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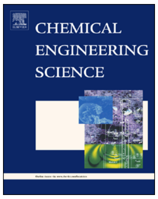

# The generalized coalescence/redispersion micromixing model. A multiscale approach

Béla G. Lakatos*

*Department of Process Engineering, University of Pannonia, PO Box 158, H-8201 Veszprém, Hungary*

## HIGHLIGHTS

- Improvement on the generalized coalescence/redispersion (gCR) micromixing model.
- Analysis of the gCR model in context of the three-scale mixing concept.
- Model of collision induced mass exchange by random parameter differential equations.
- The gCR model is validated using quasi-linear and consecutive–competitive reactions.
- A two-zone model with different micromixing rates is presented and analysed.

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## ABSTRACT

The generalised coalescence/redispersion (gCR) micromixing model is formulated in the framework of the three scale mixing concept of stirred tank reactors. It is developed on the basis of a random coefficient differential equation system describing mass exchange interactions between the Kolmogorov scale fluid elements the average behaviour of which is modelled using the population balance approach. The model provides closed terms for both the chemical reactions and micromixing and it allows computing processes with either pre-mixed or unmixed feeds of reactors. A qualitative picture of producing the fluid elements in the eddy space as Kolmogorov microscale eddies, interpreted as mesomixing of the three scale mixing model is outlined.

The population balance equation is reduced to a moment equation system for joint moments of concentrations using the standard moment method which is closed by cumulant-neglect closure.

The gCR model is validated by fitting the model to literature data of bimolecular quasi-linear chemical reactions (Miyawaki et al., 1975. J. Chem. Eng. Jpn. 8, 63–68; Takao et al., 1978. J. Chem. Eng. Jpn. 11, 481–486), and of a quasi-linear consecutive–competitive chemical reaction system (Paul and Treybal, 1971. AIChE J. 17, 718–724) in stirred tank reactors. It is pointed out by simulation that in case of non-homogeneous micromixing space of stirred tank reactor different inlets–outlet configurations of mixing experiments produce significantly different concentration responses, and using the gCR model micromixing inhomogeneities can be explored.

## 1. Introduction

Fluid-phase mixing by turbulent motion is of fundamental importance in many unit operations of chemical and process industries. The basic equation for turbulent mixing is the mass conservation equation of chemical species the numerical treatment of which exhibits all the difficulties that we found when solving the nonlinear Navier–Stokes equation describing the turbulent motion. However, taking into account that turbulent mixing can be considered as fluid motion at three scales, a molecular, eddy and bulk scales, allows treating this problem as a multi-scale process using micro-, meso- and macro-mixing.

Fluids under turbulent conditions can be considered as consisting of eddies of diverse sizes. In stirred tank reactors the largest eddies have sizes of the same order of magnitude as the vessel or the impeller whilst the smallest eddies have sizes characterized by the Kolmogorov length scale where, practically, the molecular diffusion takes place causing the homogeneity of the scalar entities. Between these extremes there exists a whole range of eddy sizes (Pope, 2000; Durbin and Pettersson Reif, 2001; Fox, 2003) which can be represented as a large population of dispersed objects therefore the population balance approach appears to be an applicable tool for modelling this process as well.

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* Tel.: +36 88 624210; fax: +36 88 624171.
*E-mail address:* lakatos@fmt.uni-pannon.hu

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Population balance mixing models in different contexts were developed by Evangelista et al. (1969) extending the coalescence/redispersion model for several chemical species in homogeneous chemical reactors, by Chen (1971) for describing micromixing in unsteady state flow reactors in terms of internal age and life expectation distributions, by Kattan and Adler (1967) combining the coalescence/redispersion model with the residual life times, by Takao et al. (1978) investigating micromixing in batch reactors using an interaction with the mean type model. Ritchie and Tobgy (1978) presented a review of population balance modelling of mixing phenomena in homogeneous chemical reactors. Madras and McCoy (2004, 2005) presented a population for turbulent mixing in stirred vessels, characterizing the process of mixing by the increasing interfacial area between the dispersed and bulk fluid. The generalized coalescence/redispersion (gCR) model (Lakatos, 2008; Lakatos et al., 2011) has also been formulated in the framework of the population balance approach.

The gCR model is a stochastic modification of the deterministic coalescence/redispersion (CR) model proposed first by Harada et al. (1962) and Curl (1963). The CR model has widely been applied also as a closure model of the mixing terms in the population density function approach (PDF). For that purpose Janicka et al. (1979) and Dopazo (1979) proposed modifications to Curl's model assuming that the extent of the mass exchange between a pair of fluid elements is determined by a uniformly distributed random variable. Pope (1982) generalized the model by Janicka et al. (1979) in one-dimensional case which was extended by Kaminsky et al. (1996) for more independent variables using the PDF framework. Multi-species CR models were applied for closing the mixing terms in PDF's by Balthasar and Mauss (2002) and Möbus et al. (2003) in modelling turbulent combustion. Several micromixing models of PDF description were compared by Meyer and Jenny (2009), and a detailed analysis of the multi-species CR models were presented by Haworth (2010).

