Proceedings of the 9th International Symposium on
Dynamics and Control of Process Systems (DYCOPS 2010),
Leuven, Belgium, July 5-7, 2010
Mayuresh Kothare, Moses Tade, Alain Vande Wouwer, Ilse Smets (Eds.)

Multi-Scale Modeling and Inventory Control of Particle Growth Processes

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Abstract: In this paper, we develop a multi-scale model to describe the particle growth in a fluidized bed reactor. Population balance model is presented to describe the dynamical behavior of particle size distribution. Stability analysis is derived to determine the control configuration for the complex particulate process. Inventory control strategy is applied to control the particle size distribution.

Keywords: Population balance, stability analysis, inventory control, multi-scale modeling, particulate processes.

1. INTRODUCTION

Particulate processes have distributed properties along both external coordinates and internal coordinates. Hulburt and Katz (1964) used the theory of statistical mechanics to develop a continuous phase space description of the particulate system behavior. The so called population balance equation describes particles evolution in phase space. The population balance equation expresses the conservation of probability in the phase space. Population balance is usually difficult to solve because it can include partial integro-differential equations. Moment transformation and discretization are two commonly used methods of solution. Those methods simplify the continuous population balance equation and then obtain a tractable approximation of the equation. One problem with such dicretization schemes is that the conservation principles may not hold exact for precise gridding. Another problem is that the methods may be computationally expensive. White (2007) developed a discrete population balance which ensures that conservation laws are maintained at all discretization levels and facilitates computation without additional discretization. In section 2, we review this newly developed model and show its relationship with classical population balance model.

Fluidized bed reactors are typical examples of processes in which this type of dynamics are characterized by the production, growth, or decay of discrete particles contained in a continuous phase. Numerous computational studies have been carried out on fluidized bed processes. However, a simplified solution strategy with a complete and detailed model which integrates CFD, chemical reaction and population balances for possible use in process scale-up, detailed design and control studies is still missing. White et al. (2007) proposed a population balance model for size distribution of particulate processes using particle mass and number balances over discrete intervals. The novel approach for solving population balance equations proposed in this paper reduces the computation time considerably. They further showed the possibility of coupling population balance with CVD calculations to capture the dynamic particle size distribution. In continuation of the above mentioned works by White et al., we present the complete multi-scale modeling approach including the effect of computational fluid dynamics along with population balance and chemical vapor deposition models.

Particulate processes can exhibit sensitive response to changes in operating conditions. The continuous phase may operate on a completely different time scale than the dispersed phase. Much work has been accomplished to apply and solve the population balance for a variety of particulate systems. Semino and Ray (1995) proposed the first controllability analysis. Christofides (2002) tested nonlinear output feedback control on a crystallization process. Diez et al. (2008) was the first to apply inventory control to the particulate process. In this paper, we apply stability analysis to show that two manipulated variables are required to control the particle size distribution. The universal storage function developed by Antonio and Ydstie (2001) is employed to show the closed loop stability. In addition, inventory control strategy is implemented to control the particle size distribution.

2. POPULATION BALANCE

Particle phase space consists of the least number of independent coordinates which provide a complete description of the properties of the particle distribution. Those independent coordinates can be separated into two categories given by external and internal coordinates. External coordinates simply describe the spatial distribution of the particles. Such coordinates are not always necessary, such as, in the description of a well-mixed particulate process. Internal coordinates refer to the properties of each individual particle by measuring its state. The prime example is particle size. In the framework of this phase space, Randolph and Larson (1988) proposed a general form of population balance to represent the dynamical behavior of the particle distributions,

$$\frac{\partial n}{\partial t} + \nabla \cdot vn - B + D = 0. \tag{1}$$

Here, n represents the particle distribution function. The rate of change of particles along internal and external coordinate axes is represented by $\nabla \cdot vn$. The birth or death of particles due to particle agglomeration or attrition is represented by the B and D terms respectively.

