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Towards multiscale modelling in product engineering

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ABSTRACT

A concept of multiscale modelling of product manufacturing based on integration of three modelling methods currently applied at different scales of length and time: process system modelling, computational fluid dynamics and computational chemistry was presented. Major features of the three key types of modelling in the chemical and process industries were briefly described. The first applications and mutual benefits of joint use of two of the three approaches were presented along with the perspectives for the full integration of all three methods. The crucial role of a universal interface, such as the CAPE-OPEN standard, was emphasized.

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1. Introduction

Progress in modelling techniques for the chemical and allied industries is closely related to the general development of chemical engineering science. Since the beginning of the last century the first paradigm associated with unit processes and operations was adopted. Around the 1960s the second paradigm was accepted (Bird, Stewart, & Lightfood, 1960) and it was focused on process engineering of momentum, heat and mass transport. Due to significant advancement in the computer and information technology, roughly from the early 1980s dynamic development of process simulation begun. The development was also reflected in applications to chemical equipment on different scales, ranging from micrometer to full industrial plant. The new research tools rendered it possible to carry out numerical simulations of non-classical fluid mechanics problems such as turbulent or/and multiphase or/and non-Newtonian flows accompanied by chemical reactions. Table 1 presents characteristic features of design calculations for each paradigm.

Nonetheless, since about 2000 problems typical for chemical product engineering have been largely investigated (Cussler & Moggridge, 2001). An important modification in the design methods was then introduced. While the traditional chemical engineering concentrates its tools on the conversion path from raw materials to products, product engineering starts from a product of market-defined properties and explores methods and tools to effectively resolve the problem (Costa, Moggridge, & Saraiva, 2006; Hill, 2009). The investigations can be carried out at various levels of resolution and often involve process analyses on different scales of time and length (Vlachos, Mhadeshwar, & Kaisare, 2006), and therefore product engineering is closely related to the multiscale approach (Charpentier, 2009; Costa et al., 2006; Hill, 2009). Currently, customers decide on the main features of market products and the essential product properties would be properly considered only then when defined and designed at the nano- or microscale. The multiscale modelling supports such design methods by starting from the molecular level and ending up at the technical scale. Chemical product design aims at novel methods to manufacture new products (Costa et al., 2006; Gani, 2004a), and the proposed multiscale modelling plays an ever-growing role in it.

The required features of a new market product usually indicate the initial scale of consideration in the procedure of product design. Increasingly since year 2000, investigations have been carried on product creation at the level of molecular structure (molecular design) (Gani, 2004a, 2004b) or on reaction modelling in nanoscale (Tyczkowski, 2007). Significant progress of molecular engineering can be noticed and product engineering is nowadays often identified with that approach (Gani, 2004a).

The designing approach proposed in product engineering is often regarded as the third paradigm of chemical and process engineering. The method is continuously progressing and still in the stage of dynamic development, idea creation and approval (Charpentier, 2002; Charpentier & McKenna, 2004; Gani, 2004b; Voncken, Broekhuis, Heeres, & Jonker, 2004) while the first and second paradigms seem already completed (see Table 1). It can also be envisaged that the design of chemical and biochemical equipment

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Nomenclature

a	acceleration (m/s^2)
c_p	specific heat (J/kg K)
c_α	molar concentration of "α" component (kmol/m^3)
D_α	kinematic diffusivity (m^2/s)
F	external force on object (N)
$\hat{H}$	Hamiltonian operator
$\hbar$	reduced Planck's constant = $h/(2\pi)$ (J s)
i	imaginary unit
m	mass of object (kg)
S_ϕ	source term
T	temperature (K)
t	time (s)
v_i, v_j	velocity components (m/s)
x_i, x_j	Cartesian coordinates (m)
ϕ	generalized variable
Γ_{eff}	effective diffusivity
λ	molecular thermal conductivity (W/(mK))
μ	molecular viscosity (kg/(ms))
ρ	density (kg/m^3)
ψ	wave function

in the coming years will be coupled with the nanoscale approach to model (bio-)chemical processes on ever-growing scale.

Expected properties of the designed chemical/biochemical product established in macroscale can nowadays be precisely predicted by means of the molecular modelling. According to Charpentier (2009), the key factor in succeeding in the final product of the desired features is the quality control at the level of nano- and/or microstructure formation.

It is widely accepted that modelling carried out at the scale of a whole production plant, together with the process simulation of selected equipment units, can lead to cost optimization of the product manufacturing. Control of the chemical and physical processes on the small-scale facilitates handling of the processes on larger scale (Yuan, 2007), which is regarded by Wei (2007) as the key role in general considerations of the problem. This paper presents an extension and updating of the previous study for chemical reactors (Jaworski & Zakrzewska, 2008).

2. Modelling methods in chemical and process industries

The modelling methods used in the prediction of phenomena and processes that are employed in the chemical and allied industries are briefly described in this chapter. Subsequently, the published application scope of pairs of those methods is presented in chapter 3.

2.1. Process system modelling (PSM)

Process system engineering is an independent scientific discipline that covers process design, process operations, process control, process intensification, chemical product design and supporting them modelling tools (Klatt & Marquardt, 2009; Grossmann and Westerberg, 2000). The modelling procedure is usually carried out in seven steps (Hangos & Cameron, 2001) consisting in: problem definition, identifying the controlling factors or mechanisms, evaluation the data for the problem, developing a set of model equations, finding and implementing a solution procedure, verifying the model solution and model validation. In a similar manner, Foss, Lohmann, and Marquardt (1998) presented the modelling process related to chemical industries. In this section, mathematical models and process simulation tools are only shortly illustrated. The tools can be employed to modelling both a single equipment unit and a whole technological system as well.

In the steady-state processes, the mathematical model of the process unit is composed by a set of algebraic equations of Eq. (1) type, which may result from transformation of the ordinary differential equations and elimination of all derivative terms dx_i/dt by setting them to zero (Hangos & Cameron, 2001). The number and form of those equations depends on the precision of the model (Jeżowski, 2002). Typical equations of this kind are those for: mass and energy balance, interfacial equilibrium, equipment hydraulics, thermodynamic properties and kinetics of chemical reactions and of transfer processes.

$$f(\boldsymbol{x}, \boldsymbol{u}) = 0 \tag{1}$$

On the other hand, dynamic simulations predict time evolution of the model outputs for imposed inputs, parameters and structure of the model. The mathematical model of the process unit is consisted of the material and energy balances (differential equations) with their associated boundary and initial conditions, and constitutive equations (algebraic equations). This results in a system of differential and algebraic equations represented by Eq. (2) (Hangos & Cameron, 2001; von Wedel, Marquardt, & Gani, 2002).

$$f\left(\frac{d\boldsymbol{x}}{dt}, \frac{d\boldsymbol{u}}{dt}, \boldsymbol{x}, \boldsymbol{u}, t\right) = 0 \tag{2}$$

In the case of optimization calculations, the mathematical model of the process system is to be supplemented with an equation defining the objective function (Marquardt, 1996; Wozny, Li, & Wendt, 2001). The model should also include inequality constrains, which define (i) process limitations such as temperature limits or product purity, Eq. (3), and (ii) limits referring to the vectors of state (dependent), $\boldsymbol{x}$, and control (independent), $\boldsymbol{u}$, variables, Eq. (4), and (iii) initial values for iterations (Wozny et al., 2001).

$$h(\boldsymbol{x}, \boldsymbol{u}) \geq 0 \tag{3}$$

Table 1
Historical development in chemical equipment design up to year 2000 (Jaworski & Zakrzewska, 2008).

Paradigm name	Unit operations	Processes of momentum, heat and mass transfer	
Publishing year	~1900	~1960	
Paradigm no.	1	2A	2B (since ~1980)
Tools of analysis	Similarity theory, π theorem	Differential balances, turbulence theory	Computational modelling
Major emphasis in design	Integration of unit operations	Process optimization, large plants	Multiscale simulations of transport processes
Design criterion	Non-dimensional numbers, scale-up	Simplified 1D, 2D models, residence time distribution	3(4)D models, local analysis
Scale of scrutiny	~10^0 m, whole apparatus (black box)	~10^{-1} m, approximate distributions	~10^{-2} m 3(4)D distributions
Typical production	Substitutes for natural products, small-medium scale	Commodity chemicals over 1000 tonne/year	Fine chemicals below 100 tonne/year
Operation mode	Batch, semi-batch	Continuous, semi-batch	Batch, semi-batch

Table 2
Alternatives of the transport equation (Jaworski, 2005).

Differential balance of	Equation type	ϕ	Γ	Sources and sinks
Total mass	Continuity	1	0	–
Force (momentum) in jth direction	Momentum transport	v_j	μ	External and body forces
Mass of "α" component	Mass transport	c_α	ρD_α	Chemical reaction
Energy	Energy (heat) transport	T	λ/c_p	Energy conversion

$$\boldsymbol{x}_{\min} \le \boldsymbol{x} \le \boldsymbol{x}_{\max}; \quad \boldsymbol{u}_{\min} \le \boldsymbol{u} \le \boldsymbol{u}_{\max} \tag{4}$$

Selection of the appropriate modelling environment always occurs at the initial stage of the modelling process and depends on the application. With the standard steady-state problems, flowsheeting tools based on the modular-oriented strategy are usually employed. In contrast, equation-oriented simulators are usually employed to nonstandard steady-state or dynamic problems (Foss et al., 1998). Other approaches to simulating a process system, which are based on the process knowledge based environments (PKBE) and the available simulation tools were thoroughly presented elsewhere (Cameron & Ingram, 2008; Hangos & Cameron, 2001; Marquardt, 1996).

