Extended abstract of "Population balance modeling and passivity-based control of particulates processes, applied to the *Silgrain*® process"

Marta Dueñas Díez

1 Addressed problem and state-of-the-art

The goal of the thesis (Dueñas Díez 2004) was to establish a systematic strategy for the development of mechanistic models of particulate processes to be used for the purposes of design and implementation of an automatic control system, and to illustrate the suggested modeling and control strategies with a real industrial particulate process, the *Silgrain*® process.

Particulate processes are characterized by the presence of a continuous phase and a disperse phase made up of entities with a distribution of properties, and where the distribution of properties strongly affects the operation of the process and the product quality. Particulate processes are encountered in a number of applications: crystallization, agglomeration, grinding, dissolution, leaching, etc. Many valuable products are obtained in these processes. Therefore, it is of great interest to focus attention on particulate processes, and how to improve their performance.

The modeling of particulate processes has been studied for several decades, and Population Balance Equations (PBE) have become the most widely used modeling approach for such processes. A complete review on the use of the PBE is given in (Ramkrishna 1985) and (Ramkrishna 2000). Despite the remarkable active research on this topic, the PBE remains being mostly a tool for the academic community, and not for the industrial community. One reason may be that the simplest way to use the PBE, i.e. assuming complete mixing, may not provide realistic models. Accounting for the spatial distribution of the properties increases realism, but the resulting models become mathematically challenging, and impractical for industrial application. Establishing a systematic strategy for the development of PBE models that both represent realistically the operation of industrial units and that are mathematically simple enough to be used in online applications, is thus a first step towards a more widely use of PBE models in industry.

Developing a control strategy for particulate processes is of interest since automatic control is known to improve process operation, and to reduce process and product variability. Despite of the rapid and remarkable advances in modeling, numerical solution, and simulation of PBE models, the field of automatic control of particulate processes has not experienced as fast a development as could be expected. Examples of advanced control strategies implemented in real industrial settings are scarce. Some of the reasons that may explain the lack of advanced controllers for particulate processes are: the nonlinear and multivariable input-output behavior of such processes, the distributed nature of the population balance models (i.e. infinite number of internal states), limited instrumentation, limited number of manipulated variables, and batch or semibatch operation. Nonlinear and multivariable control approaches are thus the most suitable for a number of particulate processes, but they are also harder to implement than linear single-input single-output approaches. One theory that has not been studied in the framework of particulate processes before, is inventory passivity-based control (Farschman, Viswanath & Ydstie 1998). Such an approach was chosen in this thesis because it has some advantageous features related to stability and robustness, and the controller design is relatively straightforward. Extending inventory passivity-based control to reactive systems and particulate systems was thus believed to be an interesting area of research.

The *Silgrain*® process is a suitable industrial case study, since the process operation and the product quality are strongly affected by the dynamic evolution of the particle size distribution of the solid phase in the system. Moreover, the process had not been modeled before, and there was clear potential to improve automation of the process. The *Silgrain*® process is a hydrometallurgical leaching process where high-purity Si metal is produced by leaching lumps of $90-94\%$ FeSi in a hot acidic solution of ferric chloride ($FeCl_3$) and hydrochloric acid (HCl). The acid attacks the crystalline structure of the FeSi, selectively dissolving the intermetallic phases containing the impurities of Fe, Al, and Ca, while leaving the Si unattached. The exact reaction mechanism is unknown, but leaching is assumed to proceed according to the following reduction-oxidation reactions (Aas 1971):

$$\mathrm{Me} + n\,\mathrm{FeCl_3} \longrightarrow \mathrm{MeCl}_n + n\,\mathrm{FeCl_2} \tag{1}$$

$$\mathrm{Me} + n\,\mathrm{HCl} \longrightarrow \mathrm{MeCl}_n + \frac{n}{2}\,\mathrm{H_2} \tag{2}$$

where Me represents a metallic impurity in the intermetallic phases, i.e. any of the species Fe, Al, and Ca, and n is the corresponding oxidation number of the metal in the resulting metallic chlorides, i.e. 2 for Fe, 3 for Al, and 2 for Ca. The most characteristic feature of the *Silgrain*® process, which differentiates it from other leaching processes, is the disintegration of particles during leaching.

