MULTI-SCALE APPROACH TO POPULATION BALANCE MODELLING OF DISPERSE SYSTEMS

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Abstract: A three-scale model is presented and analysed using the multi-scale methodology of complex systems. The micro-scale model is formulated as a set of stochastic differential equations for the individual disperse objects and it is shown that the population balance equation, containing also terms describing collision interchange of extensive quantities between the disperse elements is a meso-scale model of disperse systems. The macro-scale model is formulated by means of the moments of internal quantities. As an example a two-population model, governing the coupled behaviour of crystals and fluid elements is presented for describing micromixing in solution crystallization.

1 INTRODUCTION

Disperse systems of chemical engineering, containing large numbers of individual interacting dispersed objects such as solid particles, liquid droplets or gas bubbles, or often combinations of those are nonequilibrium and (usually) nonlinear multi-phase systems. Their characteristic property is that, not depending on the nature of disperse elements, a number of interacting size and time scales can be distinguished and identified therefore these systems, in principle, belong to the class of complex multi-scale systems .

The multi-scale structure of disperse systems has been considered relating to different modelling and computational problems (Eberard et al., 2005; Ingram and Cameron, 2002; Li et al., 2004; Wei, 2007), and also in context of population balance models. (Mazzotti, 2010; Lakatos, 2010). However, the multiscale nature of the population balance models of disperse systems has not been analysed in details yet.

In this paper the population balance models of disperse systems are analysed applying the multiscale methodology of complex systems. The microscale model is formulated as a set of stochastic differential equations for the individual disperse objects with collision interactions and it is shown that the population balance equation, containing also terms describing interchange of extensive quantities between the disperse elements is a meso-scale model of disperse systems. The macro-scale model is formulated using the moments of internal quantities. As an example a two-population model, governing the coupled behaviour of crystals and fluid elements is presented for describing micromixing in reaction crystallization.

2 MICRO-SCALE MODEL

Consider a large population of interacting disperse objects, solid or fluid particles moving stochastically in a continuous carrier phase. Let us assume that 1) the extensive quantities carried by the disperse elements, such as mass of chemical species and heat are distributed homogeneously inside those, or internal motion of those is irrelevant regarding the behaviour of system; 2) the continuous phase is modelled on kinetic scale, i.e. by means of concentrations of chemical species; 3) collision interactions of disperse elements may cause their coalescence, aggregation, breakup as well as interchange of extensive quantities between the colliding elements. Therefore the micro-scale of the disperse system can be assigned by the individual disperse elements.

Assuming that x_p and u_p denote, respectively, the space coordinates and velocities along those of a disperse element, v_p denotes its volume, c_p stands for the vector of concentrations of $K \geq 0$ relevant chemical species inside the disperse elements and T_p denotes its temperature. Then the state of a disperse

element at time t is given by the vector $(x_p, u_p, v_p, c_p, T_p) \in \mathbf{R}^{K+8}$, and, introducing the simplified notation $\chi_p = (v_p, c_p, T_p)$, the micro-scale model of disperse system, completed with the model equations of the continuous phase is given by the following set of stochastic differential equations

$$dx_p(t) = u_p(t)dt \qquad (1)$$

$$m_p du_p(t) = -\alpha(u_p)dt + \sum f dt + \sigma_{u,p} dW(t) + \int_{\mathbf{X}} \Xi_{u_p}(u_p, v_p) \mathscr{N}(dx_p, du_p, d\chi_p) \qquad (2)$$

$$d\chi_p(t) = \beta(\chi_p)dt + \sigma_{\chi,p} dW(t) + \int_{\mathbf{X}} \Xi_{\chi_p}(u_p, \chi_p) \mathscr{N}(dx_p, du_p, d\chi_p) \qquad (3)$$

where $\sum f$ are deterministic forces, $W(t)$ is a multivariable Wiener process, α and β are deterministic functions, $\sigma_{u,p}$ and $\sigma_{\chi,p}$ are the diffusion matrices, function $\mathscr{N}$ determines the conditions of collisions between the disperse elements while functions $\Xi_{(.)}$ describe the velocity, volume, concentration and temperature changes induced by collisions.

