

Modelling and Optimisation of Distributed-Parameter Batch and Semi-batch Reactor Systems

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Abstract

Macro-mixing effects in batch and semi-batch reactors are investigated by constructing 3-dimensional models using a network of zones (NoZ) discretisation. System dynamics including volume changes due to continuous feeding are successfully predicted. Detailed flow fields are calculated from phenomenological correlations which include parameters such as reactor size and configuration, impeller type and speed and fluid physical properties. The Proper Orthogonal Decomposition method is subsequently applied to extract reduced models from the large-scale NoZ-based ones that can be used for computationally efficient design, optimisation and optimal control studies.

Keywords: network of zones, non-ideal mixing, proper orthogonal decomposition

1. Introduction

Batch and semi-batch reactors are increasingly employed in the industrial production of fine and specialty chemicals, pharmaceuticals, polymers and crystals due to their economic efficiency, including low capital investment and typically high yields, and their versatility to operate with a range of reactants and products. Nevertheless, due to the complex system dynamics involving turbulent flows and mixing phenomena, optimal and safe reactor scale-up and operation are challenging tasks, increasing considerably the uncertainties in reactor design and set-up (Zaldívar et al, 1996, Brucato et al. 2000). Modelling approaches that have been proposed to predict and optimise batch reactor performance range from simple compartment-based models (David et al., 1992, Cui et al., 1996) which use only a few parameters and can yield good agreement with experimental results, but can not give detailed insight for detailed design, to more complex models involving detailed CFD-based flow field simulations (e.g. Brucato et al., 2000, Bakker et al., 2001). Turbulent CFD models can give detailed flow information, but they are computationally intensive, commonly used k - ϵ models introduce inaccuracies by assuming isotropic turbulence and more advanced models (e.g. LES) require parameter tuning through experimental validation (Vrabel et al., 2000). In this work we have adopted the network-of-zones (NoZ) approach (Nienow et al. 1992, Rahimi and Mann, 2001, Hristov and Mann, 2002), where the reactor is finely discretised in a number of cells. The NoZ model can successfully predict the effects of

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macro-mixing, reactor design and operating conditions. It can be integrated with CFD-based flow calculations (Brucato et al 2000) as well as with models describing micro-mixing and drop or bubble dispersion and movement phenomena. Here, we calculate detailed flow fields in a computationally efficient way by using complex correlations involving a large range of design parameters (Platzer and Noll, 1988) and we have developed a new network structure to deal with reaction volume change effects.

2. Network of zones model

Existing NoZ models assume that the reaction volume in every zone does not change during a batch cycle. However, in reality there are changes due to reaction, feeding or mass transport. In this work an improved model is proposed where the network of zones is constructed based on the volume of the reactor. Fig.1 depicts the structure of this new model, which involves 3 types of zones: a. Empty zones above the fluid surface; b. half-empty zones containing the fluid surface and c. Fluid-occupied zones below the surface level. If the reaction volume changes during the batch process, the surface of the fluid will move up into the empty zones or down towards the fluid-covered zones leaving more empty zones at the top. Accordingly, the mathematical description of this model includes equations describing reaction volume changes. Consider the mass transfer between one zone (i, j, k) and its adjacent zones. Equations (1), (2) and (3) are three ordinary differential equations (ODEs) describing the rate of change of volume $V_{(i,j,k)}$, molar amounts, $N_{(i,j,k)}^l$, and concentrations $C_{(i,j,k)}^l$ of each component l , in this zone.