In the modular-oriented strategy, the process system is divided into modules (blocks), which correspond to individual process units or parts of them. The modules are described by separate mathematical models. Flow of information occurs between the modules. In the simulation packages of that type, a separate segment organizing the sequence of modular calculations and another segment of thermodynamic calculation can be distinguished (Marquardt, 1996). Numerical codes based on that principle are called flowsheeting programmes. They are usually applied to compute steady-state processes, however the modern programmes can also deal with transient states (Kulikov et al., 2005) or with optimization by means of economic criteria (Zhang, Pike, & Hertwig, 1995). The simulation results for process systems can be formally regarded as extensions of the computations for single units or processes. In order to carry them out the system structure has to be established, i.e. the computing order and links between the units should be known.

Uncomplicated programming of modules and their maintenance along with easy convergence, both at the unit and flowsheet levels can be regarded as the main advantages of the modular strategy. On the other hand, its drawback lies in a rigid direction of unit computing, usually "outputs from inputs" and in difficulty in dealing with specifications for unit performance.

In the second, equation-oriented approach, the whole process system is described by the same set of equations, however it is resolved simultaneously for all state variables. The more complex the process system, the more interrelated is the equation set, often non-linear, which can introduce additional hurdle in finding its solution. The equation-oriented approach is usually applied to simulating dynamic systems and results in resolving sets of differential and algebraic equations (DAE) (Pantelides & Barton, 1993). The main benefit of that approach is the solver flexibility against the problem specifications, which may be composed of either process inputs or outputs or unit performance.

The process system modelling tools are also used in modelling a single apparatus but due to considerable simplifications the solutions delivered are less accurate than those from computational fluid dynamics (CFD). The commercial process simulator often include databases for equipment models of various precision, starting from simple short-cut models to more complex ones. In order to enhance the quality of process simulations, modifications based on empirical correlations are often introduced. However, this usually leads to problems with scaling-up of the simulations results (Kramer, Bermingham, & van Rosmalen, 1999). It should be emphasized that the standard approach in designing the systems of chemical technology consists of the consecutive steps: synthesis of system structures, structure analysis and finally optimization of one or a few structures.

Several authors refer to the process system engineering (PSE) as computer-aided process engineering (CAPE). However, current understanding of CAPE covers usually a wider area, namely all modelling methods applied to solving various problems of process engineering, ranging from the whole plant down to the nanoscale. This became especially obvious at the ESCAPE-19 conference, (e.g. Thullie, Kurowski, & Chmiel-Kurowska, 2009), even when referring to CFD as CAPE (Kurowski, Chmiel-Kurowska, & Thullie, 2009). Recently Klatt and Marquardt (2009) proposed to clearly distinguish between PSE and CAPE by using the former to analyse process systems by means of systems engineering principles and tools, especially in terms of chemical engineering. The modelling tools presented here as the process system modelling (PSM) that supports the process system engineering are further used in this study to denote modelling methods and simulations tools for large scale process systems.

2.2. Computational fluid dynamics (CFD)

The analysis method of computational fluid dynamics (CFD) renders it possible to find numerical solutions to a set of partial differential balance equations for transport processes in fluids and in some cases also in solids. In general terms, that computational method delivers an approximate numerical solution of the transport equations in selected points of space and time instead of an analytical solution, which is rather impossible to achieve. The transport equation for a generalized intensive variable, ϕ, can be presented in the form of Eq. (5) where its four components represent accumulation, convection, diffusion and source, respectively.

$$\frac{\partial(\rho\phi)}{\partial t} + \frac{\partial}{\partial x_i}(\rho v_i \phi) - \frac{\partial}{\partial x_i}\left(\Gamma_{eff}\frac{\partial \phi}{\partial x_i}\right) - S_\phi = 0 \tag{5}$$

The meaning of the generalized variable and its corresponding effective transport coefficient, Γ_{eff}, should be selected from Table 2 according to the kind of extensive variable considered.

Depending on the computed information details for turbulent flows, three general simulation techniques can be distinguished, namely the Reynolds averaging Navier–Stokes (RANS), large-eddy simulation (LES) and direct numerical simulation (DNS). Among them, the simplest and also most common is the RANS approach. The most popular types of transformation of Eqs. (5) by discretization are the finite volume (FV) and finite element (FE) methods.

Fundamental independence of the CFD analysis of the size of considered equipment is generally recognized as the main advantage provided the sub-models of the individual physical and chemical processes are correct. Other advantageous features of the CFD analysis are: (i) significant shortening of time required to the R&D work on new technologies, (ii) higher efficiency of financial investments on the simulations than on experiments, (iii) availability of technical information both on the local and global scale of the equipment, (iv) possibility of simulating conditions, which are difficult or impossible to accomplish in real experiment or production plant.

Shortcomings of the CFD methods consist in high initial investment both in training of the personnel and in license fees and also

in requirements of modelling validation for complex phenomena of which our theoretical knowledge is incomplete. The CFD analysis of transport phenomena basically assumes the continuity of matter and deals with problems on the scale significantly higher than molecular. However, bearing in mind that the size of microreactors can be of the order of 0.1 mm (Hessel, Hardt, & Löwe, 2004) then the size ratio of currently considered processes, from the smallest to the biggest equipment, can be approximately as big as $1{:}10^6$.

Recent trends in the development of the CFD methods are focused on the fully predictive, i.e. without experimental data, assessment of equipment unit performance in dependence on the unit geometry and process parameters. Significant achievements were reached in that area and it is nowadays generally accepted that the laminar and single-phase processes can be simulated in the fully predictive mode with high precision. Detailed sub-models for rheologically complex and multiphase transport have been developed for the last 20 years and several non-Newtonian and interfacial problems can now be numerically predicted with decent accuracy (Jaworski, 2005). There are also reasonable opinions that CFD is not worth applying to every single case especially when involving high computing resources. It is linked to the issue of rationality and optimum criterion applied in the design of chemical plants.

A relatively new group of CFD methods is based on the lattice-Boltzmann (LB) model (Succi, 2001; Sukop & Thorne, 2006). The fundamental equation of the model stems from the kinetic theory of gases and constitutes the basic law employed in the stochastic fluid mechanics (Heinz, 2003). That approach is especially valuable when the key assumption of the classical fluid mechanics about the continuity of the fluid matter cannot be used. The basic advantage of LB is a very high numerical efficiency of parallel processing and the main drawback is currently the failure of modelling the energy transport and multiphase flows of fluids differing in their viscosity. However, one can expect that owing to further theoretical work those limitations will be overcome.

2.3. Computational chemistry (CCH)

The term "Computational Chemistry" (CCH), which represents the third method of numerical analysis, signifies several simulation methods based on the quantum and/or statistical mechanics (Young, 2001). CCH can be used in two major ways to predict: (i) the molecule geometry and its chemical properties and/or (ii) thermodynamic macro-properties of pure species and mixtures.

Quantum mechanics (QM) allows mathematically describing electrons based on the Schrödinger equation, Eq. (6), which is the momentum equation for determining changes in time of the states of the quantum system of the total energy, E (Lucas, 2007).

$$\hat{H}\psi = E\psi \tag{6}$$

Provided both the Hamiltonian operator, $\hat{H}$, and also the wave function values are known for electrons at the starting time then it should be possible to derive the wave function value, $\psi(t)$, at any future time. However, except for the hydrogen atom, a strict solution of the Schrödinger equation (6) is not known. Thus all the practical problems of quantum chemistry are solved using approximate methods and usually either the Born–Oppenheimer approximation or the Hellman–Feynman theorem is applied in popular software. One of the solution methods is the quantum Monte Carlo, which employs numerical wave functions.

However, the most popular QM calculations are based on the Hartree–Fock approximation, which enables transforming the many-electron Schrödinger equation into several single-electron equations that are later corrected for electron interactions. The QM solvers make also use of either the perturbation theory or variational methods (McWeeny, 1973). The second type of simplifications employs semiempirical molecular parameters, especially for organic compounds. Molecular modelling/mechanics (MM) can be used in the case of large molecules assuming simple algebraic energy equations for each compound (Field, 2007). The equation set is called a force field and with this method electronic and non-equilibrium processes cannot be modelled. Another kind of simplifications in QM is related to the density functional theory that assumes the molecule energy can be derived from the electron density instead of the wave function.

Statistical (molecular) mechanics (STM) provides means of deriving thermodynamic properties of a macroscopic material. It is based on potential methods and deals with numerous sets of complete molecules and thus covers a higher range of scrutiny than that of QM (Widom, 2002). Information on molecular energy levels and conformations can be derived from molecular dynamics (MD) (Dzwinel, Yuen, & Boryczko, 2006; Gani, 2004a, 2004b; Young, 2001) or Monte Carlo (MC) simulations (Frenkel & Smit, 2002; Young, 2001). In molecular dynamics simulations for a selected system, the Newton's equations of their vibrational and Brownian motion are being solved for of a set of particles till the moment of reaching the system equilibrium. General form of the Newton's dynamics law for a molecule of mass, m, is depicted in Eq. (7)

$$F = ma \tag{7}$$

and the resultant force, F, between non-bonded and non-charged fluid molecules separated by distance, r, is usually derived by means of the Lennard-Jones energy potential, V^{LJ}, e.g. in the truncated and shifted form (Liu, Chen, Nie, & Robbins, 2007).

$$V^{LJ}(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6} - \left(\frac{\sigma}{r_c}\right)^{12} + \left(\frac{\sigma}{r_c}\right)^{6}\right] \tag{8}$$

In Eq. (8), ε is the characteristic energy, σ stands for the molecule diameter and the truncation point is set at $r_c = 2.2\sigma$ to reduce computations.