2 Thesis summary

The thesis is composed of three parts. Part I presents the suggested PBE modeling methodology, and its application to the *Silgrain*® process. Part II describes inventory passivity-based control, the extension of the theory to reactive systems and particulate systems, the application to the case study, and the analysis of certain practical issues. Part III summarizes the conclusions of the thesis.

2.1 Part I: Population Balance Modeling

The suggested systematic approach to mechanistic modeling of particulate processes consists of the following stages: establishment of model foundations, building of the model structure, determination of the constitutive relations, selection of a solution method and model validation. Such an approach is general to any type of process, but when dealing with particulate processes special care should be placed on some of these stages. Special emphasis was put on the establishment of model foundations. The widely-used assumption of complete-mixing turns out to be unrealistic in many instances, and should thus be avoided. A compartmentalization of the unit based on distinguishable regions in the unit is suggested. A macroscopic balance is used for each compartment, and the connections among compartments are defined based on the physics and hydrodynamics of the process.

Once the model is built, the model is "particularized" by defining the constitutive relations. The determination of constitutive relations corresponding to the continuous phase is fairly straightforward, since extensive research results are available on the phenomenological laws of transport phenomena and on reaction engineering. As regards the dispersed phase, general phenomenological laws for the events affecting the entities of the population have not yet been established. However, the modeler may take advantage of the existing results within each subfield of particulate processes, or tailor-made studies can be carried out.

The mathematical solution of PBE models is typically more challenging than the solution of other types of process models, mainly due to the fact that the PBE is a balance on a property distribution function. Hence, a macroscopic PBE typically results in a system of integrodifferential equations, whereas a microscopic PBE model typically results in a system of functional partial integrodifferential equations. Fortunately, extensive research results are available on this topic. In contrast, the area of parameter estimation and model validation of PBE models has not been studied to such a large extent yet. The main challenges encountered are:

- Gathering good experimental data is still difficult, particularly as regards the population density distributions.
- Computational issues are important. PBE models are large and nonlinear. Of particular importance is the fact that some parameters may be collinear for the given measurements, which results in ill-conditioning of the optimization problem, and poor parameter identifiability.

The use of a systematic parameter identifiability analysis prior to parameter estimation is discussed. Such an analysis provides a subset of parameters with two important properties: the measurements are highly sensitive to the selected parameters, and the selected parameters are not collinear with each other. In other words, the analysis provides the selection of parameters that can be identified from the available data.

Next, the methodology is illustrated by modeling the two reactors of the *Silgrain*® process. The model should capture the essence of the process and should provide realistic predictions of the process behavior. The operation of the *Silgrain*® process is as follows: FeSi lumps of relatively large size are fed in a semibatch mode at the top of the main reactor (HR) and sink towards the bottom. A relatively large flow of hot acidic solution is fed in a continuous mode at the bottom of the HR. Contact between the FeSi lumps and the hot acidic solution results in the leaching of the metallic impurities and the subsequent disintegration of the lumps. The fine grains generated in the disintegration process are displaced upwards through particle buoyancy and hydrodynamic thrust from the acid flow. The lumps that are only partially disintegrated, are still large in size, and remain in the bottom of the HR until they are further disintegrated. The stream flowing from the HR to the second reactor (UR) consists of acid and the fine disintegrated material. The top of the UR is designed as a sedimentation chamber, where most of the Si grains sediment while most of the acid leaves the reactor by the overflow on the top. Only very fine grains are entrained in the overflow stream. The settled grains in the UR react with the remaining acid in the packed bed. This is believed to proceed through pure chemical dissolution, and not through further disintegration. Tapping of the *Silgrain*® product at the bottom of the UR is carried out in a semibatch mode. After tapping, the product is subjected to diverse operations such as: filtering, drying, weighting, and packing. The overflow from UR is first circulated through a heating tank, and later on the composition of the acid is adjusted to the desired operational values before the acid is recirculated to the HR. The most characteristic feature of the *Silgrain*® process, which differentiates it from other leaching processes, is the disintegration of particles that takes place in the HR. The particle size distribution (PSD) of the feedstock affects the disintegration, and in turn, the disintegration process causes important changes in the particle size distribution. Hence, the PBE coupled to the traditional balance laws of mass, energy and momentum is the natural choice of modeling technique. According to the methodology, special emphasis was put on the establishment of the model basis and assumptions. A good compromise between model realism and model complexity was achieved when the HR and the UR were divided into two compartments, each:

- *Disintegration* region (compartment I): the bottom of the HR, where the feedstock is interacting with the leaching acid, resulting in disintegration. The disintegrated material that is fine enough to be entrained by the countercurrent acid, flows out from this region, whereas the coarse material remains in this region until it is further disintegrated.
- *Storage* region (compartment II): the top of the HR, where the fine material transported by the fluid remains for a short time before being further transported to the UR. Since the residence time is short, it can be assumed that no reaction is taking place.
- *Sedimentation* region (compartment III): the top of the UR, where separation takes place by gravity settling. Most of the acid and some of the finest grains leave the UR in the overflow stream. Meanwhile, most of the particles sink and sediment together with a small fraction of the acid.
- *Dissolution* region (compartment IV): the bottom of the UR, where the settled particles form a packed bed. The particles interact with the acid in the bed causing a further dissolution of the impurities. The dispersion of the properties in the axial direction is important and can not be neglected. There exists some dispersion in the radial direction too, but this can be neglected. Thus, a distributed model in the axial coordinate should be used; we are assuming plug-flow. Moreover, since the tapping at the bottom of the UR is carried out in a semibatch manner, the property profiles in the packed bed vary with time.

Figure 1 sketches the division in compartments for the *Silgrain*® process.

A macroscopic balance is thus written for each of these distinguishable regions. Region volumes are allowed to vary, and a force balance on the particle is used to find the cut size, which relates the PSD in the effluent of the compartment to the PSD within the compartment. Compartments I, II, and III are modeled this way. A microscopic model is used for compartment IV, since property profiles are encountered in the axial direction.

An experimental campaign at laboratory scale to determine the birth and death terms of the PBE, was carefully planned and was carried out by the thesis author at Elkem Research Centre. Raw materials from the process plant were used. The main factors affecting disintegration were investigated and quantitative equations for the constitutive equations were obtained. In view of the results, it was concluded that the disintegration rate is mainly influenced by temperature and by the initial particle size, in such a way that the smaller the initial particle size and/or the higher the temperature, the faster the disintegration is. The PSD of daughter particles fit well to a bimodal distribution with a mode located around a *fixed* particle size in the fine range and with a *mobile* mode whose location is dependent on the *mother* particle size. This led to the following theory: when the acid attacks FeSi, the intermetallic phases are dissolved, causing a particle to break up into a small number of still relatively large particles and a large number of Si grains, which are quite fine. The large daughter particles are subject to acid attack and breakup, giving rise again to grains and intermediate sizes. This process repeats until all the material is disintegrated into grains. The laboratory campaign was considered so successful by Elkem Research Centre, that the experimental procedure was adopted at other projects.