The set of differential equations (1)-(3) describes the behaviour of the population of disperse elements entirely by tracking the time evolution of the state of each disperse object individually. The first terms on the right sides of Eqs (2) and (3) describe the deterministic and stochastic disperse element-continuous environment interactions, i.e. motion of the disperse elements induced by the continuous carrier as well as the mass and heat exchange between the disperse elements and continuous environment. The integral terms in Eqs (1)-(3) represent jump-like stochastic changes of the internal properties induced by collisions, i.e. jump-like changes of the velocities, volumes, concentrations and temperatures of the disperse elements.

The system of stochastic equations (1)-(3) induces a Markov process $\{x_p(t), u_p(t), \chi_p(t)\}_{t \geq 0}$ with continuous sample paths and finite jumps (Gardiner, 1983, Sobczyk, 1991). Taking it into consideration, a multidimensional population density function $(x, u, \chi, t) \rightarrow \hat{n}(x, u, \chi, t)$ is defined where the variables (x_p, u_p, χ_p) are measured on the (x, u, χ) coordinates, and $\hat{n}(x, u, \chi, t)dxdud\chi$ provides the number of disperse elements at time t in the domain $(x, u, \chi; x + dx, u + du, \chi + d\chi)$. The population density function $\hat{n}(., t)$ provides the state function of population.

Then, in analogy with the transition probability, a transition measure and, in turn, the conditional

transition measure $\hat{P}_c(s,.;t,.|.)$ can be derived (Lakatos, 2010) by means of which variation of the state function of population of disperse elements is described by the transformation

$$\hat{n}(x, u, \chi, t) = \frac{1}{N(s)} \int_{\dot{\mathbf{X}}} \int_{\dot{\mathbf{X}}} \hat{P}_c(s, x', u', \chi'; t, x, u', \chi | x'', u'', \chi'') \hat{n}(x', u', \chi', s) \times \hat{n}(x'', u'', \chi'', s) \times dx'dx''du'du''d\chi'd\chi'' \qquad (4)$$
$$t > s; (x, u, \chi) \in \dot{\mathbf{X}}$$

where

$$N(s) = \int_{\dot{\mathbf{X}}} \hat{n}(x, u, \chi, s)dxdud\chi \qquad (5)$$

denotes the number of disperse elements in the given domain at time s.

In Eq.(4), expression

$$\frac{1}{N(s)} \hat{n}(x'', u'', \chi'', s)dx''du''d\chi'' \qquad (6)$$

is interpreted as the probability that there exists a disperse element in the state domain $(x'', u'', \chi'', x''+dx'', u''+du'', \chi''+d\chi'')$ possibly interacting with a disperse element of state (x', u', χ') and the result of this interaction event is expressed by the conditional transition measure $\hat{P}_c$.

Eq.(4) is an integral equation formulation of the population balance equation of interacting disperse elements. It provides a global description of the population of disperse elements but appears to be unpractical in computations since identification of the multivariate conditional transition measure $\hat{P}_c(s,.;t,.|.)$ is a crucial problem. However, Eq.(4) is an important intermediate state in developing the integral-differential equation form of the population balance equation.

3 MESO-SCALE MODEL

Let us now define an ε-environment around the position vector x in the physical space as

$$U_\varepsilon(x) = \{y : d(x, y) < \varepsilon\} \qquad (7)$$

where $\varepsilon > 0$ and $d(x, y)$ denotes the distance of two vectors.

Assuming that

1) The ε-environment contains sufficient number of disperse elements for defining a population density function as

$$n(\boldsymbol{x},\boldsymbol{\chi},t)= \int\limits_{U_\varepsilon(\boldsymbol{x})}\int\limits_{U} \hat{n}(\boldsymbol{y},\boldsymbol{u},\boldsymbol{\chi},t)d\boldsymbol{u}\,d\boldsymbol{y} \qquad (8)$$

by means of which

$$\frac{1}{N(\boldsymbol{x},t)}n(\boldsymbol{x},\boldsymbol{\chi},t)d\boldsymbol{\chi} \qquad (9)$$

is interpreted as the probability that there exists a disperse element in the ε-environment in the domain $(\boldsymbol{\chi},\boldsymbol{\chi}+d\boldsymbol{\chi})$ where $N(\boldsymbol{x},t)$ provides the total number of these elements in the ε-environment at time t.

