



1. Introduction

Nowadays, numerical simulation plays one of the key roles in the area of solids process engineering. It is applied to describe, to analyze and to predict process behavior and to develop control and optimization strategies.

Simulation can be performed on different time and length scales considering different phenomena which take place. This can be a microscopic scale, where for example the internal particle structure is taken into account to simulate liquid penetration into pores. Alternatively, these may be much coarser scales where the thermodynamics of a whole apparatus is calculated.

In spite of the existence of various models on the different scales, the ultimate goal of process modeling is the simulation and prediction of a plant performance (Werther et al., 2011). In order to perform a modeling of plants, which often consist of an interconnection of numerous apparatuses and process substeps, the usage of flowsheet simulation systems is state of the art for the fluid processes. The analytical solution of processes, which have complex structures and where recycled energy and mass streams exist, is in most cases impossible.

The flowsheet simulation tools have become wide applications in the area of fluid processes, where nowadays various commercial and freeware software tools does exist (Hartge et al., 2006). In comparison with it, the flowsheet calculation of particulate systems has a much shorter history. In recent years, especially for solids processes, the steady-state flowsheet simulation system SolidSim has been developed (Pogodda, 2007). This framework is applicable to general solids processes; however, the ability to perform just steady-state calculations limits considerably the application areas of it. To date, engineers do not have a general simulation tool which is able to solve the tasks concerning dynamic solids behavior effectively. This gap in the area of dynamic flowsheet simulation of particulate processes and the huge interest expressed from the side of industrial companies are the main factors of intensified researches in this direction.

The work, which is presented in this contribution, is focused on the development of a novel system for dynamic flowsheet simulation of solids. On the one hand there was an aim to develop the simulation framework, on the other hand, to derive and to implement new dynamic models for the calculation of fluidized bed granulation processes.



The central role in the simulation framework plays the calculation algorithms and methods which are used to simulate flowsheet and to calculate energy and mass balances in all streams. There can be distinguished between two main methods, namely equation-oriented and sequential-modular. Each of them has its own advantages, but the modular approach can be more effectively used for the calculation of solids (Dosta et al., 2010). Due to the high complexity of the models of different apparatuses, the equation-oriented approach is confronted with insuperable difficulties. More detailed analysis of both these methods can be found in Chapter 2.

Parallel with a development of the simulation methods and architecture of the new framework, the dynamic models of numerous apparatuses and process substeps play an important role. The fluidized bed granulator was one of the first apparatuses, which was developed and added into the library of the dynamic models. The fluidized bed spray granulation is one of the widely used production processes in the chemical, pharmaceutical, food and agricultural industries (Mörl et al., 2007). In this process different production substeps, like wetting, drying, heating, etc. are combined into one apparatus. It allows producing dust-free, free-flowing particulate products with specified properties, such as particle size distribution, compounds percentage, density, etc. The description of the fluidized bed granulation is not a trivial task, because of the intensive heat and mass transfer, the huge influence of all three phases onto the behavior and complex fluid dynamics which take place. In the majority of cases the granulation plants have a complex dynamic or even unstable behavior. That is why dynamic calculations are necessary to simulate the process.

In recent years a lot of researches were performed in the area of modeling the granulation process, whereby models with different levels of detail were developed. As a first approximation, the empirical or semi-empirical models, which are based on population balance models, are used (Heinrich et al., 2002), (Hounslow et al., 1988), (Litster et al., 1995). Using the population balances, the transient behavior of the particle distribution due to the numerous events like growth, aggregation, breakage, attrition, etc. is analyzed (Ramkrishna, 2000). To obtain a more detailed description, the model can be extended by considering the heat and mass transfer which occurs in the apparatus (Heinrich and Mörl, 1999). Nevertheless, the material microproperties and apparatus geometry are poorly considered in these models.

With a purpose to perform more detailed modeling the granulation process can be described on smaller length and time scales. On the microscale a lot of particulate systems can be effectively simulated using the discrete element method, where each particle is considered as a separate entity (Cundall and Strack, 1979). The disadvantage of this approach is a huge computational effort, which does not allow performing calculation of a real apparatus on a long time interval. A



possible solution of this problem is a combination of submodels from different time and length scales into one multiscale model (Ingram et al., 2003). Hence, to give the possibility for the user to perform the simulation of a granulation process with high detailing grade, the novel multiscale model of a fluidized bed granulator was developed in this work.