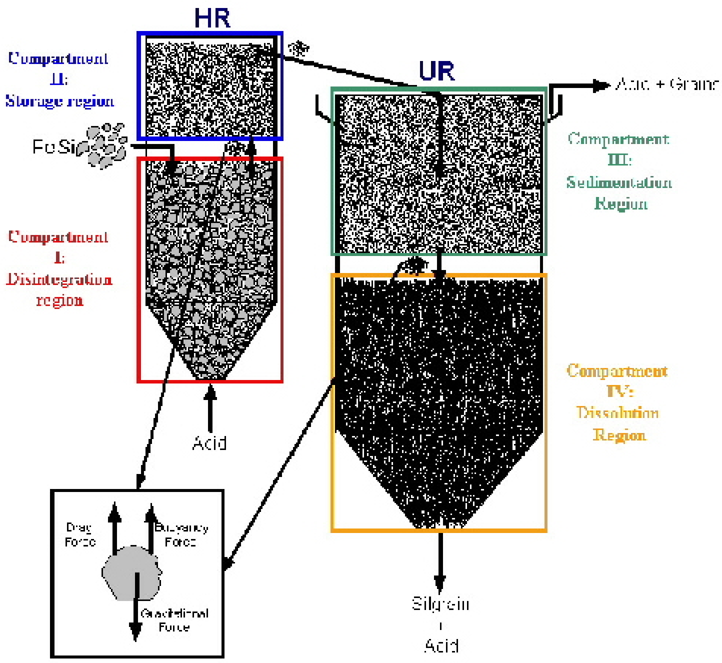

Figure 1: Compartmental modeling of the *Silgrain®* process.

The remaining constitutive relations are determined from the literature or from qualitative information of the process. Then, a solution method is chosen. The system of equations corresponding to compartments I, II, and III are reduced to an index-0 DAE (Differential and Algebraic Equations) by discretization in the particle size coordinate. A nonequidistant fixed grid is used for discretization, and discretization is implemented such that mass is preserved. As regards the model of compartment IV, the resulting functional PDAE (Partial Differential and Algebraic Equations) is first reduced to a standard PDAE by the method of moments, and subsequently reduced to a DAE by collocation.

The final stage is parameter estimation and model validation. The regular online measurements that are taken in the *Silgrain®* process are clearly insufficient to estimate parameters or validate the model. Therefore, a special measurement campaign was carefully planned (Dueñas Díez 2003) and carried out at the Elkem Bremanger plant at Svelgen (Norway) in May 2003. The campaign involved, for example, setting up indirect temperature measurements on the external wall of the UR, manually sampling and sieving feedstock sample, and sampling, sieving and analyzing product samples. Less amount of data were obtained than expected since some of the planned measurements turned out to be impossible to get. The available data could not be divided into a data set for estimation and a test data set, as was desired. Hence, the prediction abilities of the model could not be analyzed. The data were just enough to carry out the parameter identifiability and estimation analysis. The systematic method for parameter identifiability analysis was tested with the *Silgrain®* model. Such a method proved to be very useful. The parameter identifiability analysis reduced the number of parameters to be identified from 12 to 2 in the case of the HR, and from 31 to 6 in the case of the UR. After parameter estimation, the fitting of the model to the experimental data from the industrial plant was satisfactory. Although the prediction abilities could not be validated, the fact that the model fits well to the experimental data after parameter estimation is a positive result, and confirms the potential of PBE models for predictive purposes.

In addition to the original source code (MATLAB®), a graphical user interface (GUI) and an executable version of the model were delivered to Elkem ASA, such that they could use and continue developing the model once the thesis work was finalized. Indeed, the model has been used by Elkem ASA in other projects in simulation mode, to run predictions on how the process behave as conditions are changed, and to optimize process operation and process design.

2.2 Part II: Inventory Passivity-based Control

As regards automatic control, the attention was focused on a particular strategy: inventory passivity-based control. Although this control approach had never been applied to particulate processes before, and has seldom been applied to chemical processes, inventory passivity-based control was chosen in this thesis because it possesses some advantageous properties. First of all, the control is multivariable, and accounts for the nonlinearities of the process. Secondly, the method exploits directly the population balance model *as it is*, i.e. it does not require neither exhaustive manipulation nor size reduction of the model. Finally, convergence of the controlled outputs to their setpoints and stabilization of the remaining states of the system, can be ensured under certain conditions. The interconnection of systems that are rendered passive by the control law, is also passive and thus stable. The original theory for inventory passivity-based control by Farschman, Viswanath & Ydstie (1998) shows that the balance equations of process systems can be used to define a controller using feedback linearization that brings the controlled inventories to their setpoints by adjusting flows. The methodological framework is extended in this thesis to include reactive process systems and particulate process systems. The stability proof considered previously by Farschman, Viswanath & Ydstie (1998) considered only the convergence of the controlled inventories to their setpoints. The extended proof given in this thesis proves also stability of the remaining states of the system provided that certain controllability and detectability requirements are met. Proving detectability is particularly difficult for nonlinear systems. However, this thesis demonstrates how powerful theories in the field of nonlinear chemical dynamics can be exploited to check detectability in chemical reaction systems. The chemical network approach by Feinberg and Horn and Jackson (Horn & Jackson (1972), Feinberg (1980, 1991, 1995*a*, 1995*b*)) is introduced and connected with the inventory passivity-based control concept. A brief discussion about the link to nonequilibrium thermodynamics is also given. The control design methodology is illustrated with two examples: the van der Vusse reactor, and the *Silgrain*® process. It is shown that stabilization of the *Silgrain*® process can be achieved by using the following two manipulated variables: FeSi feedrate, and the temperature of the acid feed to the HR, to control the following two inventories: the internal energy and the mass of particles within compartment I (i.e. the mass of unreacted or partially reacted particles that are still too large to be transported by the acid). Hence, the Feinberg; Horn and Jackson theory proves that the internal states related to disintegration are stable, since the discretized PBE is equivalent to a zero deficiency reaction network, see Figure 2.