2) The transport of the populations of disperse elements is governed by the convection-dispersion model, then
the system is governed by the spatially distributed multi-dimensional population balance equation

$$\begin{aligned}
\frac{\partial n(\boldsymbol{x},\boldsymbol{\chi},t)}{\partial t} &= \boldsymbol{B}[n(\boldsymbol{x},\boldsymbol{\chi},t)] \\
&+ \nabla_x \boldsymbol{D}_x \nabla_x n(\boldsymbol{x},\boldsymbol{\chi},t) - \nabla_x[\boldsymbol{u}_x n(\boldsymbol{x},\boldsymbol{\chi},t)] \\
&- \nabla_{\boldsymbol{\chi}}[\boldsymbol{G}_{\boldsymbol{\chi}} n(\boldsymbol{x},\boldsymbol{\chi},t)] + \boldsymbol{M}_i^{K+2}[n(\boldsymbol{x},\hat{\boldsymbol{\chi}},t)] \\
&+ \boldsymbol{M}_b^1[n(\boldsymbol{x},\hat{\boldsymbol{\chi}},t)] + \boldsymbol{M}_a^{K+2}[n(\boldsymbol{x},\boldsymbol{\chi},t)]
\end{aligned} \qquad (10)$$

The first term on the right hand side of Eq.(10) represents the source of the disperse elements born in the continuous phase. The next two terms describe the transport of the population density function in the physical phase while the fourth term represents the rate of change of population density function due to continuous phase-disperse elements interactions

$$\nabla_{\boldsymbol{\chi}}\big[\boldsymbol{G}_{\boldsymbol{\chi}} n(\boldsymbol{x},\boldsymbol{\chi},t)\big] = \nabla_{\boldsymbol{\chi}}\bigg[\frac{d\boldsymbol{\chi}}{dt}n(\boldsymbol{x},\boldsymbol{\chi},t)\bigg] \qquad (11)$$

The next terms, in turn, represent the rates of change of the population density function because of the direct mass and/or heat exchange between the disperse elements, breakage and aggregation/coalescence of disperse elements induced by collision events.

The second integral in the expression

$$\begin{aligned}
\boldsymbol{M}_i^{K+2}&[n(v,\boldsymbol{\chi},t)] = \\
&-\frac{1}{N(t)}\int\limits_0^{c_m}\int\limits_0^{c_m}\int\limits_0^{v_m}\int\limits_0^1 \frac{\pi_{(v,\upsilon)}}{\omega p_\upsilon}\delta_{\boldsymbol{\chi}'}\bigg[\bigg(\frac{(\boldsymbol{\chi}'-\boldsymbol{\chi})}{\omega p_\upsilon}+\boldsymbol{\chi}\bigg)-\boldsymbol{\chi}''\bigg]\times \\
&\times F_{\omega|\upsilon}(d\boldsymbol{\omega})n(\boldsymbol{x},v,\boldsymbol{\chi}',t)n(\boldsymbol{x},\upsilon,\boldsymbol{\chi}'',t)d\upsilon d\boldsymbol{\chi}' d\boldsymbol{\chi}'' \\
&+\frac{1}{N(t)}\int\limits_0^{c_m}\int\limits_0^{c_m}\int\limits_0^{v_m}\int\limits_0^1 \frac{\pi_{(v,\upsilon)}}{\omega p_\upsilon}\delta_{\boldsymbol{\chi}}\bigg[\bigg(\frac{(\boldsymbol{\chi}-\boldsymbol{\chi}')}{\omega p_\upsilon}+\boldsymbol{\chi}'\bigg)-\boldsymbol{\chi}''\bigg] \\
&\times F_{\omega|\upsilon}(d\boldsymbol{\omega})n(\boldsymbol{x},v,\boldsymbol{\chi}',t)n(\boldsymbol{x},\upsilon,\boldsymbol{\chi}'',t)d\upsilon d\boldsymbol{\chi}' d\boldsymbol{\chi}''
\end{aligned} \qquad (12)$$

provides the rate of increase of the number of disperse elements having internal variables $\boldsymbol{\chi}$ due mass and heat interchange between colliding disperse

elements with volumes v and υ having internal variables χ' and χ'' while the first integral describes the rate of the number of decrease due to similar events. In Eq.(12) $\boldsymbol{\omega}$ denotes a random vector with conditional probability distribution function $F_{\omega|\upsilon}(.)$ describing the extent of equalization of intensive variables under the condition that a disperse element of volume υ is colliding with one of volume υ, $\pi_{(v,\upsilon)}$ is the frequency of collisions of such disperse elements, while $N(t)$ expresses the total number of disperse elements in the ε-environment of $\boldsymbol{x}$. In Eq.(12), the components of parameter vector $\boldsymbol{p}_\upsilon$ take the form $p_\upsilon = v/(v+\upsilon)$.