Various algorithms described in (Frenkel & Smit, 2002) are used in STM and they provide information on the location, orientation and often geometry of the molecule or collection of molecules. For a given molecular system, MC methods require in iterations less computing time than molecular dynamics. However, they also have some limitations, e.g. time-dependent information such as diffusion coefficients or viscosity could not be directly obtained. The total energy of the molecule system should reproduce the Boltzmann distribution.

Mesoscale methods were also developed for modelling materials of the structure with length scale of hundreds of nanometers – close to that of CFD. The materials are modelled as a collection of units, called beads, such as molecule, micelle or microcrystalline domain. The basic concept of the mesoscale approach is Brownian dynamics, where equations of motion are modified by random and viscous forces. Other modelling techniques based on the dissipative particle dynamics (DPD) (Groot & Warren, 1997) and dynamic mean-field density functional theory (MesoDyn) (Altevogt, Evers, Fraaije, Maurits, & van Vlimmeren, 1999; Fraaije et al., 1997) are also used to predict the mesoscopic structure and morphology of the material at nanoscale level. Input data in that approach conveys information from atomistic simulation: the bead length and structure, the Flory–Huggins parameter, χ and the bead self-diffusion coefficient (Fermeglia & Pricl, 2007, 2009). In addition, for determining the properties of multiphase materials the finite element based approach (FE) (Gusev, 2001) is used. Those computations are based on information for material morphology such as inclusions or fibres in analysed material, which were obtained from the Monte Carlo procedure or DPD, MesoDyn techniques (Gusev, 1997). Commercial packages for that modelling use unstructured tetrahedral grids, which are very similar to those used in CFD.

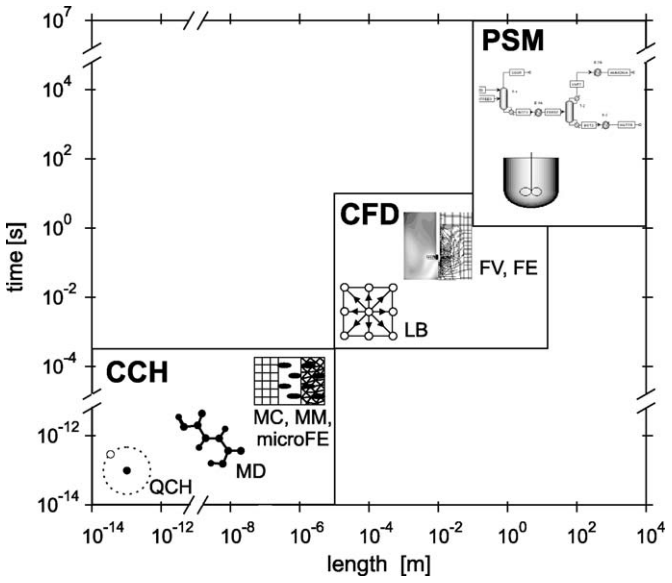

Fig. 1. Computational tools in multiscale modelling. PSM – process system modelling, CFD – computational fluid dynamics, CCH – computational chemistry, FV – finite volume, FE – finite element, LB – lattice-Boltzmann approach, MC – Monte Carlo, MM – mesoscale modelling, microFE – micro-finite element, MD – molecular dynamics, QCH – quantum chemistry.

3. Integration of modelling methods

High market requirements for new products of the process industry will promote integration of the numerical tools of fluid mechanics with the modelling tools of computational chemistry from the side of small scale (Charpentier, 2002; Dzwinel et al., 2006) and also with the dynamic process simulation (Bezzo, Macchietto, & Pantelides, 2000) on the scale higher than that typical for CFD. Fig. 1 shows the application ranges of the described three modelling method of product manufacturing. The application ranges of the PSM and CFD methods partly overlap owing to scaling down of the modelled equipment in PSM and to progressing integration of the two numerical tools. However, at present the CFD and CCH application scales do not have a common range.

The three modelling techniques, i.e. PSM, CFD and CCH, were developing independently up to roughly year 2000. During last few years the first studies on integration of the CFD and PSM methods were published and the methods have been continuously developed and improved. Fig. 1 shows schematically the ranges of both length and time characteristic for individual means of numerical analysis in process engineering. However, generally approved criteria and tools of their joint application have not yet been elaborated adequately. Along with the progress in molecular engineering, new reports on numerical modelling in nanoscale were published. Nevertheless, a small gap in the integration of CCH and CFD still exists (Dzwinel et al., 2006) and the authors of this study express the opinion that the lattice-Boltzmann (LB) approach seems the best candidate to link CFD with CCH. In addition, another potential option is the integration of the mesoscale techniques with microFE, which could provide information on properties of pure components or complex material with CFD approach.

Along with the progress in molecular engineering, new reports on numerical modelling in nanoscale were published. The two boundary modelling methods, i.e. PSM and CCH, deliver and use different types of information, which are complementary to different extent with that required for use in CFD simulations or resulted from them. A brief list of typical features of the three modelling tools is presented in Table 3.

The versatility of information from the three methods was the most likely reason, which gradually led several researchers (Braatz, Alkire, Rusli, & Drews, 2004; Fermeglia & Pricl, 2007; Vlachos et al., 2006) to the concept of integrated, multiscale modelling in chemical and process engineering, developing the general idea of Charpentier (2002). Raimondeau and Vlachos (2002) mention two equivalent approaches to the multiscale modelling: the bottom-up approach (mostly used) and the other where strong coupling between methods for different scales exists and information flow is bi-directional. Those integration steps of the three techniques along with the associated scales of scrutiny are schematically shown in Fig. 2. The first integration stages were creation of common platforms of CFD with PSM and also PSM with CCH to result in "computational process engineering" and "computational chemical systems", respectively. In a similar way, CFD with CCH are likely to form "computational transport modelling", what is depicted by broken lines in Fig. 2. The last stage would probably be the final integration of the three bi-method platforms to create "computational product manufacturing".

The proposed name of the multiscale method of computational product manufacturing, CPM, emphasizes the aim of integration of different modelling methods at largely differing scales of scrutiny. CPM involves different scales of modelling and its definition is therefore similar to that of CAPE by Klatt and Marquardt (2009), however with emphasis on product engineering. Analysing processes of such a growing complexity will involve significant increase in the computational resources demanded by CPM. That aspect is a key reason for searching for more efficient modelling methods. Details of the integration aspects of two numerical methods, CFD with PSM and CCH with CFD and also with PSM are presented in the next chapter.

The proposed multiscale approach identifies framework of methodologies and modelling tools aiming at the design of the product manufacturing accounting for customer required properties. Such an approach is characteristic of chemical product design, which integrated with manufacturing process design constitutes the core of chemical product engineering (Costa et al., 2006).

4. Applied integration of numerical modelling

4.1. PSM with CFD

Starting roughly from year 1995 and in parallel to independent development of the analysis tools of both computational fluid dynamics and process system modelling, papers were published on joint use of the two methods to enhance the modelling quality both of the phenomena and of processes taking place in process equipment. Those attempts were undertaken equally for achieving products of required description and also to optimize the whole technological system.

The last generation of the PSM codes is very advanced and enables simulations of complex systems and processes involving many components, several phases and accompanying chemical reactions. However, in that approach significant simplifications and assumption of some process quantities are employed (Kramer, Dijkstra, Neumann, Meadhra, & van Rosmalen, 1996; Zitney & Syamlal, 2001). On the other hand, CFD is usually applied when detailed modelling results are required. It is especially important when the yield of simulated equipment is strongly dependent on the transport kinetics in fluids. Up to now, the most popular applications of CFD in process engineering are gas-sparged reactors, stirred reactors, crystallizers and combustion processes (Jaworski, 2005).

A potential joint application and integration of the CFD and PSM codes is numerical simulation of process conditions of complete technological systems. In the case of incorporation of several equip-

Table 3
Basic characteristics of the integrated modelling methods.

Modelling tool	Computing stage	Input information	Output information
PSM	Generating system structure (flowsheet)	Selected unit operation blocks (models) and connecting streams	The flowsheets
	Simulation computing	Choosing thermodynamic model for process media properties, input of given data for: (i) stream data: components, P, T, mass flow rate, heat flow rate; (ii) equipment unit models with specific information for the unit characteristics	Data for: (i) material streams: P, T, mass flow rate, heat flow rate, composition of individual streams, their enthalpy and entropy; (ii) energy streams; (iii) T, P in equipment units, heat duty, flow profiles
	Steady-state optimization of streams and equipment units along with cost optimization	In sensitivity analysis: carrying out several simulations for different values of optimized variables based on the existing flowsheet. In cost optimization – selection of objective function and optimization method	One optimum system or more sub-optimum systems
CFD	Preprocessing – generating equipment geometry & numerical grid	Definition of process equipment geometry and boundary conditions	Numerical grid of equipment with implemented boundary conditions
	Solving – numerical iterations to resolve sets of differential equations	Physical properties of process media, numerical grid, selecting: sub-models, solution algorithms, initial guess and convergence strategy	Local numerical data for: hydrodynamic parameters; heat transfer, species concentration, chemical reactions and interfacial processes
	Postprocessing – adapting simulation data	Output data from solving stage	Average values and visualised distributions of selected process parameters
CCH	Quantum mechanics	Mass and charges of both nuclei and electrons of molecule, structural formula. Selection of the approximation method	Electron energy density, natural orbitals, molecule optimum geometry and properties
	Atomistic scale methods (MM, MD and MC)	Initial position and momentum of molecules, their interaction parameters, e.g. Lennard-Jones	Equilibrium state of molecules delivering thermodynamic macro-properties and molecular structure
	Mesoscale techniques (DPD, MesoDyn)	From atomistic simulations: the bead size and Gaussian chain architecture, the Flory–Huggins parameter, χ, and bead self-diffusion coefficient (MesoDyn)	Morphology and structure of the matter at nanoscale level, volume fractions of components
	microFE	Positions of the interfaces separating the inclusions and the matrix and periodic boundary conditions from Monte Carlo or mesoscale techniques (Gusev, 2001)	Effective properties of multiphase materials

ment units and/or unit operations in the simulated production process, usually a key unit in the system, e.g. a reactor, determines the efficacy of the entire system. With the unit already indicated, one can evaluate its performance in detail by employing a CFD package for modelling all important characteristics such as productivity, selectivity, etc. The CFD results can be exported to the PSM module and then imported back with varied parameters of the key unit for optimizing the whole production process.