The gCR model was developed under the assumption that production of fluid elements, identified in the model as the smallest eddies in a turbulent process is an instantaneous process. Although this modelling reduction often proves to be justified but this process, in principle, occurs by transformation of macro-scale eddies across scales to the micro-scale ones therefore considering the problem accounting for the multi-scale characteristics of turbulence (Li and Kwauk, 2003; Li et al., 2004; She, 2007) allows setting the gCR model in a new perspective of a complex three-scale mixing model.

The aim of the paper is to formulate and analyse the generalized coalescence/redispersion model of micromixing in the framework of multi-scale approach of complex systems taking into consideration the transformation process of turbulent eddies across the scales. Micro-scale interactions between the fluid elements defined as micro-scale objects are described as a dynamic process with random coefficients while production of the micro-scale objects is treated as mesoscale mixing. An infinite order moment equation model is derived for the joint moments of concentrations of chemical species from which a second order moment equation reduction is obtained by means of cumulant-neglect closure. The properties and behaviour of the model are studied by simulation, and predictions of the gCR model are evaluated and validated using experimental data of quasi-linear bimolecular and consecutive–competitive chemical reactions taken from the literature.

## 2. Characterization of micro-scale

Consider a continuous isothermal chemical reactor in which fluid mixing is assumed to occur by intensive stirring. Stirring the reactor as well as the fluid fed through the inlet pipe generate a complex turbulent mixing process which is modelled as a superimposed eddy field in depth from the largest, energy containing eddies to the smallest ones under the given turbulent conditions. This eddy field is composed of a continuous distribution of length scales the behaviour of which is governed mainly by the small scale of flow. Here, two characteristic scales play important roles: the energy-containing eddies are of scale of impeller $L_{\mathrm{imp}}$ and/or the feed pipe $L_{\mathrm{in}}$, and the smallest eddies in the system the characteristic size range of which is given by the Kolmogorov length scale of turbulence.

$$\eta \propto \left(\frac{\nu^3}{\varepsilon}\right)^{1/4} \tag{1}$$

where $\nu$ denotes the average kinematic viscosity and $\varepsilon$ stands for the energy dissipation rate. In the paper, the Kolmogorov scale eddies will be termed fluid elements.

When the process of production and distribution of large scale eddies occurs uniformly over the whole fluid volume of the vessel then the eddy space becomes homogeneous. If the turbulence is assumed to be locally isotropic then the motion of fluid elements in the physical phase can be treated with no orientation effects and inertia-dominated fully stochastic behaviour. In this case the fluid elements are treated as individual objects of the eddy space having properties given by the vector of concentrations $\boldsymbol{c}_j \in \boldsymbol{R}^K$ of possible $K \geq 1$ chemical species carried by the fluid elements $j = 1,2,3\ldots$. Since the volume of fluid elements is assumed to be constant and the system is isothermal the volume and temperature as properties of fluid elements are not denoted here.

The fluid elements moving randomly in the physical space interact with each other by collisions and may exchange some mass of species while chemical reactions may occur inside. This process can be described formally by the set of stochastic differential equations

$$d\mathbf{c}_j(t) = \boldsymbol{R}_r(\boldsymbol{c}_j)dt + \int_{\boldsymbol{X}} \Xi_{\mathbf{c}_j}(\boldsymbol{c}_j) n(dt, d\boldsymbol{u}), \quad j = 1,2,3\ldots \tag{2}$$

where $\boldsymbol{R}_r(\boldsymbol{c}_j)$ denotes the vector of reaction rates and the second terms on the right hand side of Eq. (2) represent random counting integrals in which $n$ is a Poisson process determining the conditions of collisions in the physical space $X$ while functions $\Xi_{(,)}$ describe the values of collisions induced short-time changes of concentrations.

Process by Eq. (2), due to interchanges of mass of species induced by collisions generates distribution of concentration over the fluid elements the resultant intensity of which depends on the local diffusional mass transfer rates during the course of collisions. Since the fluid elements are of constant volume this mass exchange is treated as a coalescence/redispersion event in which motion of the $k$th species is described by the pair of linear differential equations.

$$\frac{d}{dt}\begin{bmatrix} c_j^k(t) \\ c_l^k(t) \end{bmatrix} = \begin{bmatrix} \alpha^k & -\alpha^k \\ -\alpha^k & \alpha^k \end{bmatrix} \begin{bmatrix} c_j^k(t) \\ c_l^k(t) \end{bmatrix} \tag{3}$$

subject to the initial conditions

$$\begin{bmatrix} c_j^k(0) \\ c_l^k(0) \end{bmatrix} = \begin{bmatrix} c_{j0}^k \\ c_{l0}^k \end{bmatrix} \tag{4}$$

where $c_j^k$ and $c_l^k$ denote the concentrations of $k$th species in fluid elements, $\alpha^k = \beta^k a / \upsilon_\eta$, $\beta^k$ is a diffusion-type mass transfer coefficient, $a$ denotes the contact surface and $\upsilon_\eta$ stands for the volume of a fluid element.

The contact surface $a$ between two colliding fluid elements is, since it is a function of velocity fluctuations and depends on the actual value of velocity causing the impact of fluid elements, is a random variable while the mass transfer coefficient $\beta^k$, because of the randomness of the actual driving force of mass transfer can