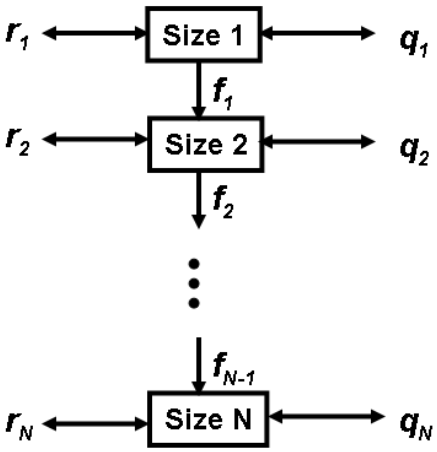

Fig. 1. Size interval characterization of particles

White et al. (2006) developed a finite dimensional population balance model to track the dynamic size distribution. They assumed that the particles are distributed along N discrete size intervals, characterized by an average mass m_i for $i = 1, ..., N$ seen in Figure 1. The discretized model consists of mass balance and number balance on each size interval. The relationship between the total mass of particles (M_i) in an interval and the number of particles in each interval (n_i) is given by

$$M_i = m_i n_i \tag{2}$$

The mass balance over size intervals is written as:

$$\frac{dM_i}{dt} = f_{i-1} - f_i + r_i + q_i + a_i \tag{3}$$

The rate of material transfer from the continuous phase to the particle is represented by r_i. The rate of transition of particles from one size interval to the next, caused by particle growth, is represented by f_{i-1} for flow into interval i and f_i for flow out of interval i. The external flow rate is q_i. a_i represents the rate of agglomeration or breakage of particles.

The discretized population balance model is closed by number balance model. The relationship(2) implies that the time derivatives of number can be written as

$$\frac{dn_i}{dt} = \frac{1}{m_i}\frac{dM_i}{dt} \tag{4}$$

The particle number balance over each size interval is represented as

$$\frac{dn_i}{dt} = \frac{f_{i-1}}{m_i} - \frac{f_i}{m_{i+1}} + \frac{a_i}{m_i} \tag{5}$$

They have shown that the discrete model approaches the continuous population balance (1) as the number of size intervals increases. Therefore the discrete population balance can be viewed as a physically based discretization scheme for continuous population balance.

3. STABILITY ANALYSIS FOR POPULATION BALANCE

In industry, many dispersed phase particulate systems are carried out in one or more regions which can be considered as well-mixed. The particle growth process we are interested is such case. The system can be represented only by the distribution of particles in the internal phase space. As reactor is assumed as well mixed, spatial distribution is not our main concern and hence the particle distribution function is only a function of internal coordinate axes. The macroscopic population balance represents the transient particle size distribution in a continuous, isothermal, well-mixed and constant volume crystallizer.

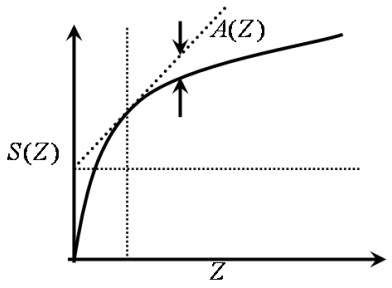

Fig. 2. Storage function for chemical processes

The particle behavior along the internal and external coordinate axes is described by the population balance equation, in the general form(1). Multiplying (1) by dV and integrating over V gives

$$\int_V (\frac{\partial n}{\partial t} + \nabla \cdot v_e n + \nabla \cdot v_i n - B + D) dV = 0 \tag{6}$$

Here, v_i is the internal coordinate velocity and v_e is the external coordinate velocity. The second term, an integral over the volume of the spatial divergence of the population flux can be transformed into the boundary condition of the population flux flowing into and outside the system. Hence,

$$\int_V \nabla \cdot v_e n dV = -Qn \tag{7}$$

Therefore the spatial averaged population balance can be written as

$$\frac{\partial n}{\partial t} + \nabla \cdot v_i n = B - D + \frac{Qn}{V} \tag{8}$$

As the internal coordinate is taken as particle size x and assume that particle growth rate is independent of size, an empirical observation known as McCabe's law holds true, the population balance equation (8) is reduced as,

$$\frac{\partial n}{\partial t} + G\frac{\partial n}{\partial x} = B - D + \frac{Qn}{V} \tag{9}$$

Here, $G \equiv \partial x/\partial t$, it is always positive as particle sizes keep increasing along time due to chemical reaction and G is the convective velocity of a particle along the x axis. Q is positive for flow into system and negative for flow out of system.