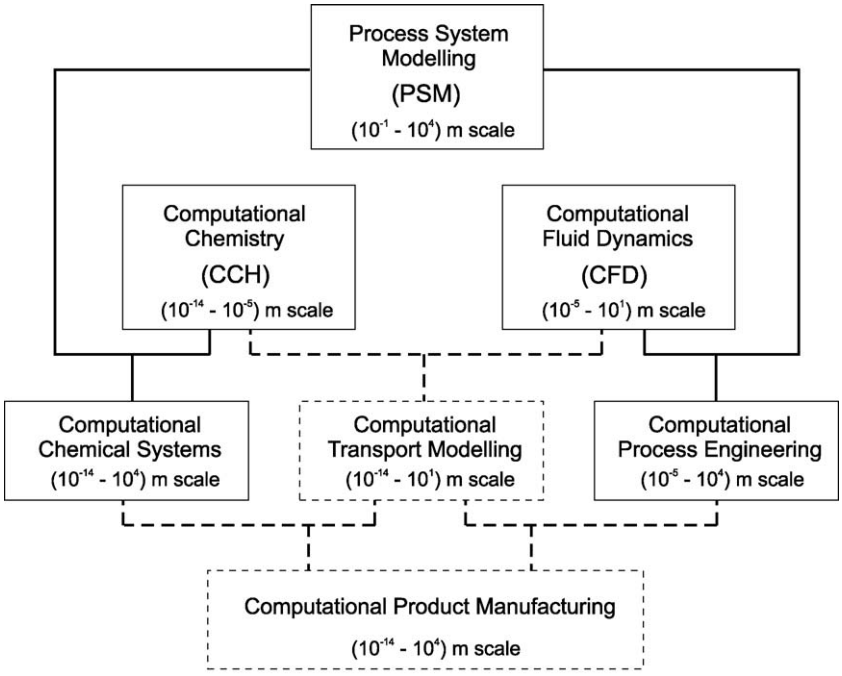

Fig. 2. Scheme of multiscale modelling integration.

Perhaps first examples of such integration in a successful joint modelling of a chemical plant with CFD and PSM codes were published by Zitney and Syamlal (2001, 2002). The system consisted mainly of chemical reactors, heat exchangers and distillation columns. The role of key unit was assigned to a stirred reactor with a reaction strongly dependent on the flow patterns. The CFD programme used values of the input process variables delivered from the PSM model. After completion of iterations, the CFD code returned to the PSM code the results on the flow rate and reactant concentration in the outflow stream from the reactor. The alternate use of the two codes was continued till the prescribed system convergence was reached.

The published results suggest that the integrated use of the two different modelling methods enable us to analyse and optimize overall plant performance with respect to mixing and fluid flow behaviour. A similar way of a combined application of the two numerical tools proposed by Bezzo et al. (2000), however to a small-scale system. Since then that approach has been developed in applications to model fuel cell systems (Syamlal, Madsen, Zitney, & Rogers, 2003) or advanced power systems modelling (Lang, Biegler, Munteanu, Madsen, & Zitney, 2007; Swensen, Zitney, & Bockelie, 2007).

Integration of the two modelling tools was also applied to single units with complex processes initially analysed by means of a process simulation tool. Nowadays the PSM programmes are increasingly advanced and the accuracy of unit models used in them is being improved. However, especially if a unit considered is a bottleneck of the system then it is usually beneficial to make use of more accurate CFD predictions of the transport processes within the unit as an alternative to the simplified models in PSM. For example, significant problems can arise when such a process, e.g. crystallization, is to be scaled up from the laboratory dimension to full technical scale and its kinetic model was experimentally derived (Kramer et al., 1996, 1999). This is because constants of those models are usually derived in hydrodynamic conditions present in the small-scale equipment and are hardly representative to the conditions of the full scale equipment. A good example here is the conclusion of Jones, Rigopoulos, and Zauner (2005) that the assumption of "well-mixed" conditions throughout the whole crystallizer leads to significant inaccuracy in estimating the rate of crystal growth, nucleation and agglomeration. Nevertheless, application of CFD methods became very helpful both in deriving local variables and in proper averaging them for the whole unit. On the other hand, one can find that exclusive use of CFD for modelling such complex processes is often problematical (Jones et al., 2005) due to involvement of substantial computer resources. It results from resolving by CFD codes of a set of several differential equations, which usually are non-linear. Therefore, the integrated use of the two numerical tools, CFD and PSM is a good compromise and that hybrid technique was used for modelling processes of various type in single equipment units, for example in crystallizers (Bermingham, Trivellato, Vernier, & Bezzo, 2007; Bezzo, Macchietto, & Pantelides, 2004; Choi, Chung, Oh, & Kim, 2005; Kougoulos, Jones, & Wood-Kaczmar, 2006; Kramer et al., 1999; ten Cate, Bermingham, Dersken, & Kramer, 2000; Urban & Liberis, 1999), sparged reactors (Rigopoulos & Jones, 2003), precipitators (Jones et al., 2005; Zauner & Jones, 2002) or reactive separators (Klöker, Kenig, & Górak, 2003). Nonetheless, the hybrid approach is also associated with some degree of simplification (Jones et al., 2005).

Separate numerical grids for CFD and PSM were used in the hybrid method. Relatively dense numerical grids were chosen in CFD to obtain good quality of flow predictions. Quite the opposite lumped models or crude numerical mesh are usually applied in the PSM codes for faster simulating of complex chemical processes. Various ways of partitioning the working volume into zones of ideal mixing were applied, e.g. Zauner and Jones (2002), Kagoshima and Mann (2005), Bermingham et al. (2007), Kramer et al. (1999), Rigopoulos and Jones (2003), Urban and Liberis (1999). One can also observe a growing role of the CFD computations in the recent work (Choi et al., 2005; Kougoulos et al., 2006). Due to rapid development and increasing capabilities of CFD codes, designating the computing task mainly to CFD enables improving the predictions of scale-up of the simulated process (Kougoulos et al., 2006) and, in consequence, quality of the designed product.

It is generally approved that a significant obstacle in the modelling with a joint application of two or more numerical codes is a low efficiency of data exchange between the packages usually of different architecture and programming language. To achieve a well-organized integration of the codes, both the correct communication and computing sequence must be realized. According to Bezzo et al. (2000), the simplest way of integration of the CFD and PSM codes is their sequential execution. Usually the CFD code is applied first to predict the hydrodynamic conditions and then the obtained data are introduced to the model created in the process simulation software. However, in many practical applications the process hydrodynamics can evolve as a result of changes in composition and temperature. Therefore, more precise results of hybrid simulations are achieved by exchange of data between the two codes subsequent to each iteration (Bezzo et al., 2000). Bermingham et al. (2007) propose, after each completed iteration, transferring information on mass flow rate and important hydrodynamic quantities from the employed CFD code to the process simulation code. Updated viscosity and density should be passed in the reverse direction after completion of the PSM computations. It is very likely that this would be the main investigation area in the near future for the hybrid applications in complex processes.

A technique of integrating packages of CFD with a process simulator for modelling an adsorption storage tank was proposed by Mota, Esteves, and Rostam-Abadi (2004). They used a CFD code (Fluent) for predicting gas flow and heat transfer and the process simulator (AngTank) for simulating the dynamics of the adsorbent beds. The two codes worked independently and shared memory for mutual communication. A user defined function in Fluent activated alternatively and controlled data via a boundary interface between the two computational domains. A more advanced link, called foreign object interface, FOI, was presented by Bezzo et al. (2000) to support integration of the PSM package gPROMS for process simulation with the CFD package Fluent. The authors suggested that the FOI interface should be incorporated within the PSM package for a better overview and control of the whole process. In the methodology, the CFD code was equipped with the used defined function only to provide hydrodynamic data to gPROMS.

A general scheme of data exchange between the PSM and CFD programmes is depicted in Fig. 3. The middleware, which participates in data exchange and management, can be a part of either a PSM package (Bezzo et al., 2000) or a CFD (Mota et al., 2004) or an independent interface. Other characteristic features of the middleware will be described in Section 4.3.

