behavior could be totally different from the expected behavior of a larger system, e.g., 1,000×1,000 cells lattice.

Finally, the study of small systems by direct simulation using stochastic simulations could give us insight in how an open CSTR sys-
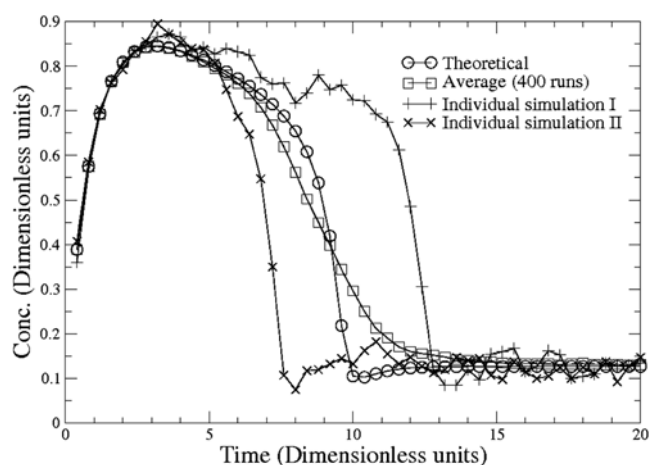

Fig. 5. Different concentration behaviors of chemical A for systems of the same size (20×20) that are treated by the same method and could be the underlying dynamic characteristic of microreactors. Two individual simulations corresponding to two different initial distributions of the chemical A over the cells are displayed together with the average curve of 400 individual simulations as well as the theoretical curve corresponding to the ODE-based model. The initial configuration and the stochasticity and discreteness introduced in the model lead to a time-distributed behavior. The initial values are: X_1=0.1, X_2=0.1, X_3=0.1. The time step is 0.001; one temperature diffusion step per time step.

tem could behave when the statistical fluctuations and the initial configuration are important. In Fig. 5 one can see that the possible behaviors of a system of size 20×20 have large deviations from the average and the theoretical values.

This kind of variability is not provided by pure ODEs (without a

stochastic term). Moreover, the discreteness of the CA procedure adds an important contribution to these differences.

Both the stochastic part and the discreteness part are intrinsic features of small scales. We think that this stochastic CA approach could be an important simulating tool for this small-scale variability and for testing control strategies in reduced environments. This is because CA approaches could be seen as an intermediate step between the ODEs models and the specific experimental situation.

CONCLUDING REMARKS

A cellular automata approach for a CSTR with a cooling jacket has been presented in this paper. It is able to reproduce the CSTR dynamical behavior calculated by ODE's with a good approximation and in an easy way. The presented stochastic model allows us to study what could be the behavior of the variables of the tank when the reaction probability depends on the local temperature. It also gives us an approach to study systems with few elements, such as micro and nanoreactors, for example catalytic membranes separating two phases. The main advantages of the CA approach presented here are its stochastic nature and the direct involvement of a spatial structure. This also represents a tool for studying the role of initial configuration and stochastic fluctuations in systems with few elements. Additionally, the CA approach is a clear improvement in CSTR modeling, and moreover can be applied to different reactor and jacket geometries, as well as for considering in more detail the real mass flow in the tank reactor-geometry.

We have found that in our CA implementation at lattice dimensions beyond 400×400 and below time steps of 0.001 the ODE-based solutions (curves) are very well reproduced, thus indicating that for those values the stochastic noise is very small. On the other hand, at small lattice dimensions the CA procedure gives strong fluctuations which are due to the CA discretization that sometimes could match the discretization of nature at those scales.

Finally, we mention that CA-type models can be used to analyze local microorganism densities which are directly related to the production yields of important fermentation products, such as bacterial cellulose obtained in generalized stirred reactors [12].

ACKNOWLEDGMENT

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