Ydstie and Antonio (1997) have proven that as long as dissipative effects dominate the chemical process, the process is guaranteed to be stable. The storage function referred as generalized availability is developed to show the stability of system. The geometry of the storage function is illustrated in Figure 2.

$$b(n, n^*) = [s(n^*) + A^*(n - n^*)] - s(n) \tag{10}$$

where $s(\cdot)$ is the entropy function and $A = \frac{\mu}{T}$ called intensive variable of the system. Here we assume $s(\cdot)$ is strictly concave in order to guarantee that the map from $n \rightarrow A$ is bijective. The first term on the right side inside the brackets corresponds to the supporting hyperplane tangent to $s(n)$ at $n = n^*$. Since $s(z)$ is concave, it follows that the difference is positive for any $n \neq n^*$. As $b(\cdot)$ is convex itself, it is easy to show that $b(\cdot)$ has a upper bound given as:

$$0 \leq b(n, n^*) \leq [A(n) - A(n^*)]^T (n - n^*) \tag{11}$$

where n is arbitrarily chosen. Therefore we have shown that $b(\cdot)$ is qualified as a storage function for a dissipative system.

$$\frac{\partial b}{\partial t} = (A - A^*)^T \frac{\partial (n - n^*)}{\partial t} = \overline{A}^T \frac{\partial \overline{n}}{\partial t} \tag{12}$$

$$\overline{A} = \frac{\mu - \mu^*}{T} \tag{13}$$

$$\overline{n} = n - n^* \tag{14}$$

where overbar stands for deviations with respect to the reference.

Substitute (9) into (12) we obtain:

$$\frac{\partial b}{\partial t} = \overline{A}^T (-G \frac{\partial \overline{n}}{\partial x} + \overline{B} - \overline{D} + \frac{\overline{Q}\overline{n}}{V}) \tag{15}$$

Assume that no agglomeration and breakage phenomena take place in the reactor such that $\overline{B}$ and $\overline{D}$ terms can be set to zero.

As we are interested in the overall stability, we integrate the storage function b over the whole size intervals$[x_1, x_n]$ such that:

$$\int_{x_1}^{x_n} \frac{\partial b}{\partial t} dx = \int_{x_1}^{x_n} \overline{A}^T (-G \frac{\partial \overline{n}}{\partial x} + \frac{\overline{Q}\overline{n}}{V}) dx \tag{16}$$

let

$$B = \int_{x_1}^{x_n} b dx \tag{17}$$

$$\begin{aligned} \frac{dB}{dt} &= -G \overline{A(n(x))}^T \overline{n(x)}|_{x_1}^{x_n} + \int_{x_1}^{x_n} \frac{\overline{QA}^T \overline{n}}{V} dx \\ &= -G \overline{A(n(x_n))}^T \overline{n(x_n)} + G \overline{A(n(x_1))}^T \overline{n(x_1)} \\ &+ \int_{x_1}^{x_n} \frac{\overline{QA}^T \overline{n}}{V} dx \end{aligned} \tag{18}$$

Here G is always positive,$\overline{A}$ and $\overline{n}$ have the same sign such that the product of them always nonnegative. Therefore, Inventory control is required to be implemented on the boundary condition which is the particle inlet and outlet flow rate to enforce that $\frac{dB}{dt} < 0$ so that the population balance converges to the stationary point.

4. APPLICATION

4.1 Process Description

Silicon based solar cells are expected to play an important role in meeting future energy demand and new processes have been introduced to increase production rate and reduce cost. Thermal decomposition of silane (SiH_4) in a fluidized bed reactor as shown in Figure 3 is one new technology for solar grade silicon production. The fluidized bed reactor considered in this study is shown in Figure 1. The silicon seed particles are fed into the system at rate S. The silane and hydrogen gas (reactants) are fed from the bottom of the reactor. The reactants are fed at the rate of F_{in} to maintain the fluidization and control the residence time to ensure maximum conversion. The feed consists of silane diluted by hydrogen or argon which acts as the fluidization gas. Silicon particles grow in the reactor due to the deposition of silicon and are removed from the bottom (product) at rate P. Some of the fine silicon particles (powders) escape the reactor along with hydrogen (F_{out}) at the top exit. The powder production should be minimized to maximize yield. The reactor is heated