2. Flowsheet simulation of solids

2.1 General methodology

Flowsheet simulation is used to perform quantitative modeling of industrial production processes and allows to predict properties, compositions and flowrates in the streams and main operating conditions. During flowsheet simulation the numerical solutions of energy and mass balances as well as intensive process variables for arbitrary materials and process structures can be calculated.

The investigated process is decomposed and represented as a set of individual submodels, which are coupled by energy and mass streams. In Figure 2.1 the process-flow diagram of an example structure is illustrated. The energy and raw material are introduced into the system through the input streams and after their transformation the resulting product leaves the flowsheet.

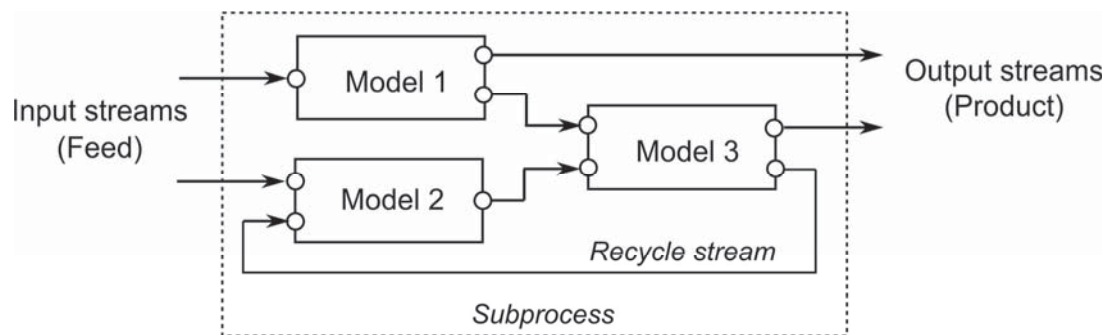


Figure 2.1: General process flowsheet

The scheme depicted in Figure 2.1 has simplified topology, while real industrial production processes can have a structure with much higher degree of complexity. In this case the main demands, which are related to the numerical simulation, are caused by the knotted network of material and energy streams and large amount of apparatuses and production substeps. In Figure 2.2 as an example, the general structure of industrial granulation process is shown. This process is used for fluidized bed granulation of urea (Uhde Fertilizer Technology). Because of the existence of a recycled material stream of milled granules the analytical prediction of the process behavior and its optimization is an unfeasible task. Therefore, the numerical solution with help of flowsheet simulation systems plays an important role in the industry.