4.2. CCH with CFD and PSM

Direct and joint applications of typical CCH and CFD packages were published only recently in the subject literature. Numerical studies in that area were initially devoted to hybrid schemes linking molecular particle dynamics with theoretical continuous dynamics. Perhaps the first successful generic algorithm, for coupling non-equilibrium molecular dynamics (MD) in one region with continuum dynamics (Navier–Stokes, NS) in another, was published by O'Connell and Thompson (1995). Their hybrid scheme assumed the continuity of stress by introduction of an overlap region—a

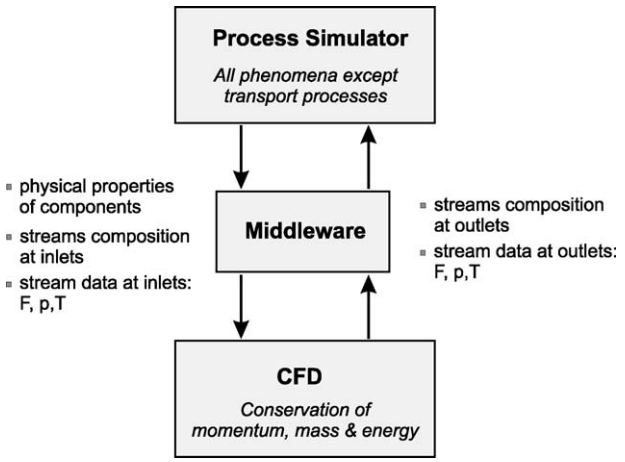

Fig. 3. Data exchange between PSM and CFD software packages.

hybrid solution interface where thermodynamic properties and the velocity field was consistent with both MD and continuum. They validated the hybrid approach for a uni-directional flow and concluded their algorithm can be applied to other flows of full complexity with the restriction that the overlap region must reflect the range of the shifted Lennard-Jones potential.

A further development of the hybrid MD-continuum method was reported by Nie et al. (2004). The coupling was achieved through constrained dynamics in the overlap region and consistent boundary conditions for fluxes at both sides of the overlap. The MD simulations used the shifted Lennard-Jones potential and the Navier–Stokes with continuity equations represented the fluid continuity. The approach was validated for sudden-start Couette flow and compared very well with exact solutions. The state-of-the-art in 2004 in the hybrid molecular-continuum fluid dynamics was summarized by Delgado-Buscalioni and Coveney (2004) who proposed a coupling scheme with two sub-cells overlap region where two-way exchange of hydrodynamic information occurs. They indicated that the coupling-through-fluxes scheme by Flekkøy, Wagner, and Feder (2000) delivers correct results. They also indicated the lattice-Boltzmann method as a candidate to interface with the MD domain. A similar hybrid MD-continuum approach with an overlap region was applied by Yen, Soong, and Tzeng (2007) to model flows in nano/mesoscale channels for resolving Couette and Poiseuille flows. They proposed a new way of force evaluation in the hybrid data exchange. A model for indirect coupling of MD simulations with the continuum NS equations was also proposed (Wang & He, 2007). The authors decomposed the computational domain into three regions: atomistic, continuum and their overlap with the particle momentum constraint.

A hybrid method combining MD with LB for simulating several types of colloidal (macroion) systems was described by Chatterji and Horbach (2005). Both the positive and negative features of that approach were expressed. In a later study, Chatterji and Horbach (2007), accounted for hydrodynamic interactions by applying a hybrid simulation scheme, where the charged ions propagated via molecular dynamics were coupled to a lattice-Boltzmann fluid. A general hybrid model to simulate dynamics of macromolecules in electrolyte solution with microions was also proposed by Giupponi, De Fabritiis, and Coveney (2007). The hydrodynamic forces were computed by a special fluctuating hydrodynamics solver coupled with macromolecular dynamics.

An advanced coupling of CFD and non-equilibrium MD simulations was proposed by Yasuda and Yamamoto (2008) who developed a multiscale hybrid method based on a local sampling idea. Typical CFD simulations based on lattice-mesh were used with CFD as a fluid solver and the MD simulations with a simple Lennard-Jones fluid. MD delivered local properties by carrying out local statistical sampling at all CFD nodes. Each lattice was associated with a small MD cell, which created local stresses according to the local flow field conveyed from CFD. It was found that in order to avoid the influence of fluctuations developing in MD, proper ratio of the time steps and length sizes in CFD and MD must be applied. The authors expect their hybrid method can be applied to complex rheology fluids.

Perhaps the most comprehensive and coherent summary of the status of the multiscale approach, which involves the microscopic models, was presented by E et al. (2007), Raimondeau and Vlachos (2002) and Vlachos et al. (2006). The authors also defined the key feature of the hybrid approach as designing multiscale algorithms that are driven by the data. They defined general top-down strategies and underlined the advantage of coupling models of different scale and utilizing the specific features of them, e.g. scale separation for the fine scale problems. Vlachos et al. (2006) discuss the problem which scale and model should be applied for the design and control of complex systems. The challenges and requirements which face the multiscale systems tools are listed by Braatz et al. (2004). Then again, Ingram, Cameron, and Hangos (2004) propose framework class definitions for two-scale models in terms of microscale and macroscale balance volumes and add examples of the model applications to catalytic packed bed reactor.

Other examples of two-scale applications refer to the integration of the CCH and PSM tools. For example, the CCH models are then used for determining the missing thermodynamic data in process simulators (Fermeglia et al., 2003). Fermeglia and Pricl (2009) applied atomistic and mesoscale simulations to polymeric mixtures and showed how the CCH methods can be used in predicting macroscopic properties in materials science. In a similar way Stefanovic and Pantelides (2001) integrated molecular dynamics with the classical Newtonian equations of motion, which allowed them to compute the macroscopic properties of the bulk molecular system. Integration of the two computational tools is enhanced by employing the CAPE-OPEN standard interfaces (Fermeglia, Longo, & Toma, 2009; Morales-Rodriguez, Gani, Déchelotte, Vacher, & Baudouin, 2008; Sandrock & de Vaal, 2009).

Another approach to the multiscale modelling, however still within the CCH methods, was presented in a review by Fermeglia and Pricl (2009). The review was supported by several applications to the nanostructured material design with the use of the mesoscale techniques (DPD, MesoDyn). In the work, special attention deserved the finite element based approach (microFE) to predicting structures and properties of complex compounds. Due to similarity of the unstructured numerical grids applied in the FE method (Guseva & Gusev, 2008) to those used in CFD it seems likely that integration of the two modelling methods is one of the possible ways to link CFD with CCH. Another example of a successful integration of CFD with mesoscale modelling was reported by Raimondeau and Vlachos (2002) who applied multiscale integration hybrid algorithms for molecular-continuum coupling, where fluid phase (macroscopic scales) was modelled by direct numerical simulations (DNS) whereas the surface (mesoscopic scales) with a stochastic MC simulator. Values of the spatially averaged surface rates obtained from the MC simulation were input to the gas-phase equations and the output from them was the updated gaseous mole fractions needed in MC. The use of coarse-grained Monte Carlo (CGMC) methods for diffusion-reaction in microporous materials, on catalyst surfaces and pattern nucleation and evolution (Chatterjee, Snyder, & Vlachos, 2004) and also in permeation through a microporous membrane (Snyder, Chatterjee, & Vlachos, 2005) can serve as further illustration of employment of the existing modelling techniques to reach larger length and time scales.

Computations by means of the CCH and CFD methods require high computing power, usually accomplished on several CPUs in parallel. Recently however, high computational efficiency of the Graphics Processing Units (GPUs) to non-graphic or general-purpose applications has also been used. Perhaps the pioneering papers on the applications to CCH and CFD were those by Li, Wei, and Kaufman (2003) and Wei et al. (2004) where the lattice-Boltzmann problems were solved on graphics hardware. The enormous numerical capacity of the GPUs results from parallel processing on several CPUs. On the other hand they are quite inelastic and therefore unattractive in some applications. Employment of the GPU computing power was made easier since the application of the Compute Unified Device Architecture (CUDA) developed by NVIDIA as a parallel computing architecture with a new programming and instruction set architecture. It can be used with several programming languages such as C, Fortran, OpenCL and DirectCompute. That technology was also employed in molecular dynamics simulations (Chen, Ge, & Li, 2009; Liu, Schmidt, Voss, & Müller-Witting, 2008), Monte Carlo simulations (Preis, Virnau, Paul, & Schneider, 2009), calculations of the fluid flow and CFD-based analysis (Kampolis, Trompoukis, Asouti, & Giannakoglou, 2010).

4.3. CAPE-OPEN standard

The necessity of standardization of links between various programmes that are used in the wide area of computer-aided process engineering induced the CAPE-OPEN standard for interfaces to create an effective integration of different modelling approaches. Currently, the standard is being developed by the CAPE-OPEN Laboratories Network (CO-Lan) consortium (www.colan.org). CAPE-OPEN defines rules and common interfaces for integration and interoperability of process modelling software components. The standard has important advantages of: (i) an easy plug-and-play link between software components from various sources, (ii) possibility of employing those interfaces as parts of a common application framework for the flow of process information within the process modelling environment (Barrett & Yang, 2005).

According to the CAPE-OPEN standards, the computer programmes interchanging data are divided into two groups called software components. The higher scale component is usually labelled process modelling environment (PME) and it is represented by a simulation engine, external software and external simulator (Morales-Rodriguez et al., 2008). The second group is called process modelling components (PMC), which are programmes and/or databases usually for use within a PME. The PMC packages can be used for computing thermophysical properties (Radermecker, Dumont, & Heyen, 2009; Sandrock & de Vaal, 2009; Yang et al., 2008); simulation of a particular unit operation (Fermeglia et al., 2009; Floquet, Joulia, Vacher, Gainville, & Pons, 2009; Morales-Rodriguez & Gani, 2007; Morales-Rodriguez et al., 2008) and for solving of mathematical problems arising in process simulation or optimization. The CFD codes are assigned to the PMC group and currently there are a few studies on the integration of a process simulator with CFD (Lang et al., 2007; Swensen et al., 2007; Syamlal et al., 2003) presented at the CAPE-OPEN US Conferences.