Some issues that are relevant for the practical implementation of inventory passivity-based control are also discussed. In the practical implementation of control strategies, the manipulated variables are often constrained. The presence of such constraints may make certain sets of setpoints unfeasible, and the controller performance may deteriorate. These issues come up on all control designs since the constraints are imposed

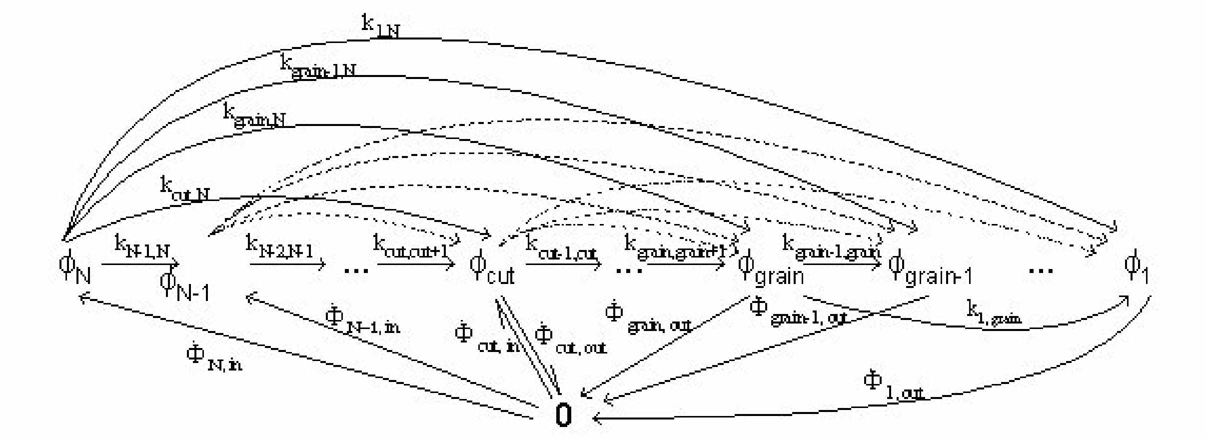

Figure 2: Feinberg diagram of the discretized HR dynamics.

by the process equipment and the process physics. Therefore, a prior analysis of setpoint feasibility is necessary, regardless of the method used for control. It is shown that inventory passivity-based control ensures convergence of the controlled inventories to their setpoints, and stabilization of the remaining states, provided that the selected setpoints are feasible and ϵ-controllable. For chemical reaction networks of zero deficiency, the closed-loop is stable even in the case of unfeasible setpoints. In such case, the system stabilizes but an offset is obtained. The effect of disturbances and model errors is also analyzed. Wang & Ydstie (2004*b*) suggested a method which combined inventory passivity-based control and sliding mode control. The method has been tested on the *Silgrain*® process with satisfactory results. The inventories converge to the desired setpoints and the remaining states stabilize, even in the presence of disturbances and model errors, as long as certain conditions are met. Moreover, the design of the controller is nearly as simple as in the nominal design. One of the main limitations of inventory passivity-based control is the need of knowing the state of the system in order to calculate the control law. Unfortunately, full state measurements may not be available. However, relatively realistic models are available or can be developed for most particulate processes. Therefore, observers can be constructed to calculate the unmeasured states. Many particulate processes operate in semibatch, such as the case under study. It is shown that inventory passivity-based control can be used in a straightforward way to automate such a semibatch operation. A brief discussion of the role of inventory passivity-based control within the overall operation of a plant is given. Inventory passivity-based control is just an element in the hierarchy of control functions in a chemical plant. It is mentioned how the stability of interconnections that characterizes passivity-based control can be advantageous in the framework of plantwide control. Finally, a brief discussion is given about the potential benefits of combining inventory passivity-based control with statistical process monitoring.