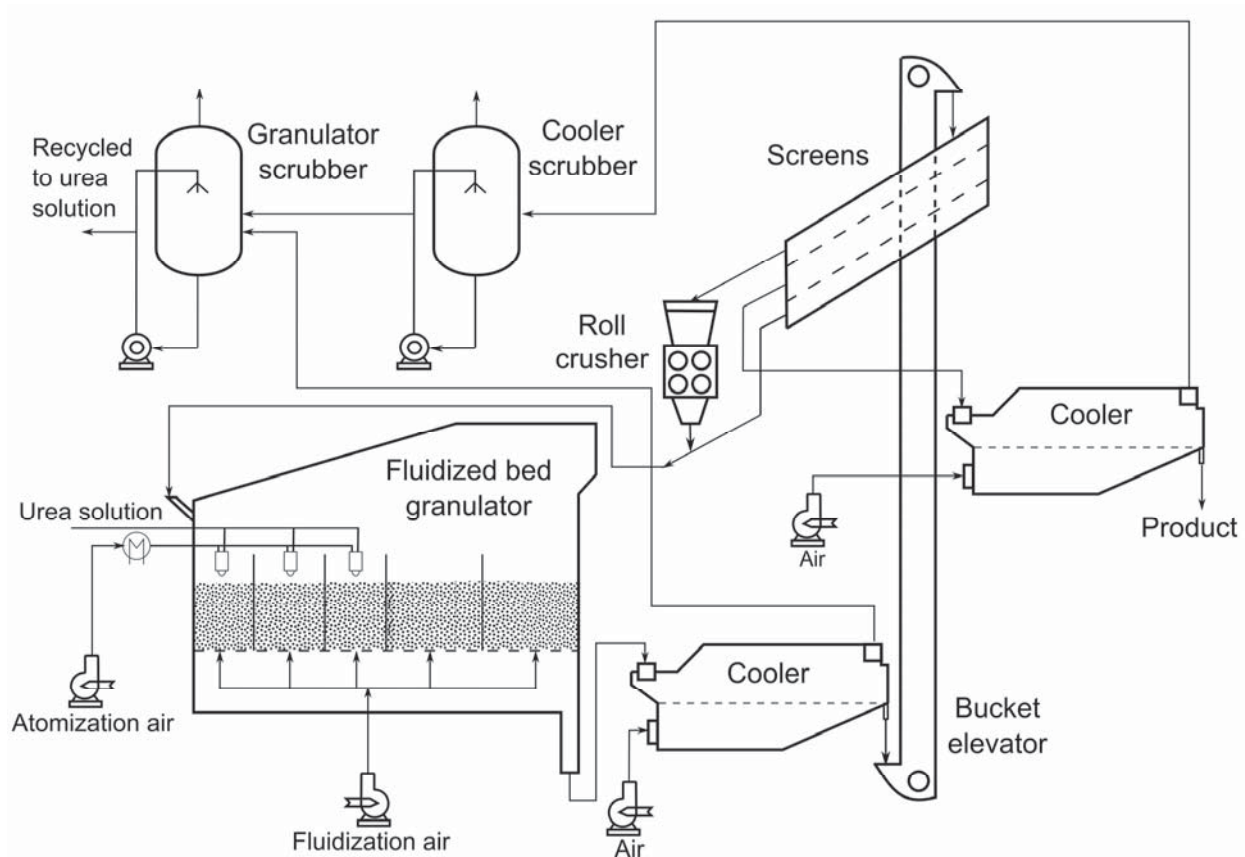


Figure 2.2: Flowsheet of urea granulation process

The flowsheet simulation can be performed in steady-state and dynamic modes. The dynamic simulation serves to obtain time dependent behavior and is much more demanding regarding computational effort. Despite the easier calculation procedure, the steady-state modeling not always allows to achieve sufficient results.

For example, transient process behavior during starting-up or shutting down phases often leads to the increased consumption of energy and raw material and cannot be modeled by a steady-state simulation tool. As a consequence, the usage of the steady-state simulation makes effective optimization infeasible. Another case, where just a dynamic simulation can be applied, is an unsteady process such as agglomeration or crystallization, which can be unsteady or can possess constant or damped oscillating behavior.

As mentioned, from the computational point of view two general classes of calculation strategies can be distinguished, which can be applied for the dynamic flowsheet simulation (Marquardt, 1991):

- equation-oriented (simultaneous) approach
- modular (sequential-modular) approach.



It should also be mentioned that a combination between the methods listed above can serve as effective calculation strategy, which is used for instance in the Aspen Dynamics framework (Aspen). In the Aspen Dynamics, the modular approach is used to find consistent initial conditions (Aspen Plus) and equation-oriented approach (Aspen Custom Modeler) to perform dynamic modeling.

In the case of the simultaneous approach, equations for the description of all units (physical properties, thermodynamics, mass and energy balances, stream connectivity, etc.) are combined into one homogeneous system of Differential Algebraic Equations (DAE's) or Ordinary Differential Equations (ODE's). In most cases this is a large set of equations, where small fraction of variables is included into any single equation (sparse). This system is solved simultaneously by a suitable integration method. Usually the Newton or quasi-Newton algorithms are used. That results in iterative calculation of Jacobian matrices and solution of large non-linear equation systems (Hindmarsh et al., 2005).

This equation-oriented approach can be applied for a flowsheet process which consists of open-form models (Schopfer et al., 2004). These types of models provide all information about the internal equations set, as it is required by an external numerical algorithm.

By the modular approach (Hillestad and Hertzberg, 1986), (Helget, 1997) the units are represented as "black box" models and every unit is solved independently from each other by its own calculation procedure. The calculation sequence corresponds to the flow of material on the actual process and the connectivity equations are solved implicitly by direct data transfer from output of one unit to the input of another.

This approach can be easily applied, when the flowsheet has a simple topology and does not contain recycle streams. In the case of complex structures with a material and energy feedback the modeling procedure becomes more complex. In the first stage the structural loops should be torn to render acyclic network topology. Afterwards, iterative calculation is performed until the convergence is reached. As convergence criteria the deviations of the tear stream on successive iterations is examined.