A schematic linkage between the programmes, based on the proposal of Morales-Rodriguez et al. (2008), can be presented in Fig. 4. However, it does not show additional links that may exist, e.g. connecting CFD with a database (Osawe, 2005). The structure of liaising codes is formed of separate modules in which individual computations take place, both for steady state and dynamic simulations. The CAPE-OPEN standards are also relevant to the equation-oriented process modelling tools (Braunschweig, Pantelides, Britt, & Sama, 2000). Information on the required equations is transferred to the executive module where a set of equations is assembled and then they are resolved by interacting with appropriate numerical solvers (Braunschweig et al., 2000).

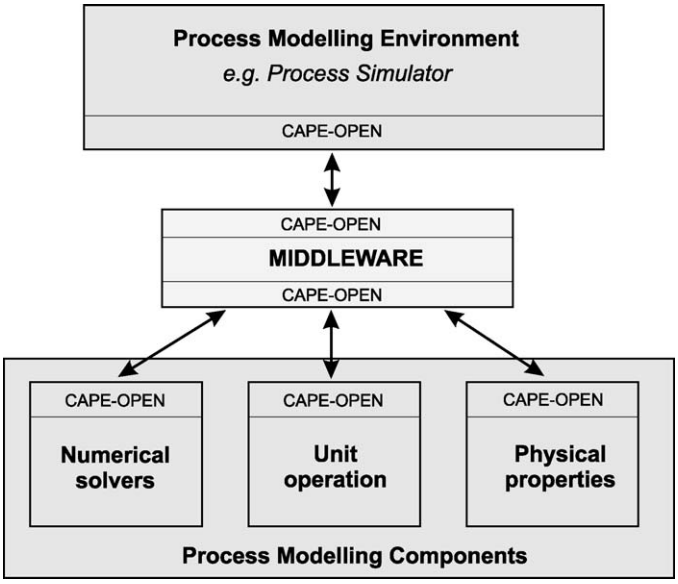

Fig. 4. Software integration based on CAPE-OPEN strategy.

Information transfer between a process simulator (PME) and individual modules (PMC) is accomplished through middlewares such as CORBA from Object Management Group for Unix and COM or .NET, both from Microsoft for Windows. The COM-CORBA Bridge software was generated for packages using the two different operating systems.

Another option was recently employed by using the Microsoft .NET as middleware for communicating between different software components. An overview of interoperability for COM/NET based CAPE-OPEN was enlightened by Barrett, Pons, von Wedel, and Braunschweig (2007). Possible errors that encounter in the process of integration of software components from different vendors using CAPE-OPEN can be debugged by the CAPE-OPEN Logging and Testing Tool (COLTT) provided by CO-LaN Consortium (Pons, Banks, & Braunschweig, 2007).

The CAPE-OPEN standards were successfully applied in packages of chemical and process engineering. The standards have already been introduced in some of the commercial process simulator tools to their source codes. At present, it is possible to employ several internal modules (COM models) in the Aspen simulations and also to register an own module in the "dll" format as a "CAPE-OPEN COM unit operation". Various types of control middleware were also used to data transfer and bridging the PME and PMC programmes. Among them COM and CORBA (Bezzo et al., 2000; Osawe, 2005; Yang et al., 2008) and also .NET (Barrett & Yang, 2005; Fermeglia & Parenzan, 2007) were applied most frequently. An important introduction of Advanced Process Engineering Co-Simulator (APECS) by Zitney (2005) should also be reported. Owing to APECS process/CFD co-simulations, computations are more efficiently and effectively carried out. Integration of CFD and PSM tools by that software was presented at a conference (Swensen et al., 2007; Lang et al., 2007).

5. Concluding remarks

The reviewed literature on modelling in the process industries shows clear trends towards coupling of the design of chemical and biochemical equipment/plants with the nanoscale approach to model (bio)chemical processes. It is generally expected that multiscale modelling can lead to cost optimization of the product

manufacturing. Three major types of auxiliary modelling software were used in the design, i.e. process system modelling, PSM, computational fluid dynamics, CFD, and computational chemistry, CCH. The software categories focused respectively on the efficacy of the production system, transport processes in individual equipment units, molecule structure and thermodynamics of the molecular processes. The modelling tools have independently been developed and nowadays have reached the maturity level enabling to successfully integrating pairs of them.

The application ranges of the PSM and CFD methods partly overlap but the CFD and CCH scales do not have a common range of application. In the last decade the first studies on integration of the CFD with PSM and of CFD with CCH were published, however, without generally approved criteria and tools of their joint application. Nevertheless, the original concept of integrated, multiscale modelling in chemical and process engineering (Charpentier, 2002) begun its implementation, first with PSM coupled with CFD and separately CFD with CCH and PSM with CCH. An efficient candidate for successful linking the three package types seems the CAPE-OPEN standard. Information transfer between a process simulator and individual modules can be carried out through software tools for interfacing packages applied in different operating systems.

It is therefore highly probable that the final integration of the two hybrid approaches will materialize soon to form the PSM–CFD–CCH system. The new system, called here computational product manufacturing, will involve significant increase in the computational resources demanded and more efficient modelling methods as well. The most probable use of the triple-system software is alternate use of the three components with gradual updating of the data transferred between them.

Currently, market customers decide on the required product quality, especially when polymers and nanotechnology products are concerned. It implies that research and development start from the molecular level and end up at the technical scale. However, practical implementation of a new technology is preceded by advanced designing procedures. Those activities are usually supported by computational modelling/optimization at various scales of scrutiny, both in time and length. Therefore, several literature examples of integration of two of the three described modelling tools were presented in this paper. Results of this study confirm common opinion that viable integration of the three modelling tools (PSM–CFD–CCH) will allow to speeding up and decreasing costs of the design of technological systems for manufacturing modern products. In addition, such the approach may contribute to optimization of the existing production lines.

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References

Altevogt, P., Evers, O. A., Fraaije, J. G. E. M., Maurits, N. M., & van Vlimmeren, B. A. C. (1999). The MesoDyn project: Software for mesoscale chemical engineering. *Journal of Molecular Structure (Theochem)*, *463*, 139.

Barrett, W. M., Pons, M., von Wedel, L., & Braunschweig, B. (2007). An overview of the interoperability roadmap for COM/.NET-based CAPE-OPEN. In V. Plesu, & P. S. Agachi (Eds.), *17th ESCAPE, Computer Aided Chemical Engineering, Vol. 24* (pp. 165–170).

Barrett, W. M., & Yang, J. (2005). Development of a chemical process modeling environment based on CAPE-OPEN interface standards and the Microsoft.NET framework. *Computers & Chemical Engineering*, *30*, 191.

Bermingham, S. K., Trivellato, S., Vernier, M., & Bezzo, F. (2007). Hybrid CFD-multizonal modelling of polymorphs and agglomeration phenomena in crystallisation processes. In *The eight international conference on chemical & process engineering* ICheaP-8, 24–27 June 2007,

Bezzo, F., Macchietto, S., & Pantelides, C. C. (2000). A general framework for the integration of computational fluid dynamics and process simulation. *Computers & Chemical Engineering*, *24*, 653.

Bezzo, F., Macchietto, S., & Pantelides, C. C. (2004). A general methodology for hybrid multizonal/CFD models. Part I. Theoretical framework. *Computers & Chemical Engineering*, *28*, 501.

Bird, R. B., Stewart, W. E., & Lightfood, E. N. (1960). *Transport phenomena*. New York: Wiley.

Braatz, R. D., Alkire, R. C., Rusli, E., & Drews, T. O. (2004). Multiscale systems engineering with applications to chemical reaction processes. *Chemical Engineering Science*, *59*, 5623.

Braunschweig, B. L., Pantelides, C. C., Britt, H. I., & Sama, S. (2000). Process modelling: The promise of open software architectures. *Chemical Engineering Progress*, *96*, 65.

Cameron, I. T., & Ingram, G. D. (2008). A survey of industrial process modelling across the product and process lifecycle. *Computers & Chemical Engineering*, *32*, 420.

Charpentier, J.-C. (2002). The triplet "molecular processes-product-process" engineering: The future of chemical engineering? *Chemical Engineering Science*, *57*, 4667.

Charpentier, J.-C. (2009). Perspective on multiscale methodology for product design and engineering. *Computers & Chemical Engineering*, *33*, 936.

Charpentier, J. C., & McKenna, T. F. (2004). Managing complex systems: Some trends for the future of chemical and process engineering. *Chemical Engineering Science*, *59*, 1617.

Chatterjee, A., Snyder, M. A., & Vlachos, D. G. (2004). Mesoscopic modeling of chemical reactivity. *Chemical Engineering Science*, *59*, 5559.

Chatterji, A., & Horbach, J. (2005). Combining molecular dynamics with lattice Boltzmann: A hybrid method for the simulation of (charged) colloidal systems. *Journal of Chemical Physics*, *122*, 184903, 1.

Chatterji, A., & Horbach, J. (2007). Electrophoretic properties of highly charged colloids: A hybrid molecular dynamics/lattice Boltzmann simulation study. *Journal of Chemical Physics*, *126*, 064907, 1.

Chen, F., Ge, W., & Li, J. H. (2009). Molecular dynamics simulation of complex multiphase flow on a computer cluster with GPUs. *Science in China Series B-Chemistry*, *52*, 372.