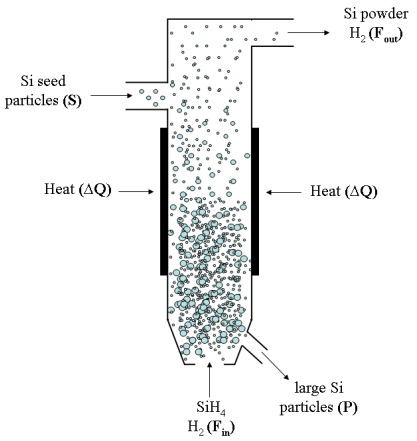

Fig. 3. Fluidized bed reactor

at specified locations as shown in the figure to heat the reactants to the reaction temperature. Product particles are withdrawn at a rate such that the solid particle hold-up is controlled. Additionally, seed particles are added in such a way that the desired product size distribution is achieved. Thus, the size distribution of the silicon particles at the exit (product) can be controlled by manipulating the reactant flow rate F_{in}, product flow rate P and silicon seed rate S.

4.2 Multi-Scale Modeling

The reaction between silane and hydrogen is almost instantaneous and the flow and mixing regimes reach steady state within few seconds of operation. However, based on the inlet conditions, temperature reaches steady state only after few minutes. These systems level phenomena are captured well by solving the mass, momentum and energy balance equations simultaneously using a two-dimensional heterogeneous model. On the other hand, the micro-level deposition of silicon particles produced in gas phase on to the silicon seed particles is captured by CVD equations. The particle size distribution and the dynamics of the particle growth are represented through particle mass and number balances. This particular dynamics is slow (in the order of hours). Thus, in modeling such a complex system which is characterized by different space and time scales, multi-scale approach is employed as illustrated in this section.
The multi-scale modeling strategy consists of three steps:

Define framework to integrate submodels
In this work, we follow multi-domain framework **?**) where the micro-scale and macro-scale parts occupy adjacent, non-overlapping parts of the system domain S. Interface regions exist between domains where both models apply. Micro-scale models define particle nucleation growth and agglomeration which determine the size distribution of particles in the reactor. The macro-scale model represents homogenous reaction in the gas phase. The micro-scale model is represented by a population balance module while the macro-scale model is represented by a fluid dynamics module. Heterogeneous reaction takes place in the gas phase. The interface between them is developed by Chemical Vapor Decomposition which describes particle growth process caused by depositing long chain silicon polymers and scavenging powder produced through homoge-

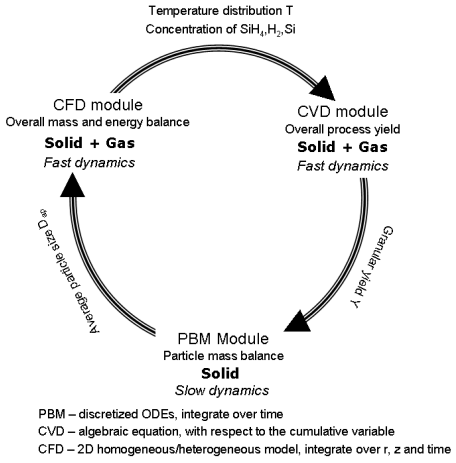

Fig. 4. Multi-scale Modeling scheme

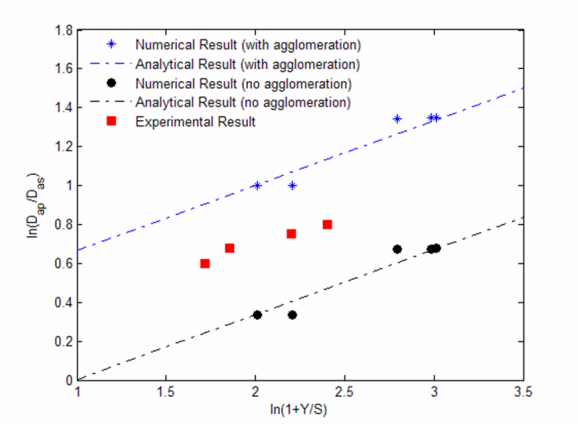

Fig. 5. Model Validation.

neous reaction in the gas phase.