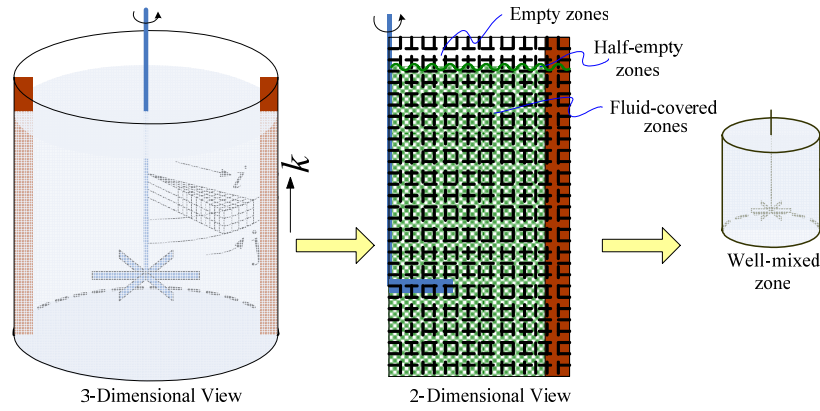


Figure 1. Structure of the network of zones model

$$\begin{aligned} \frac{dN_{(i,j,k)}^l}{dt} = & Vol_{-} flow_{-} in_{(i-1,j,k)} \times C_{(i-1,j,k)}^l + Vol_{-} flow_{-} in_{(i+1,j,k)} \times C_{(i+1,j,k)}^l + \\ & Vol_{-} flow_{-} in_{(i,j-1,k)} \times C_{(i,j-1,k)}^l + Vol_{-} flow_{-} in_{(i,j+1,k)} \times C_{(i,j+1,k)}^l + \\ & Vol_{-} flow_{-} in_{(i,j,k-1)} \times C_{(i,j,k-1)}^l + Vol_{-} flow_{-} in_{(i,j,k+1)} \times C_{(i,j,k+1)}^l - \\ & Vol_{-} flow_{-} out_{(i,j,k)} \times C_{(i,j,k)}^l \pm Vol_{(i,j,k)} \times \sum_{p=1}^{n-r} Rct_{-} rate_p^l + \sum_{q=1}^{n-f} Fd_{-} rate_q \end{aligned} \quad (1)$$

$$\frac{dV_{(i,j,k)}}{dt} = \sum_{l=1}^{n-c} \text{Molar_vol}^l \times \frac{dN_{(i,j,k)}^l}{dt} \quad (2)$$

$$\frac{dC_{(i,j,k)}^l}{dt} = \frac{1}{V_{(i,j,k)}} \left(\frac{dN_{(i,j,k)}^l}{dT} - C_{(i,j,k)}^l \times \frac{dV_{(i,j,k)}}{dt} \right) \quad (3)$$

Vol_flow_in and Vol_flow_out are the volumetric convective flow rates through the surfaces of the zone, calculated by integrating the flow field across each surface. Rct_rate and Fd_rate and $Molar_vol^l$ are the reaction rate, feed rate and component l molar volume, respectively. Each zone is assumed to be well-mixed, its volume is determined by the level of discretisation and remains fixed during the simulation. For the fluid-covered zones, reaction volume is the same as the zone volume, so the volume changes from equation (2) are used to compute the new level of the fluid surface. Beyond a volume change threshold the flow field is re-computed. Note that only equations (2) and (3) need to be solved being both expressed in terms of equation (1).

2.1 Flow field computation

In several works employing NoZ models, the flow rate through surfaces of zones was calculated only based on impeller overall circulation convective capacity (Nienow et al., 1992; Rahimi and Mann, 2001; Hristov and Mann, 2002). This flow rate was then equally distributed through all the zones. There are several drawbacks in this method. It can not capture velocity variations at different regions inside the reactor vessel and does not fully exploit the benefits of the NoZ model. Also, it uses too few reactor configuration parameters, thus it cannot be used for reactor design and optimisation. Usually other parameters like tank size, impeller type, baffle number and size are of great interest in reactor engineering. Here, a sophisticated correlation system (Desouza and Pike, 1972; Platzer and Noll, 1988) is used which divides the flow field into three characteristic model parts: (1) rotational flow due to impeller rotation; (2) Circulating flow in the “bulk” cells; (3) distinct jet flow (near the impeller). Different correlations have been developed for these 3 parts. The correlations can give detailed 3-D flow field information, i.e. velocity vectors at every cell in the vessel. The parameters involved include reactor configuration (tank size, impeller type, size and position and baffle number and size), operating conditions (impeller speed, fluid depth and feed position) and fluid properties (density and viscosity). This model has been tested against CFD-based calculations giving results of comparable accuracy for a wide range of parameters (Zheng et al 2004). Furthermore it is much more computationally efficient.

3. Case study

A system proposed by Nienow et al. (1992) in order to characterise imperfect macro-mixing and partial segregation in a stirred semi-batch reactor is considered. The predictions of our model were compared with experimental results and simulation results reported in the literature. The chemistry is based on a pair of parallel reactions



where A (a diazonium salt) is initially charged in the reactor and B (a pyrazolone) is added continuously at a constant rate. R, a dyestuff is the product and S the unwanted

by-product. The reaction kinetics, operating conditions and kinetic parameters are reported in (Nienow et al., 1992). The height and diameter D of the vessel are both 0.3m. Four 0.1D strip baffles were used with a Rushton turbine with diameter $D_1=D/3$ and clearance $C=D/3$. A 3-D NoZ model was constructed using 20 zones in each direction (axial, radial and circumferential -8000 zones in total) resulting in a system of 48000 ODEs which were integrated in time using DASPK (Maly and Petzold, 1996). Fig.2 shows a comparison between results from the 3-D network (diamonds) experimental results (squares) and simulation results from the literature (triangles- Nienow et al 1992) for a range of impeller rotation speeds. Simulations assuming ideal mixing (circles) over-predict the yield. Our 3-D simulations agree very well with the experiments and are in better agreement than the literature results, which are, however, close since the volume change effects are small in this case. Further parametric studies have shown that better yield can be achieved by supplying both feeds continuously from the same feed position near the tip of the impeller. These results along with results from a second case study where volume change effects were more pronounced (Paul and Treybal 1972) are presented in a forthcoming publication (Zheng et al, 2004).