In Figure 2.3 the general principle of both approaches is illustrated. The exemplary flowsheet consists of a fluidized bed granulator and a screen apparatus and contains one recycle stream. By simulation with an equation-oriented (EO) approach, the equations are homogenized and calculated by one solver. The modular approach gives more flexibility in the choice of the calculation procedure, because each unit is solved separately and different solvers and calculation procedures can be used simultaneously.

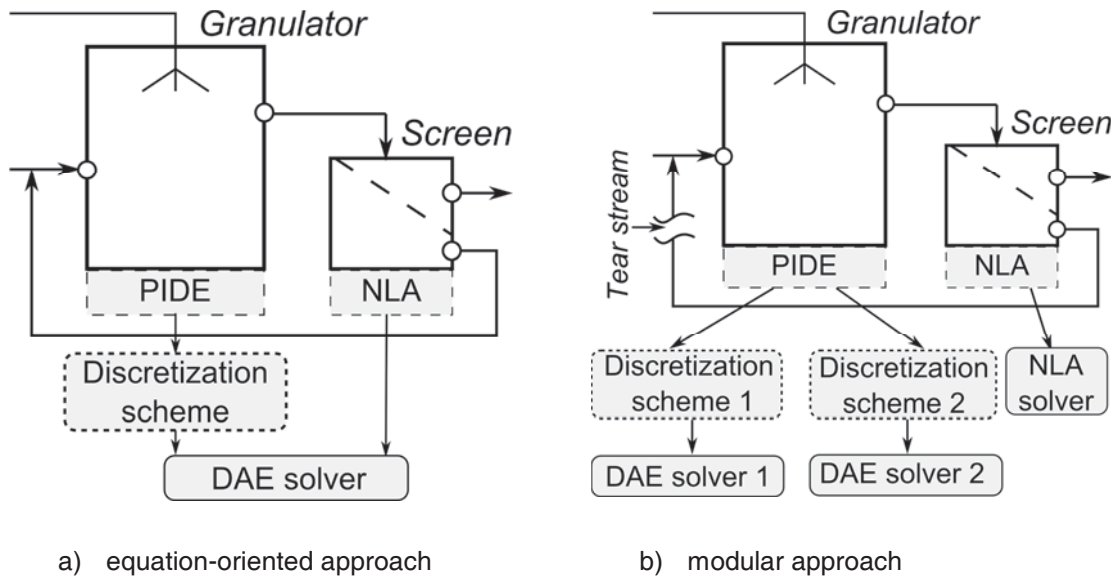


Figure 2.3: Difference between simultaneous and modular approaches

One of the main advantages of the equation-oriented approach, compared to the modular one, are better convergence properties, which can be reached especially in the situations when the flowsheet contains large number of recycle streams (Morton, 2003). However, heterogeneity of the mathematical models, which are developed in the area of solids processes, complicate the usage of simultaneous approach. These models can be consisted of ODE's, PDE's, linear and nonlinear algebraic equations as well as they can also have implicit and explicit (time events) discontinuities, which can modify the structure of the model. Further complexity arises from various particulate processes, for instance crystallization, granulation, agglomeration, drying. Commonly these processes can be described by a population balance model (PBM) and contain the partial integro-differential equations (PIDE) (Ramkrishna, 2000).

After the comparison of both above described strategies, the conclusion can be drawn that modular strategy, according to the set of advantages, can be more effectively applied for dynamic flowsheet simulation of solids processes. From the significant advantages of sequential-modular approach the following can be marked out:

- higher flexibility in the process of model development: Any closed-form model can be added to the flowsheet;
- only this approach can be used when a certain appropriate solver for simulation of the whole system does not exist;
- it leads to an easier procedure of the consistent initialization (Biegler et al., 1997): In the case of modular simulation, the process units are executed in the sequence according



to the structure of the flowsheet, which provides a reasonably good starting point for simulation;

- it allows to implement the effective methods for parallelization (Borchardt et al., 1999).

2.2 Complexity of solids processes

For many years, the usage of the flowsheet simulation frameworks is a state of the art in the area of Computer Aided Process Engineering (CAPE) (Schuler, 1995). However, in spite of the importance of solids processes, previous researches have been more focused on the fluid systems. Exactly, for the fluid processes the development of a flowsheet simulation methods was first started (Hlavacek, 1977), (Shacham et al., 1982), (Marquardt, 1991) and the first software tools for the flowsheet simulation were implemented.