Define length scales in each models
Different scales are identified based on the nature of the process and the characteristic length and time scales.
1. **Particle scale:** The important phenomena are the growth of each particle due to the reaction taking place.
2. **Granule scale:** Powder formation, granule nucleation, consolidation and coalescence are the main processes at this level.
3. **Vessel scale:** Temperature, concentration and velocity profile of both the gas and solid phase are crucial at this level.

The hydrodynamics are modeled using CFD, which provides a basis for a simplified reactor flow model as illustrated in Figure 2. The kinetic terms and the reactor temperature and concentrations can be expressed as functions of reactor dimensions, void volume and time in the CFD module. Reactor temperature and concentration from the CFD module provide inputs to the CVD module. The CVD module calculates the overall process yield which provides an input to the population balance module. The average particle diameter is then calculated by population balance module and imported into the CFD module for further calculation. The population balance module and CVD module are solved using MATLAB and the fluid dynamic module is solved using the Multi-physics modeling software COMSOL. The interlink between COMSOL and MATLAB is exploited well to integrate the different modules in a single computational platform. The details about the multi-scale model is presented in Balaji et al. (2009).

4.3 Multi-scale Modeling Results

The results obtained by coupling all the mechanisms based on the multi-scale approach (Figure 4) is discussed in this section. In Figure 5, we compare the numerical results with the analytical and the experimental results. As per the experiments conducted at REC Inc., the agglomeration of particles within the system does exist and the proposed model does capture the agglomeration phenomenon. The analytical expressions are obtained from White (2007). The relationship between process flow rates and average particle size is given by

$$1 + \frac{W}{S} = \frac{n_p}{n_s}\left(\frac{D_{ap}}{D_{as}}\right)^3 \tag{19}$$

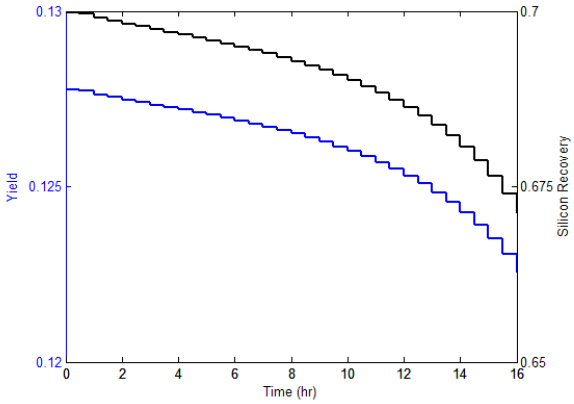

Fig. 6. Total yield and the percentage recovery of silicon.

where W is the product flow rate and S is the seed flow rate. D_{ap} is the average particle diameter of product and D_{as} is the average particle diameter of seed. n_p is the number of particles withdrawn and n_s is the number of particles added.

$\ln(1 + P/S) = 3\ln(D_{ap}/D_{as})$, then it is likely that $n_p/n_s = 1$, which means no nucleation, agglomeration, or breakage is present. If data indicate that $n_p/n_s < 1$, particle agglomeration takes place in the reactor.Here P is the product withdraw flow rate and S is the seed inlet flow rate. D_{ap} is the average particle diameter of product and D_{as} is the average particle diameter of seed. The detail of derivation process is described explicitly in White et al. (2006) From the results obtained, the numerical results agree satisfactorily with the analytical and experimental results and hence the proposed multi-scale model can be used for further studies.