The rate of increase of the number of disperse elements because of breakage is expressed by the second integral of the term

$$\begin{aligned}
\boldsymbol{M}_b^1[n(\boldsymbol{x},v,\hat{\boldsymbol{\chi}},t)] &= -\frac{1}{N(t)}\int\limits_0^{v_m}\int\limits_0^{v_m}\phi_b(\upsilon'',v)S_b(v|\upsilon')\times \\
&\pi_{(v,\upsilon')}n(\boldsymbol{x},v,\hat{\boldsymbol{\chi}},t)n(\boldsymbol{x},\upsilon',\hat{\boldsymbol{\chi}},t)d\upsilon' d\upsilon'' \\
&+\frac{1}{N(t)}\int\limits_0^{v_m}\int\limits_v^{v_m}\phi_b(v,\upsilon')S_b(\upsilon'|\upsilon'')\pi_{(\upsilon',\upsilon'')}\times \\
&n(\boldsymbol{x},\upsilon',\hat{\boldsymbol{\chi}},t)n(\boldsymbol{x},\upsilon'',\hat{\boldsymbol{\chi}},t)d\upsilon' d\upsilon''
\end{aligned} \qquad (13)$$

where $S_b(\upsilon'|\upsilon'')$ is the probability of breakage of disperse element of volume υ' collided with one of volume υ'', and $\phi_b(v,\upsilon')$ denotes the ratio of disperse elements of volume v resulted from breakage of disperse elements of volume υ'. The first integral provides the rate of decrease due to similar events. Here it was assumed that the effect of a breakage event on the extensive quantities carried by the disperse elements is negligible. Note that writing Eq. (13) the notation $\boldsymbol{\chi} = (v,\hat{\boldsymbol{\chi}})$ was used.

When the breakage of disperse elements occurs because of their collisions with some solid surface or, as in the case of fluid droplets and bubbles induced by turbulence then Eq.(13) can be written in the form

$$\begin{aligned}
\boldsymbol{M}_b^1[n(\boldsymbol{x},v,\hat{\boldsymbol{\chi}},t)] &= -\int\limits_\Theta\int\limits_0^v \phi_b(\upsilon'',v)S_b(v|\theta)\pi_{(v,\theta)}\times \\
&n(\boldsymbol{x},v,\hat{\boldsymbol{\chi}},t)F_\theta(d\theta)d\upsilon''+\int\limits_\Theta\int\limits_v^{v_m}\phi_b(v,\upsilon')S_b(\upsilon'|\theta)\times \\
&\pi_{(\upsilon',\theta)}n(\boldsymbol{x},\upsilon',\hat{\boldsymbol{\chi}},t)F_\theta(d\theta)d\upsilon'
\end{aligned} \qquad (14)$$

where θ is a random variable with the probability distribution function $F_\theta(.)$ characterizing the frequency of random events inducing breakage of the disperse elements.

In the case of aggregation or coalescence, the rate of increase of the number of disperse elements is given by the second integral of the term

$$
\begin{aligned}
&\boldsymbol{M}_a^{K+2}\big[n(\boldsymbol{x},v,\hat{\boldsymbol{\chi}},t)\big]= \\
&-\int_0^{c_m}\!\!\int_0^{c_m}\!\!\int_0^{v_m}\frac{S_a(v,\upsilon)}{\boldsymbol{p}_\upsilon}\pi_{(v,\upsilon)}\delta_{\boldsymbol{\chi}'}\!\left[\!\left(\frac{(\boldsymbol{\chi}'-\boldsymbol{\chi})}{\boldsymbol{p}_\upsilon}+\boldsymbol{\chi}\right)-\boldsymbol{\chi}''\right]\times \\
&n(\boldsymbol{x},v,\hat{\boldsymbol{\chi}},t)n(\boldsymbol{x},\upsilon,\hat{\boldsymbol{\chi}}',t)d\upsilon d\hat{\boldsymbol{\chi}}' \\
&+\int_0^{c_m}\!\!\int_0^{c_m}\!\!\int_0^{v}\frac{S_a(v-\upsilon,\upsilon)}{\boldsymbol{p}_\upsilon}\delta_{\boldsymbol{\chi}}\!\left[\!\left(\frac{(\boldsymbol{\chi}-\boldsymbol{\chi}')}{\boldsymbol{p}_\upsilon}+\boldsymbol{\chi}'\right)-\boldsymbol{\chi}''\right]\times \\
&\pi_{(v-\upsilon,\upsilon)}n(\boldsymbol{x},v-\upsilon,\hat{\boldsymbol{\chi}}',t)n(\boldsymbol{x},\upsilon,\hat{\boldsymbol{\chi}}'',t)d\upsilon
\end{aligned}
\tag{15}
$$