Nowadays solids processes play an important role in chemical, pharmaceutical, agricultural and food industries. More than 60% of all products, which are sold by such companies as DuPont or BASF to the customers, are amorphous, crystalline or polymeric solids (Wintermantel, 1999). In order to satisfy the quality standards, these products should consist not only of specified chemical compounds, but they should also have a specific clearly defined size distribution, shape and physical microproperties. For instance, the flowability and stability of solids products play a decisive role in the minimization of transportation and storage costs.

For the transformation of the raw material into the final product, various types of processes with different conversion operations can be used. According to the classification proposed by Rumpf (1975) five mechanical treatment techniques can be categorized:

- splitting (grinding, cutting, deagglomeration, etc.);
- separation (classification, screening, sedimentation, etc.);
- agglomeration (compacting, tableting, granulation, etc.);
- mixing (homogenization, stirring, dispersing, etc.);
- transport, storage and dosing of disperse material.

The above described basic operations sequentially or simultaneously appear in the industrial processes. In Table 2.1 some process examples from different industries are listed.



Table 2.1: Examples of solids production processes

Process description	Industry
alumina calcination process	chemical
fluidized bed combustion process (Ratschow, 2009)	energy
urea granulation process (Uhde Fertilizer Technology) (see Figure 2.2)	agricultural
separation of contaminated dredged material into clean sand fraction and silt fraction with high contamination degree (Detzner, 1995)	environmental

Each of the listed examples consists of a complex interconnection of different apparatuses and basic process steps (unit operations), like screening, crushing, solids transportation, etc. To minimize the production costs and to create effective plant structures the material and energy streams are often re-used in the processes. Because of the existence of additional recycled streams this leads to an increase of the structure complexity. That is why in most cases even coarse analytical prediction of process steady-state and transient behavior are not possible. To solve this problem, the numerical calculations in form of flowsheet simulation should be used.

The necessity to distinguish the solids processes from liquid-vapor systems is not just by an additional phase, but rather by new simulation methods, was pointed out by numerous authors (Rossister and Douglas, 1986), (Barton and Perkins, 1988), (Evans, 1989), (Hartge et al., 2006), etc.). In the case of solids processes it is necessary to handle with a set of multidimensional distributed properties, which describe particle distribution by size, shape, habit, solid moisture content, etc. According to the complexity of the data, properties can be divided into three main categories (Pogodda, 2007) :

- distributed properties generally used for all types of distributed parameters such as PSD or stream composition;
- single-value properties have following data fields: numeric value, dimension and name. Each stream has at least four single-value properties such as temperature, pressure, mass flow and phase fractions of each phase;
- dependent single-value properties can have a different value for each individual interval of certain distributed property. For example, moisture content or density can be for different for different size fractions.



Application of the simulation methods developed for liquid-vapor systems can cause an incorrect process modeling or even the loss of required information. In Figure 2.4 a schematic representation of the calculation of the screen unit and appeared incorrectness are shown. Here, the separation diameter of the screen is 2 mm and the inlet solid stream consists of particles which are distributed by dissimilar color and size. To illustrate the appeared incorrectness it is assumed that the inlet stream consists of just four fractions, which are depicted in Figure 2.4.

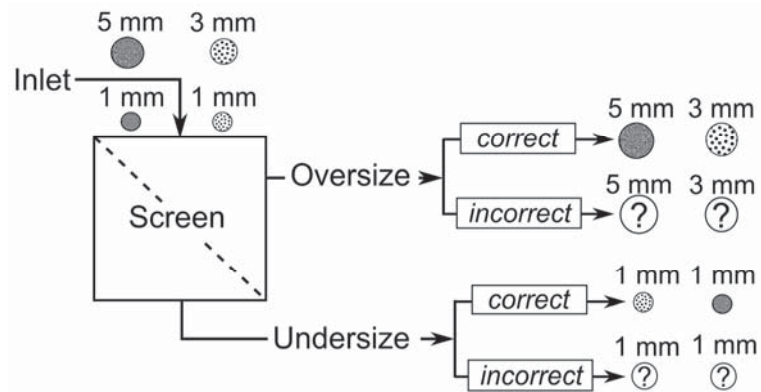


Figure 2.4: Example of incorrect solids treatment

If the inlet stream in Figure 2.4 is treated on the same manner like a fluid, the multidimensional properties will be mixed and as a consequence incorrect results will be received. One of the possible solutions of this problem was proposed and implemented in the SolidSim simulation environment (Pogodda, 2007). In order to achieve correct handling of multidimensional parameters the approach based on the stream transformation was developed. Instead of explicit calculation of models, in the SolidSim system a movement matrix is generated for every unit. This matrix is used to perform transformation of inlet to outlet stream.