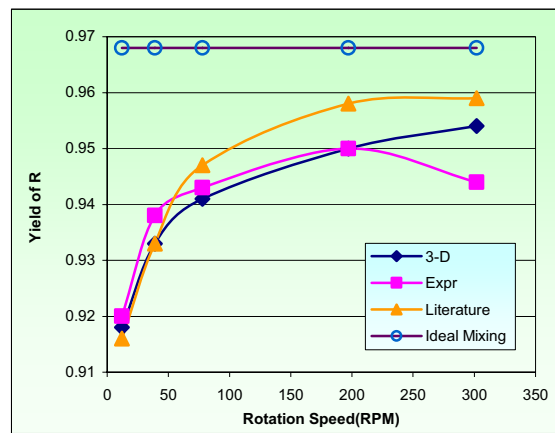


Figure 2. Comparison between yield predictions from our 3-D model, and experimental and simulation results from the literature.

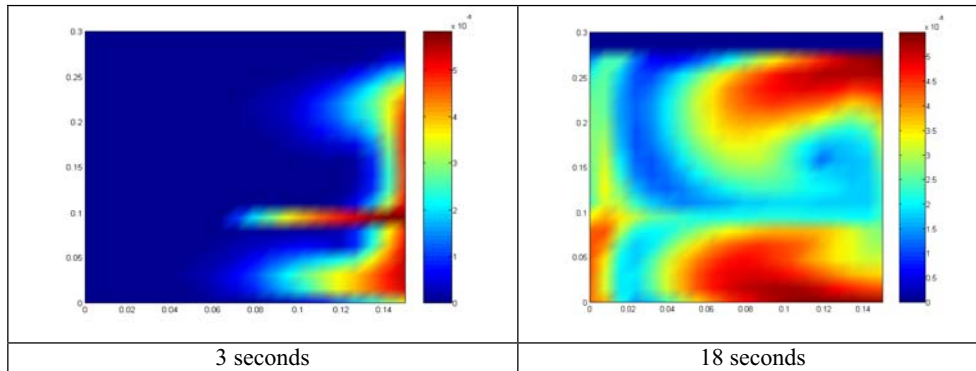


Figure 3. Concentration snapshots of the product R on a vertical plane in the reactor vessel at $t=3$ and $t=18$ s. The blue (red) colour denotes low (high) concentration.

Fig. 3 shows concentration distribution profiles of species R at a vertical plane inside the reactor at $t=3$ and 18 s. The right side is the reactor centreline. The impeller rotation speed was 78 RPM. Blue (red) colour denotes low (high) concentration. The concentration at the top empty zones is zero. As it can be seen, areas of lower mixing intensity are the corners of the reactor, the impeller shaft and the circulating zones. As time progresses reactants in these parts eventually participate in the reactions and are converted to products or by-products.

3. Reduced model

The NoZ model coupled with flow correlations typically results to systems containing (hundreds of) thousands of ODEs. The simulation of large-scale ODE-based systems is nowadays achievable in realistic CPU times with large yet reasonable memory requirements. Nevertheless, optimisation studies and optimal control design and implementation cannot be based on such large-scale systems since a huge number of function evaluations is required. In this work we have employed the Proper Orthogonal Decomposition method (POD) (Holmes et al., 1996) to extract accurate low-order models from the full-scale ones. In POD a small number of semi-empirical eigenfunctions are computed from a database of detailed full-scale simulations (or even experiments) that can capture the energy of the system i.e. can accurately describe the system in the parametric range of interest. The dynamic low-order model can then be obtained by a Galerkin projection of the governing equations onto these few basis functions. POD has been used successfully in a number of works (e.g. Rowley et al, 2004; Cizmas et al, 2003; Shvartsman et al 2000). Here the *scalar-valued* method is employed (Rowley et al. 2004) computing POD modes for each variable (concentrations and reaction volume). We have constructed a simulation database for the case study presented above, by performing simulations using the NoZ model at 3 different rotation speeds: 39 RPM, 197 RPM and 302 RPM recording snapshots every 0.5s. It was found that 20 basis functions for each species (100 in total) and only 1 basis function for the volume were sufficient to capture 99.9 % of the energy of the system. A Galerkin projection of equations (1)-(3) onto these eigenfunctions produced a reduced model of only 101 ODEs that can accurately predict the system behaviour.