where $S_a(v-\upsilon,\upsilon)$ denotes the probability of agglomeration or coalescence of the colliding elements having volumes v-υ and υ. Here it is assumed that agglomeration or coalescence of two disperse elements leads to full equalization of their intensive quantities therefore $\boldsymbol{\omega}=\mathbf{1}$.

4 MACRO-SCALE MODEL

A mathematical model of a disperse system, not depending on the method of development must satisfy the requirements of the first principle models, i.e. the balances of conservative extensive quantities have to be fulfilled as concerns the whole system. From one side this requirement provides strong limitations on the forms of the constitutive expressions of Eqs (10)-(15). From the other side, the population density function playing a centred role in the meso-scale model (10)-(15) is not an extensive quantity by itself. Extensive quantities for the population of disperse elements needed for the balances are formulated only by the multivariate joint moments of internal variables defined as

$$
\mu_{l,k_1\dots k_K,m}(\boldsymbol{x},t)=\int_{\boldsymbol{\chi}} \upsilon^l c_1^{k_1}\dots c_K^{k_K} T^m n(\boldsymbol{x},\boldsymbol{\chi},t)d\boldsymbol{\chi}
\tag{16}
$$

As a consequence, for instance, the total mass involved in the population of disperse elements is given as

$$
\langle m\rangle(\boldsymbol{x},t)=\rho\langle v\rangle(\boldsymbol{x},t)
\tag{17}
$$

where $\langle v\rangle(\boldsymbol{x},t)$ denotes the mean volume of the population expressed as

$$
\langle v\rangle(\boldsymbol{x},t)=\frac{1}{N(t)}\int_{\boldsymbol{\chi}} \upsilon n(\boldsymbol{x},\boldsymbol{\chi},t)d\boldsymbol{\chi}
\tag{18}
$$

while ρ is the density of the disperse elements.

Similarly, the total heat involved in the population is expressed as

$$
C\rho\langle T\rangle(\boldsymbol{x},t)=\frac{C\rho}{N(t)}\int_{\boldsymbol{\chi}} Tn(\boldsymbol{x},\boldsymbol{\chi},t)d\boldsymbol{\chi}
\tag{19}
$$

while C is the heat capacity of the disperse elements.

Finally, the mean value of kth species is given as

$$
\langle c_k\rangle(\boldsymbol{x},t)=\frac{1}{N(\boldsymbol{x},t)}\int_{\boldsymbol{\chi}} c_k n(\boldsymbol{x},\boldsymbol{\chi},t)d\boldsymbol{\chi}
\tag{20}
$$

As an example the heat balance from Eq.(10) takes the form

$$
\begin{aligned}
&\frac{\partial Q(\boldsymbol{x},t)}{\partial t}=\nabla_x \boldsymbol{D}_x\nabla_x Q(\boldsymbol{x},t)-\nabla_x\big[\boldsymbol{u}_x Q(\boldsymbol{x},t)\big] \\
&C\rho\int_{\boldsymbol{\chi}}\frac{dT}{dt}n(\boldsymbol{x},\boldsymbol{\chi},t)d\boldsymbol{\chi}+C\rho\int_{\boldsymbol{\chi}} T\boldsymbol{B}\big[n(\boldsymbol{x},\boldsymbol{\chi},t)\big]d\boldsymbol{\chi}
\end{aligned}
\tag{21}
$$

Where

$$
Q(\boldsymbol{x},t)=N(t)C\rho\langle T\rangle(\boldsymbol{x},t)
\tag{22}
$$

The third term on the right hand side represents the population-continuous phase interaction, while the last term describes the heat effect of source of disperse elements.

5 TWO-POPULATION MODEL

The multidimensional population balance equation (10)-(15) is cognitive-type model of populations of disperse elements. When modelling real processes appropriate reductions of this cognitive-type model, the so called purpose-oriented models are applied. These models contain necessary and sufficient information to provide an adequate description of the process to be modelled. Here as an example a two-population model is presented aimed to describe micromixing in reaction crystallization.