Choi, Y.-J., Chung, S.-T., Oh, M., & Kim, H.-S. (2005). Investigation of crystallization in a jet Y-mixer by a hybrid computational fluid dynamics and process simulation approach. *Crystal Growth & Design*, *5*(3), 959.

CO-LaN Consortium. (2003). *Methods & tools integrated guidelines*. Available at: http://www.colan.org/Spec%2010/Methods&Tools%20Integrated%20Guidelines.pdf.

Costa, R., Moggridge, G. D., & Saraiva, P. M. (2006). Chemical product engineering: An emerging paradigm within chemical engineering. *AIChE Journal*, *52*, 1976.

Cussler, E. L., & Moggridge, G. D. (2001). *Chemical product design*. Cambridge: University Press.

Delgado-Buscalioni, R., & Coveney, P. V. (2004). Hybrid molecular-continuum fluid dynamics. *Philosophical Transactions of the Royal Society of London A*, *362*, 1639.

Dzwinel, W., Yuen, D. A., & Boryczko, K. (2006). Bridging diverse physical scales with the discrete-particle paradigm in modeling colloidal dynamics with mesoscopic features. *Chemical Engineering Science*, *61*, 2169.

E, W., Engquist, B., Li, X., Ren, W., & Vanden-Eijnden, E. (2007). Heterogenous multiscale methods: A review. *Communications in Computational Physics*, *2*, 367.

Fermeglia, M., Longo, G., & Toma, L. (2009). Computer aided design for sustainable industrial processes: Specific tools and applications. *AIChE Journal*, *55*, 1065.

Fermeglia, M., & Parenzan, M. (2007). CAPE OPEN interface revisited in term of class-based framework: An implementation in.NET. *Chemical Engineering Transactions*, *11*, 235.

Fermeglia, M., & Pricl, S. (2007). Multiscale modeling for polymer systems of industrial interest. *Progress in Organic Coatings*, *58*, 187.

Fermeglia, M., & Pricl, S. (2009). Multiscale molecular modeling in nanostructured material design and process system engineering. *Computers & Chemical Engineering*, *33*, 1701.

Fermeglia, M., Pricl, S., & Longo, G. (2003). Molecular modeling and process simulation: Real possibilities and challenges. *Chemical & Biochemical Engineering Quarterly*, *17*, 19.

Field, M. J. (2007). *A practical introduction to the simulation of molecular systems*. Cambridge: Cambridge University Press.

Flekkøy, E., Wagner, G., & Feder, J. (2000). Hybrid model for combined particle and continuum dynamics. *Europhysics Letters*, *52*(3), 271.

Floquet, P., Joulia, X., Vacher, A., Gainville, M., & Pons, M. (2009). Numerical and computational strategy for pressure-driven steady-state simulation of oilfield production. *Computers & Chemical Engineering*, *33*, 660.

Foss, B. A., Lohmann, B., & Marquardt, W. (1998). A field study of the industrial modeling process. *Journal of Process Control*, *8*, 325.

Fraaije, J. G. E. M., van Vlimmeren, B. A. C., Maurits, N. M., Postma, M., Evers, O. A., Hoffmann, C., et al. (1997). The dynamic mean-field density functional method and its application to the mesoscopic dynamics of quenched block copolymer melts. *Journal of Chemical Physics*, *106*, 4260.

Frenkel, D., & Smit, B. (2002). *Understanding molecular simulation. From algorithms to applications*. London: Academic Press.

Gani, R. (2004a). Chemical product design: Challenges and opportunities. *Computers & Chemical Engineering*, *28*, 2441.

Gani, R. (2004b). Computer-aided methods and tools for chemical product design. *Chemical Engineering Research & Design*, *82*, 1494.

Giupponi, G., De Fabritiis, G., & Coveney, P. V. (2007). Hybrid method coupling fluctuating hydrodynamics and molecular dynamics for the simulation of macromolecules. *Journal of Chemical Physics*, *126*, 154903, 2

Groot, R. D., & Warren, P. B. (1997). Dissipative particle dynamics: Bridging the gap between atomistic and mesoscopic simulation. *Journal of Chemical Physics*, *107*, 4423.

Grossmann, I. E., & Westerberg, A. W. (2000). Research Challenges in Process Systems Engineering. *AIChE Journal*, *46*, 1700.

Gusev, A. A. (1997). Representative volume element size for elastic composites: A numerical study. *Journal of the Mechanics and Physics of Solids*, *45*, 1449.

Gusev, A. A. (2001). Numerical identification of the potential of whisker- and platelet-filled polymers. *Macromolecules*, *34*, 3081.

Guseva, O., & Gusev, A. A. (2008). Finite element assessment of the potential of platelet-filled polymers for membrane gas separations. *Journal of Membrane Science*, *325*, 125.

Hangos, K. M., & Cameron, I. T. (2001). *Process modelling and model analysis*. London: Academic Press.

Heinz, S. (2003). *Statistical mechanics of turbulent flows*. Berlin: Springer-Verlag.

Hessel, V., Hardt, S., & Löwe, H. (2004). *Chemical micro process engineering. Fundamentals, modelling and reactions*. Weinheim: Wiley–VCH Verlag GmbH & Co. KGaA.

Hill, M. (2009). Chemical product engineering—The third paradigm. *Computers & Chemical Engineering*, *33*, 947.

Ingram, G. D., Cameron, I. T., & Hangos, K. M. (2004). Classification and analysis of integrating frameworks in multiscale modelling. *Chemical Engineering Science*, *59*, 2171.

Jaworski, Z. (2005). *Numeryczna mechanika płynów w inżynierii chemicznej i procesowej Numerical fluid mechanics in chemical and process engineering*. Warszawa: Akademicka Oficyna Wydawnicza EXIT. [in Polish].

Jaworski, Z., & Zakrzewska, B. (2008). Multiscale modelling of chemical reactors. *Chemical and Process Engineering*, *29*, 567.

Jeżowski, J. (2002). *Wprowadzenie do projektowania systemów technologii chemicznej Introduction to designing systems of chemical technology*. Rzeszów: Oficyna Wydawnicza Politechniki Rzeszowskiej. [in Polish].

Jones, A., Rigopoulos, S., & Zauner, R. (2005). Crystallization and precipitation engineering. *Computers & Chemical Engineering*, *29*, 1159.

Kagoshima, M., & Mann, R. (2005). Effects of convection, feed-separation and macromixing on particle size distributions for double-jet semi-batch precipitation in a stirred vessel. *Chemical Engineering Science*, *60*, 2155.

Kampolis, I. C., Trompoukis, X. S., Asouti, V. G., & Giannakoglou, K. C. (2010). CFD-based analysis and two-level aerodynamic optimization on graphics processing units. *Computer Methods in Applied Mechanics and Engineering*, *199*, 712.

Klatt, K.-U., & Marquardt, W. (2009). Perspectives for process systems engineering—Personal views from academia and industry. *Computers & Chemical Engineering*, *33*, 536.

Klöker, M., Kenig, E. Y., & Górak, A. (2003). On the development of new column internals for reactive separations via integration of CFD and process simulation. *Catalysis Today*, *79–80*, 479.

Kougoulos, E., Jones, A. G., & Wood-Kaczmar, M. W. (2006). Process modelling tools for continuous and batch organic crystallization processes including application to scale-up. *Organic Process Research & Development*, *10*, 739.

Kramer, H. J. M., Bermingham, S. K., & van Rosmalen, G. M. (1999). Design of industrial crystallisers for a given product quality. *Journal of Crystal Growth*, *198/199*, 729.

Kramer, H. J. M., Dijkstra, J. W., Neumann, A. M., Meadhra, R. O., & van Rosmalen, G. M. (1996). Modelling of industrial crystallizers, a compartmental approach using a dynamic flow-sheeting tool. *Journal of Crystal Growth*, *166*, 184.

Kulikov, V., Briesen, H., Grosch, R., Yang, A., vonWedel, L., & Marquardt, W. (2005). Modular dynamic simulation for integrated particulate processes by means of tool integration. *Chemical Engineering Science*, *60*, 2069.

Kurowski, L., Chmiel-Kurowska, K., & Thullie, J. (2009). Numerical simulation of heat transfer in nanofluids. In J. Jeżowski, & J. Thullie (Eds.), *19th ESCAPE, Computer Aided Chemical Engineering, Vol. 26* (pp. 967–972).

Lang, Y.-D., Biegler, L. T., Munteanu, S., Madsen, J., & Zitney, S. E. (2007). Advanced Process Engineering Co-Simulation (APECS) using CFD-based reduced order models. In *AIChE annual meeting* November 4th–9th, 2007, Salt Lake City, Utah,. Session TD001. Presentation available at:. http://www.colan.org/Conferences/Y07_4thCOUS_398c.pdf

Li, W., Wei, X., & Kaufman, A. (2003). Implementing lattice Boltzmann computation on graphics hardware. *Visual Computer*, *19*, 444–456.

Liu, J., Chen, S., Nie, X., & Robbins, M. O. (2007). A continuum-atomistic simulation of heat transfer in micro- and nano-flows. *Journal of Computational Physics*, *227*, 279.

Liu, W., Schmidt, B., Voss, G., & Müller-Witting, W. (2008). Accelerating molecular dynamics simulations using Graphics Processing Units with CUDA. *Computer Physics Communications*, *179*, 634.

Lucas, K. (2007). *Molecular models for fluids*. Cambridge: Cambridge University Press.

Marquardt, W. (1996). Trends in computer-aided process modeling. *Computers & Chemical Engineering*, *20*, 591.

McWeeny. (1973). *Quantum mechanics: Methods and basic applications*. *The international encyclopedia of physical chemistry and chemical physics, Topic 2. Classical and quantum mechanics* Oxford: Pergamon Press Ltd.