3 Applications/implementations/results

The contributions of this thesis regarding PBE modeling are:

- The PBE model of the *Silgrain*® process is entirely original, including the determination of the constitutive equations describing particle disintegration. Two experimental campaigns were specifically designed for model development: one at laboratory scale for the determination of constitutive equations, reported in (Dueñas Díez 2001) (confidential), and one at the plant scale for gathering data for parameter estimation and model validation, reported in (Dueñas Díez 2003) (confidential). Diverse versions of the model have been presented in (Dueñas Díez & Lie 2000), (Dueñas Díez, Ausland, Fjeld & Lie 2002) and (Dueñas Díez, Ausland & Lie 2003*b*).

- The importance of compartmentalization based on distinguishable physical phenomena in the process in order to achieve realistic yet simple models of industrial particulate processes is demonstrated in this thesis. The work on PBE modeling and compartmentalization has been presented in (Dueñas Díez, Ausland, Fjeld & Lie 2001) (later published in (Dueñas Díez, Ausland, Fjeld & Lie 2002)), (Dueñas Díez & Lie 2003*c*), (Dueñas Díez, Ausland & Lie 2003*a*), and (Dueñas Díez et al. 2003*b*).

- The importance of parameter identifiability analysis prior to parameter estimation of PBE models of industrial processes is illustrated with the *Silgrain*® process. This part of the work was presented in (Dueñas Díez, Andersen, Fjeld & Lie 2004), and has later on been published in (Dueñas Díez, Fjeld, Andersen & Lie 2006).

The contributions of this thesis regarding inventory passivity-based control are:

- The application of inventory passivity-based control to particulate processes is new. This work has been reported in (Dueñas Díez, Lie & Ydstie 2001), (Dueñas Díez, Ydstie & Lie 2002*a*), (Dueñas Díez, Ydstie & Lie 2002*b*), (Dueñas Díez & Lie 2003*a*) and (Dueñas Díez & Lie 2003*b*).

- The stability proof reported in (Farschman et al. 1998) has been extended in this thesis to account for systems with chemical reaction and particulate systems. Such systems are typically rectangular, i.e. systems with less manipulated variables than inventories. A detectability requirement is introduced in this thesis, and a way to check detectability, based on a published technique of nonlinear chemical dynamics, is suggested. It is the intention to submit such new theoretical results for publication in an international journal in the field of process control.

- This is the first time that the Feinberg and Horn and Jackson theory is applied to particulate processes, and connected to passivity-based control.

- Some issues that are important for the practical implementation of inventory passivity-based control, such as the presence of input constraints and robustness, are discussed, and some methods taken from literature are tested in simulation with the *Silgrain*® process, being thus tested for the first time on a particulate process.

The industrial partner in this project, Elkem ASA, has benefited from this thesis work as follows:

- The implementation of the PBE model of the *Silgrain*® process was delivered to Elkem ASA, both in the original source code in MATLAB® language, and in executable form including a GUI for easier use. Elkem ASA has been using the model for independent simulation studies.

- The disintegration experiments at laboratory scale and following data analysis suggested in this work has been adopted by Elkem ASA in at least one other project.

- The experimental campaign at the plant scale, together with the results from the identifiability analysis and inventory control, were used as basis to recommend improvements in the instrumentation and control of the industrial process.

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