Figure 6 shows the total yield of silicon obtained from the reactor and the corresponding recovery. In the model equations, the amount of silicon particles to be removed from the system to maintain the void fraction and the extent of reaction in the reactor is unknown. Hence, the simulations are carried out at unsteady state since the product withdrawal rate is smaller than the seed flow rate. This study shows the effect of void fraction and the mass hold up on the average diameter of the silicon particles. From this Figure, we can see void fraction

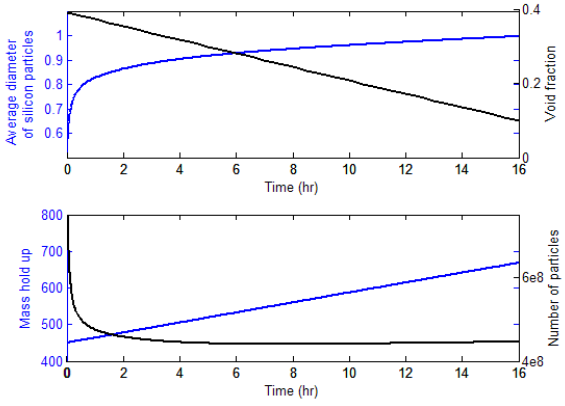

Fig. 7. Effect of mass hold up or the product flow rate.

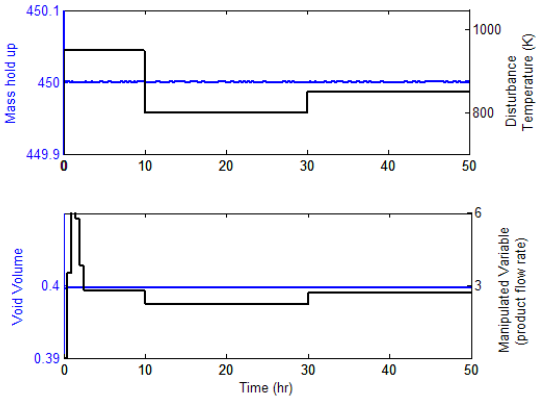

Fig. 8. Closed loop simulation results.

decreases as silicon mass hold up is increasing such that yield is decreasing along reaction taking place. Figure 7 shows that the as mass hold up increases, the void fraction slowly decreases leading to a decrease in the overall yield and recovery (Figure 6). However, there is an increase in the average diameter of the silicon particles. Thus, the mass hold up and in turn the product flow rate should be maintained in such a way that the desired particle size can be obtained.

4.4 Inventory Control

In this section, we apply the inventory control technique to a previously validated discrete population balance model for silicon production in a fluidized bed reactor. Simplifying the size interval mass balance of the model produces

$$\frac{dM_i}{dt} = g_i + \sum_{\gamma} q_{i,\gamma}, \tag{20}$$

where g_i represents production and internal flow terms (between size intervals), and q_i represents external particle flow terms. Summing (20) over specified size intervals allows definition of process inventories as defined in Farschman et al. (1998). The inventory balance equation then has the general form

$$\frac{dM_k}{dt} = \sum_{i \in \Omega_k} \left(g_i + \sum_{\gamma} q_{i,\gamma} \right), \tag{21}$$

where Ω_k represents the set of specified size intervals. These intervals may, for example, represent the amount of feed particles or the total mass of the system. In a continuous CVD process such as fluidized bed production of silicon, particles continuously increase in size. To operate at relatively constant conditions, it is necessary to maintain a constant particle hold-up and size distribution. The total hold up of particles can be held constant by withdrawing product particles at a rate equivalent to that of particle addition and growth. The particle size distribution can be held constant by regenerating the system with seed particles. To mimic this operation, we assume seed particles correspond to the smallest sized intervals, $i = 1, ..., I_s$, where I_s is the largest seed size interval. This results in the definition of seed hold-up

$$M_{\text{seed}} = \sum_{i=1}^{I_s} M_i.$$

The total mass is simply the sum over all intervals so this gives

$$M_{\text{total}} = \sum_{i=1}^{N} M_i.$$

To ensure the process inventories are kept at a desired level (M^*), we choose control parameters to obtain

$$\frac{dM_k}{dt} = -C \left(\sum_{i \in \Omega_k} M_i - M^* \right). \tag{22}$$

where C is an arbitrary strictly input passive operator. The inventory balance of (21) then requires us to choose controls so that

$$\sum_{i} q_i = - \sum_{i \in \Omega_k} g_i - K \left(\sum_{i \in \Omega_k} M_i - M^* \right). \tag{23}$$

Here, the external particle flow rates, q_i are manipulated input that cause the system to track given set points, M^*. This control law is guaranteed to be stable and obtain convergence of the particle size distribution if the zero dynamics are stable.