Morales-Rodriguez, R., & Gani, R. (2007). Computer-aided multiscale modelling for chemical process engineering. In V. Plesu, & P. S. Agachi (Eds.), *17th ESCAPE, Computer Aided Chemical Engineering, Vol. 24* (pp. 207–212).

Morales-Rodriguez, R., Gani, R., Déchelotte, S., Vacher, A., & Baudouin, O. (2008). Use of CAPE-OPEN standards in the interoperability between modeling tools (MoT) and process simulators (Simulis® Thermodynamics and ProSimPlus). *Chemical Engineering Research & Design*, *86*, 823.

Mota, J. P. B., Esteves, I. A. A. C., & Rostam-Abadi, M. (2004). Dynamic modeling of an adsorption storage tank using a hybrid approach combining computational fluid dynamics and process simulation. *Computers & Chemical Engineering*, *28*, 2421.

Nie, X. B., Chen, S. Y., E, W. N., & Robbins, M. O. (2004). A continuum and molecular dynamics hybrid method for micro- and nano-fluid flow. *Journal of Fluid Mechanics*, *500*, 55.

NVIDIA CUDA™ Programming Guide v. 3.0. (2010). http://developer.download.nvidia.com/compute/cuda/3_0/toolkit/docs/NVIDIA_CUDA_ProgrammingGuide.pdf.

O'Connell, S. T., & Thompson, P. A. (1995). Molecular dynamics-continuum hybrid computations: A tool for studying complex fluid flows. *Physical Review E*, *52*(6), R5792.

Osawe, M. (2005). Fluent CAPE-OPEN COM/CORBA Bridge and CO-Compliant Unit Operation. In *2nd annual U.S. CAPE-OPEN meeting* May 25–26, 2005, Morgantown, WV, [Paper 6.1]

Pantelides, C. C., & Barton, P. I. (1993). Equation-oriented dynamic simulation current status and future perspectives. *Computers & Chemical Engineering*, *17*, S263.

Pons, M., Banks, P., & Braunschweig, B. (2007). A deliverable from CO-Lan to CAPE-OPEN developers and users: The CAPE-OPEN Logging and Testing Tool (COLTT). In V. Plesu, & P. S. Agachi (Eds.), *17th ESCAPE, Computer Aided Chemical Engineering, Vol. 24* (pp. 225–230).

Preis, T., Virnau, P., Paul, W., & Schneider, J. J. (2009). GPU accelerated Monte Carlo simulation of the 2D and 3D Ising model. *Journal of Comparative Physiology*, *228*, 4468.

Radermecker, E., Dumont, M.-N., & Heyen, G. (2009). Adaptation and testing of data reconciliation software for CAPE-OPEN compliance. In J. Jeżowski, & J. Thullie (Eds.), *19th ESCAPE, Computer Aided Chemical Engineering, Vol. 26* (pp. 303–308).

Raimondeau, S., & Vlachos, D. G. (2002). Recent developments on multiscale, hierarchical modeling of chemical reactors. *Chemical Engineering Journal*, *90*, 3.

Rigopoulos, S., & Jones, A. (2003). A hybrid CFD-reaction engineering framework for multiphase reactor modeling: Basic concept and application to bubble column reactors. *Chemical Engineering Science*, *58*, 3077.

Sandrock, C., & de Vaal, P. L. (2009). Dynamic simulation of Chemical Engineering systems using OpenModelica and CAPE-OPEN. In J. Jeżowski, & J. Thullie (Eds.), *19th ESCAPE, Computer Aided Chemical Engineering, Vol. 26* (pp. 859–864).

Snyder, M. A., Chatterjee, A., & Vlachos, D. G. (2005). Net-event kinetic Monte Carlo for overcoming stiffness in spatially homogeneous and distributed systems. *Computers & Chemical Engineering*, *29*, 701.

Stefanovic, J., & Pantelides, C. C. (2001). Molecular dynamics as a mathematical mapping. II. Partial derivatives in the microcanonical ensemble. *Molecular Simulation*, *26*, 167.

Succi, S. (2001). *The lattice Boltzmann equation for fluid dynamics and beyond*. Oxford: Clarendon Press.

Sukop, M. C., & Thorne, D. T. (2006). *Lattice Boltzmann modeling. An introduction for geoscientists and engineers*. Berlin: Springer Verlag.

Swensen, D. A., Zitney, S. E., & Bockelie, M. (2007). Development of Cape-Open unit operations for advanced power systems modeling. In *AIChE 2007 annual meeting* November 4th–9th, 2007, Salt Lake City, Utah,. Session TD001. Presentation available at:. http://www.colan.org/Conferences/Y07_4thCOUS_398e.pdf

Syamlal, M., Madsen, J. I., Zitney, S. E., & Rogers, W. A. (2003). Application of an integrated process simulation and CFD environment to model fuel cell systems. In *AIChE 2003 spring national meeting* March 30–April 3, New Orleans, LA.

ten Cate, A., Bermingham, S. K., Dersken, J. J., & Kramer, H. M. J. (2000). Compartmental modeling of an 1100L DTB crystallizer based on Large Eddy flow simulation. In *10th European conference on mixing* Delft, the Netherlands, 2–5 July 2000, (pp. 255–264).

Thullie, aJ., Kurowski, L., & Chmiel-Kurowska, K. (2009). A new experimental relation for effective thermal conductivity of nanofluids. In J. Jeżowski, & J. Thullie (Eds.), *19th ESCAPE, Computer Aided Chemical Engineering, Vol. 26* (pp. 949–954).

Tyczkowski, J. (2007). On the road to molecular engineering (in Polish). In *Proceedings of XIX Polish conference of chemical and process engineering, plenary lectures* (pp. 25–29).

Urban, Z., Liberis, L. (1999). Hybrid gPROMS-CFD modelling of an industrial scale crystalliser with rigorous crystal nucleation and growth kinetics and full population balance. *Chemputers 1999*. Dusseldorf, Germany, 21–22.10.1999, Session 12-Process Simulation (unpublished).

Vlachos, D. G., Mhadeshwar, A. B., & Kaisare, N. S. (2006). Hierarchical multiscale model-based design of experiments, catalyst, and reactors for fuel processing. *Computers & Chemical Engineering*, *30*, 1712.

von Wedel, L., Marquardt, W., & Gani, R. (2002). Modelling frameworks. In B. Braunschweig, & R. Gani (Eds.), *Software architectures and tools for computer aided process engineering. Computer-aided chemical engineering* (pp. 89–125). Amsterdam: Elsevier Science B.V.

Voncken, R. M., Broekhuis, A. A., Heeres, H. J., & Jonker, G. H. (2004). The many facets of product technology. *Chemical Engineering Research & Design*, *82*, 1411.

Wang, Y., & He, G. (2007). A dynamic coupling model for hybrid atomistic-continuum computations. *Chemical Engineering Science*, *62*, 3574.

Wei, J. (2007). Coordination of multi-scales in chemical engineering. *Chemical Engineering Science*, *62*, 3326.

Wei, X., Zhao, Y., Fan, Z., Li, W., Qiu, F., Yoakum-Stover, S., et al. (2004). Lattice-based flow field modeling. *IEEE Transactions on Visualization and Computer Graphics*, *10*, 719–729.

Widom, B. (2002). *Statistical mechanics. A concise introduction for chemists*. Cambridge: University Press.
Wozny, G., Li, P., & Wendt, M. (2001). Optimization in process engineering: State of the art and future trends. *Chemical and Process Engineering*, *22*, 137.
Yang, A., Braunschweig, B., Fraga, E. S., Guessoum, Z., Marquardt, W., Nadjemi, O., et al. (2008). A multi-agent system to facilitate component-based process modeling and design. *Computers & Chemical Engineering*, *32*, 2290.
Yasuda, S., & Yamamoto, R. (2008). A model for hybrid simulations of molecular dynamics and computational fluid dynamics. *Physics of Fluids*, *20*, 113101, 1.
Yen, T. H., Soong, C. Y., & Tzeng, P. Y. (2007). Hybrid molecular dynamics-continuum simulation for nano/mesoscale channel flows. *Microfluidics and Nanofluidics*, *3*(6), 665.
Young, D. (2001). *Computational chemistry: A practical guide for applying techniques to real world problems*. New York: J. Wiley & Sons, Inc.
Yuan, W.-K. (2007). Targeting the dominating-scale structure of multiscale complex system: A methodological problem. *Chemical Engineering Science*, *62*, 3335.
Zauner, R., & Jones, A. G. (2002). On the influence of mixing on crystal precipitation processes—Application of the segregated feed model. *Chemical Engineering Science*, *57*, 821.
Zhang, Z., Pike, R. W., & Hertwig, T. A. (1995). An approach to on-line optimization of chemical plants. *Computers & Chemical Engineering*, *19*(Suppl.), S305.
Zitney, S. E. (2005). CAPE-OPEN integration for Advanced Process Engineering Co-Simulation. In *2nd annual U.S. CAPE-OPEN meeting* May 25–26, 2005, Morgantown, WV, [Paper 2.3].
Zitney, S. E., & Syamlal, M. (2001). Integration of CFD and process simulation for improved plant design. In *AIChE 2001 annual meeting* [Paper 271g].
Zitney, S. E., & Syamlal, M. (2002). Integrated process simulation and CFD for improved process engineering. In J. Grievink, & J. van Schijndel (Eds.), *12th ESCAPE, Computer Aided Chemical Engineering, Vol. 10* (pp. 397–402).