Crystallization from solution is an important fluid-solid disperse system in which the disperse phase is formed by solid particles. Let us assume that the crystallizer is isothermal and the supersatura-tion is generated by the chemical reac-tion $A+B\rightarrow C\downarrow$. When the continuous phase can be treated in the ε-environment of $\boldsymbol{x}$ as a homogeneous continuum with respect to scalar quantities then the composition and temperature environment is the same for all crystals. Consequently, crystals born and growth in the same composition and temperature environment hence the behaviour of crystals can be modelled adequately by using the population balance equation (10). This crystallizer is considered perfectly mixed on micro-scale.

When, however, solution exhibits varying in time spatial inhomogeneites of scalars even on micro-scale then these changes modulate not only the pro-

cesses in solution (mixing, reaction, nucleation) but also the crystal-solution and crystal-crystal interactitions since crystals moving randomly in the fluid phase meet diverse composition and temperature regions even inside the ε-environment of x. This phenomenon can be modelled making use of the generalized coalescence/re-dispersion (gCR) model developed in the context of multi-scale structure of (Lakatos, 2008; Lakatos et al., 2011).

In the gCR model, the Kolmogorov-scale eddies of solution are treated as a large population of fluid elements having identical volume v_η. Then, two different interacting populations are identified in the crystallizer, i.e. the population of crystals and that of fluid elements. Since, however, kinetic processes are also determined by micro-scale phenomena, the rate expressions of crystallization kinetics may be influenced significantly by stochastic interactions of these two populations. Therefore, the mathematical model of the crystallizer consists of two population balance equations and of averaged kinetic and constitutive equations describing their interactions.

Since the concentration of species in crystals is negligible then the population density function of crystals is given as

$$n(x,v,t)dv = \int_{X} \delta(c)\delta(T - \langle T \rangle)n(x,\chi,t)d\chi \quad (23)$$

while that of fluid elements is defined as

$$p(x,c,t)dc = \int_{X} \delta(v - v_\eta)\delta(T - \langle T \rangle)n(x,\chi,t)d\chi \quad (24)$$

where $\langle T \rangle$ denotes the temperature of process.

Assuming that 1) the motion of populations in the vessel is described by the 1D axial dispersion model, 2) no breakage of crystals occurs then the population balance equation for crystals is given as

$$\frac{\partial n(\xi,v,\tau)}{\partial \tau} + \bar{t}\frac{\partial[\langle G \rangle(\xi,v,\tau)n(\xi,v,\tau)]}{\partial v}$$
$$= \frac{1}{Pe}\frac{\partial^2 n(\xi,v,\tau)}{\partial \xi^2} - \frac{\partial n(\xi,v,\tau)}{\partial \xi} \quad (25)$$
$$+ \bar{t}\langle B_n \rangle(\xi,\tau)\delta(v - v_n) + \bar{t}\langle B_a \rangle(\xi,\tau)$$

while that for fluid elements takes the form

$$\frac{\partial p(\xi,c,\tau)}{\partial \tau} + \frac{\partial}{\partial c}\left[\frac{dc}{dt}p(\xi,c,\tau)\right]$$
$$- \bar{t}\frac{\partial[\langle R \rangle(\xi,\tau)p(\xi,c,\tau)]}{\partial c_c} = \bar{t}M_i^3[p(\xi,c,\tau)] \quad (26)$$
$$+ \frac{1}{Pe}\frac{\partial^2 p(\xi,c,\tau)}{\partial \xi^2} - \frac{\partial p(\xi,c,\tau)}{\partial \xi}$$

where $\xi = x/L$ is the dimensionless axial coordi-

nate scaled with the length of vessel L while $\tau = t/\bar{t}$ denotes the dimensionless time scaled with the mean residence time $\bar{t}$. In Eqs (25)-(26), $\langle R \rangle$ denotes the mean rate of consumption of solute in solution due to nucleation and growth of crystals, $\langle G \rangle$, $\langle B_n \rangle$ and $\langle B_a \rangle$ denote, respectively, the rates of crystal growth, nucleation and agglomeration, all averaged over the ensemble of fluid elements in the ε_ξ-environment of ξ. These kinetic equations take the forms as follows.

$$\langle G \rangle = \frac{1}{N_p}\int_0^{c_m} G(c_c,c_e,v)p(\xi,c,\tau)dc \quad (27)$$

where c_c and c_e denote the solute and the equilibrium saturation concentrations,

$$\langle B_n \rangle = \frac{1}{N_p N_n}\int_0^\infty \int_0^{c_m} B_n(c_c,c_e)\times$$
$$\times p(\xi,c,\tau)n(\xi,v,\tau)dcdv \quad (28)$$

where N_p and N_n stand for the total numbers of fluid elements and crystals in the ε_ξ-environment of ξ.