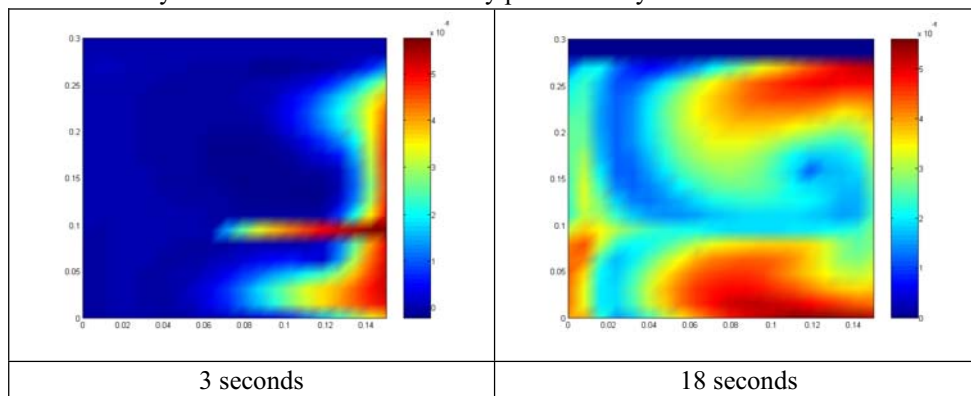


Figure 4. Concentration snapshots of the product R at $t=3$ & 18 s on a vertical plane in the reactor obtained from the reduced model. The blue (red) colour denotes low (high) concentration.

In Fig. 4 concentration profiles obtained from the reduced model at the same conditions as the profiles showed in Fig. 3 are depicted. As it can be seen the agreement between the full-scale and the reduced model results is excellent both for the short term (3s) and for the longer term (18s) dynamics. It is worthwhile to note that the case simulated here (impeller speed 78 RPM) is not included in the simulation database. Results of this reduced model at other conditions also show the same agreement with results from the full model. It can be concluded that the reduced model can predict the system behaviour very well requiring much less computer memory and CPU time.

4. Conclusions

We have constructed 3-D models of batch and semi-batch reactors using a network of zones discretisation. The computational domain is discretised in an appropriately large number of cells and local velocity distributions are computed by detailed flow correlations. Mass balances coupled with volumetric changes are then superimposed onto the computed flow resulting in large-scale ODE-based systems. The model can successfully predict the effects of non-ideal macro-mixing and includes a large number of important design and operating parameters than can be used for system scale-up, optimisation and control. The POD method was subsequently used to extract reduced computationally-amenable models from the full-scale ones that can be efficiently employed in parametric studies, model-based optimisation and optimal control.

References

- Bakker, A., A.H. Haidari and L.M. Oshinowo 2001, Chem. Eng. Prog., 97, 45.
 Brucato A., M. Ciofalo, F. Grisafi, R. Tocco 2000, Chem. Eng. Sci. 55, 291.
 Cizmas, P.G., A. Palacios, T. O'Brien and M. Syamlal 2003, Chem. Eng. Sci. 58, 4417.
 Cui, Y.Q., R.G.J.M. van der Lans, H.J. Noorman and K. Luyben 1996, Chem. Eng. Res. Des. 74, 261.
 David, R., H. Muhr and J. Villiermaux 1992, Chem. Eng. Sci. 47, 2841.
 Desouza, A. and R.W. Pike 1972. Can. J. Chem. Eng. 50, 15.
 Holmes P., J.L. Lumley and G. Berkooz 1996, Turbulence, coherent structures, dynamical systems and symmetry, Cambridge University Press.
 Hristov, H.V. and R. Mann 2002, IChemE, 80, 872.
 Maly, T. and L.R. Petzold 1996, Appl. Numer. Math. 20, 57.
 Nienow, A.W., S.M. Drain, A.P. Boyes, R. Mann, A.M. El-Hamouz, and K.J. Carpenter 1992. Chem. Eng. Sci. 47, 2825.
 Paul, E. L. and R.E. Treybal 1971 AIChE J. 17, 718.
 Platzer, B. and G. Noll 1988. Chem. Eng. Proc. 23, 13.
 Rahimi, M. and R. Mann 2001 Chem. Eng. Sci. 56, 763.
 Rowley, C.W., T. Colonius and R.M. Murray 2004 Physica D: Nonlin Phen. 189, 119.
 Shvartsman, S. Y., C. Theodoropoulos, R. Rico-Martínez, I.G. Kevrekidis, E.S. Titi and T.J. Mountziaris 2000 J. Proc. Control, 10, 177.
 Vrabel P, R.G.J.M van der Lans, K.Ch.A.M. Luyben, L. Boon and A.W. Nienow 2000, Chem. Eng. Sci. 55, 5881.
 Zaldívar, J. M., H. Hernández and C. Barcons 1996 Thermochemica Acta, 289, 267.
 Zheng X., R. Smith and C. Theodoropoulos, *Manuscript in preparation*.