To briefly illustrate this control technique, we use the silicon example. Equation (23) allows us to design flow rates to achieve the required control of particle hold-up. The summation shown in (21) is performed over all size intervals (1 to N) to derive the product flow rate (P) required to maintain a constant hold-up in the reactor (M^*_{total}).

$$P = - \sum_{i=1}^{N} g_i - K_t \left(\sum_{i=1}^{N} M_i - M^*_{\text{total}} \right) \tag{24}$$

Here we used proportional control to illustrate. Another summation of (21) is performed over the seed size intervals, 1 to I_s, to determine the seed flow rate (S) required to maintain a constant mass of seed particles in the reactor (M^*_{seed}).

$$S = - \sum_{i=1}^{I_s} g_i - K_s \left(\sum_{i=1}^{I_s} M_i - M^*_{\text{seed}} \right) \tag{25}$$

Simulation of controlling the total and seed particle hold-up is shown in Figures 9 and 10. The hold-up of particles in the system is shown in Figure 9. The product and seed flow rates required to achieve the control are also shown. The first steady state (SS1) represents operation when $M^*_{\text{total}} = 75$ and

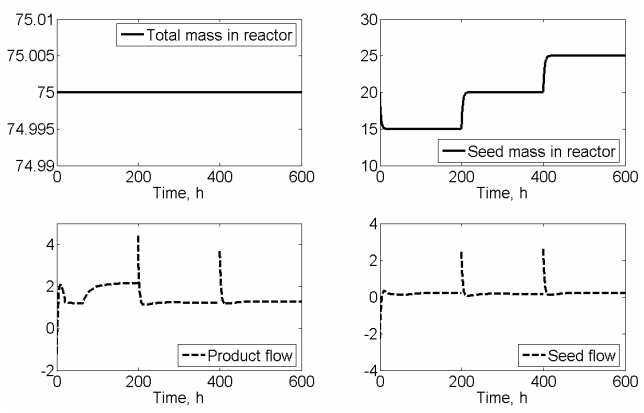

Fig. 9. Control total and seed hold up in CVD system

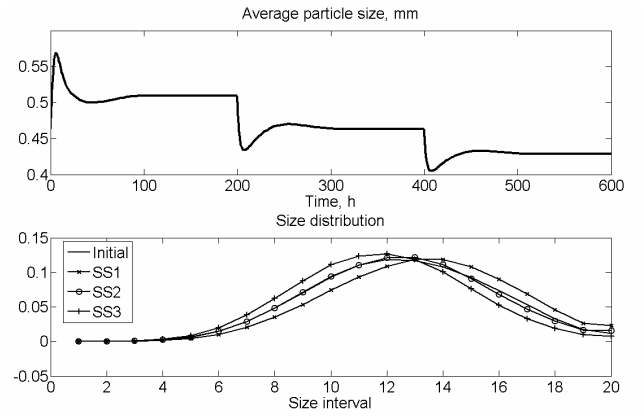

Fig. 10. Particle size achieved during seed control of CVD

$M^*_{\text{seed}} = 15$. The subsequent steady states are achieved when M^*_{seed} is increased to 20 and 25.

The average particle size and size distribution achieved during each steady state are shown in Figure 10. This simulation shows we can control the average product size as well as the product distribution. As the hold up of seed particles increases relative to the total hold up, the average size decreases. The interval representation of the size distribution supports this result. In this simulation, we assumed that the largest seed size interval, I_s, was interval 10 out of 20 and that the distribution of seed particles flowing into the system was constant.

5. CONCLUSION

In this paper, a comprehensive multi-scale model is implemented to describe the growth of silicon particles in a fluidized bed reactor. Population balance is used to represent the growth and aggregation of silicon particles. The simulations are successfully carried out and validated against the experimental and analytical results. Stability analysis is achieved in order to determine the control configuration for the particulate process. In addition an inventory based control is implemented to control the particle size distribution.

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