The rate of agglomeration is given by a simplified form of Eq.(15) since the system is assumed to be isothermal, and there is no mass exchange between the crystals:

$$\langle B_a \rangle = -\frac{1}{N_p}\int_0^{c_m}\int_0^\infty S_a(c_c,c_e)\times$$
$$\times \pi_{(v,v')}n(\xi,v,\tau)n(\xi,v',\tau)p(\xi,c,\tau)dv'dc$$
$$+ \frac{1}{2N_p}\int_0^{c_m}\int_0^\infty S_a(c_c,c_e)\pi_{(v-v',v')}\times \quad (29)$$
$$\times n(\xi,v-v',\tau)n(\xi,v',\tau)p(\xi,c,\tau)dv'dc$$

In general case, the model equations of the two-population model (25)-(29) can be solved only by numerical method, but when the exponents b and g are positive integers and the agglomeration kernel provides closed moment terms then, since the moment terms of micromixing operation are always closed (Lakatos, 2011), a closed finite set of moment equations can be obtained. Indeed, assuming the following forms for the intrinsic kinetic rates $S_a \equiv 1$, $B_b = k_b(c_c - c_e)^3 \mu_1$, $\pi_{(v,v')} = b_0(v + v')$ and applying the cumulant-neglect closure (Lakatos, 2010) a set of 13 ODE's was generated for the joint moments of concentrations c_a, c_b and c_c and for the raw moments of crystal volume v up to the second order.

Figure 1 presents the time evolution of axial distribution of the number of crystals for the case of small seeding and micromixing intensity 0.15. In this case the feed of species A and B was segregated. Figure 1 shows well the effects of increasing nuclea-
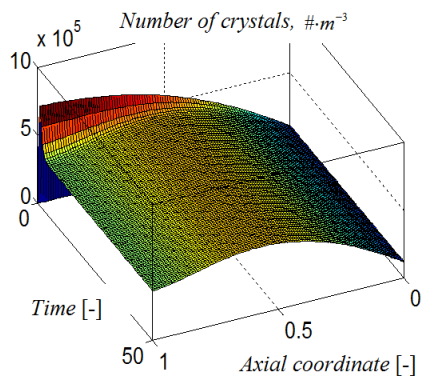
Figure 1: Time evolution of the axial distribution of population density function of crystals for *Pe*=10 and $\bar{t}=1$.

tion rate as the state of mixing was increased along the axial coordinate. At the same time, the number of crystals is decreased due to crystal agglomeration.

Figure 2 shows the effects of micromixing intensity on the steady state axial distribution of crystals illustrating the strong influence of micromixing on the number of crystals produced in the process.
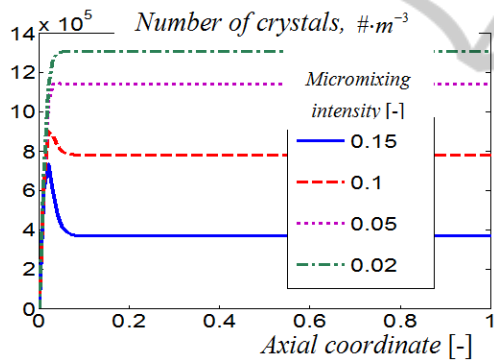

Figure 2: Effects of micromixing intensity on the steady state axial distribution of population density function of crystals for *Pe*=10 and $\bar{t}=1$.

6 CONCLUSIONS

The population balance approach was applied to develop a three-scale model for disperse systems using the multi-scale methodology of complex systems. It was shown that marking out the individual disperse objects for microlevel of the system the population balance equation, containing also terms describing collision interchange of extensive quantities between the disperse elements and motion in the physical space is a meso-scale model. In this interpretation, the macro-scale model is formulated by means of the moments of internal quantities of disperse elements. As an example, a two-population model, governing

the coupled behaviour of crystal and fluid element populations is presented for describing micromixing in reaction crystallization.

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