A further question is how the distributed solid properties, as, for instance, the particle size distribution (PSD), are represented in the system. The conventional way to describe such parameters is the usage of the discretized form of PSD's. In this case, the variables are represented by a set of intervals along internal coordinates and assigned to them values. The application of a coarse grid can lead to a large simulation error, induced by numerical diffusion. Hounslow and Wynn (1992) have pointed out the disadvantages of the usage of the discretized PSD and have proposed a functional description of PSD as continuous parameter. However, in the case of flowsheet simulation the exact representation of parameter values in terms of functions is a challenging task (Töbermann, 1999). Furthermore, the discretization by a sufficiently fine grid decreases the numerical error to the values, which are significantly smaller in comparison with the inaccuracies, which arise due to the process simplification in the used empirical models.



Further challenge of processing of solids processes is induced by the complexity of the existing models and the necessity to use different numerical techniques to solve them. For instance, to describe the time evolution of the particle assemblies during various production processes, the usage of the population balance models (PBM's) is a state of the art (Ramkrishna, 2000) (Gerstlauer et al., 2006). They have been introduced into the area of chemical engineering by Hulburt and Katz (1964) and since that time they have experienced wide expansion and have been applied for many processes like granulation, crystallization, grinding, drying, etc. The general form of the PBM is represented as a partial integro-differential equation, the numerical calculation of which can be rather tedious.

Similar to other listed above demands, in the case of solids processes the apparatus geometry has a bigger influence on to the process behavior then in the case of fluids. For instance, the number of nozzles and their spraying angle or exact positions in the fluidized bed granulator can have a decisive influence on the growth kinetics of granules. Another example is a hydrocyclone, which separation characteristics of which depends on the apparatus diameter.

Werther et al., (2004) have formulated a set of requirements to the flowsheet simulation system of solids processes. The main necessary criteria are:

- general application to all types of processes which involve solids;
- consideration of liquid and vapour phases;
- existence of a unit library for numerous apparatuses and subprocesses;
- user-friendly graphical interface;
- documentation with information about implemented models and calculation methods.

Over the last few years, various flowsheet simulation programs have been developed particularly for solids processes and already existing systems were modified to handle solids. In Table 2.2 the most well-known systems are listed.

Despite the large number of entries listed in Table 2.2, neither of the above mentioned systems can be effectively used for dynamic simulation of general solids processes. Some of the programs are applicable just for processes with fixed structure or for a strongly limited number of unit operations, while other software tools can be used just for steady-state calculations. Therefore, after analysis of the state of the art, the conclusion about the necessity to develop a new modeling environment for the dynamic flowsheet simulation of solids processes can be drawn.



Table 2.2: Flowsheet simulation systems which are able to simulate solids

Program	Simulation type	Main application area	Last release
Metsim (Metsim)	Steady-state (some process can be modelled dynamically)	Chemical industry	2001 (Metsim ver. 19.01)
PMP FS Sim PMP FS Disp (Grainsoft)	Steady-state	Different size-reduction processes with classification units	2004
CHEOPS (Kulikov et al., 2005)	Tools integration framework for dynamic simulation	General solids processes	2005
JKSimMet (Morrison and Richardson, 2002)	Steady-state	Mining processing	2006
USimPack (Brochot et al., 2002)	Steady-state	Mineral processing	2007
Parsival (Wulkow et al., 2001)	Dynamic	Industrial crystallization, granulation processes	2009
Pro II (Invensys)	Steady-state	Polymer, fine-chemical and food industries	2010
FBSim	Dynamic	Fluidized bed granulation with predefined process structure	2006
gSolids (PSE)	Steady-state and dynamic	General solids processes	2012
AggFlow (AggFlow)	Steady-state	Mining industry	2011
SolidSim (Hartge et al., 2006)	Steady-state	General solids processes	2012
Aspen Plus, Aspen Dynamics, Aspen Custom Modeler	Steady-state, dynamic	Fluid processes with solid and gas phases	2012