Modeling and control of size distribution for fluidized bed silane decomposition

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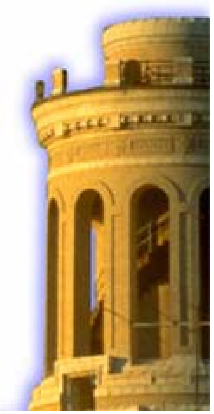
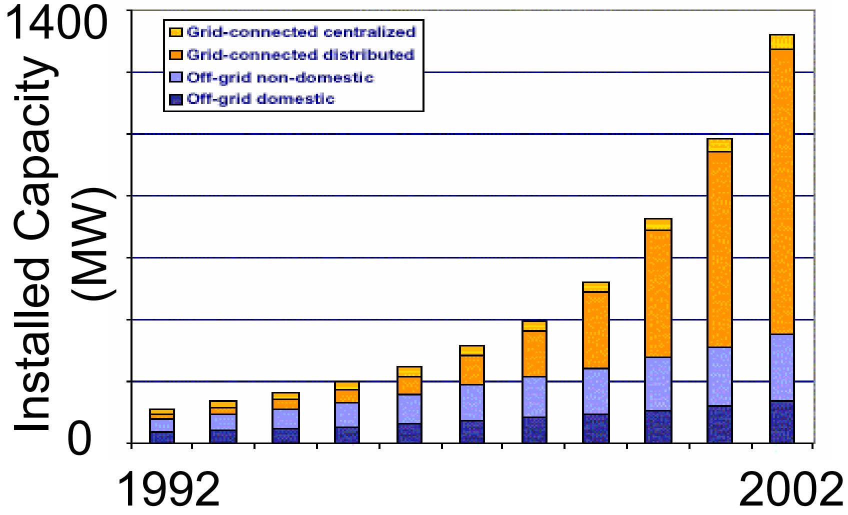


Photovoltaic Industry

Industry Growth

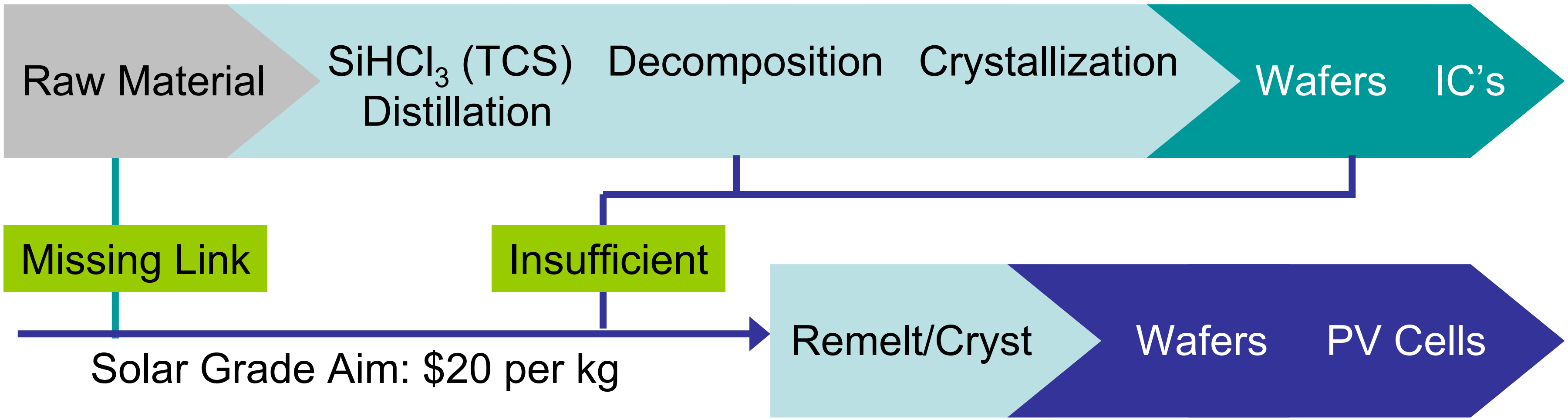
IEA-PVPS, '03



Solar Cell Production

Metallurgical Grade \$3-5 per kg

Electronic Grade \$40-60 per kg



Raw Material

$SiHCl_3$ (TCS) Distillation

Decomposition

Crystallization

Wafers

IC's

Missing Link

Insufficient

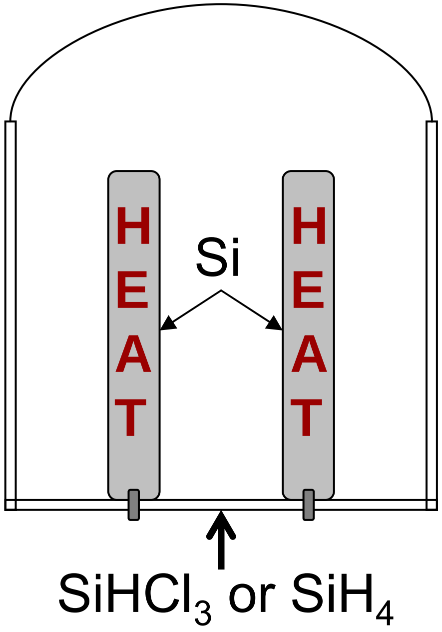
Remelt/Cryst

Wafers

PV Cells

Solar Grade Aim: \$20 per kg

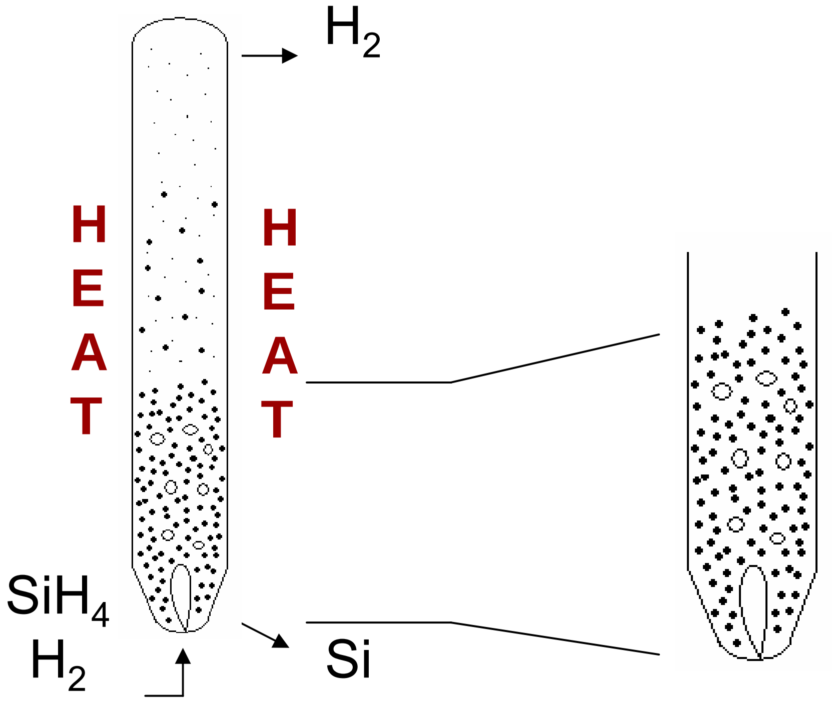
Silicon Production



Siemens Reactor

Batch Process

1100°C



Fluid Bed Reactor

Continuous Process

650°C

Large surface area

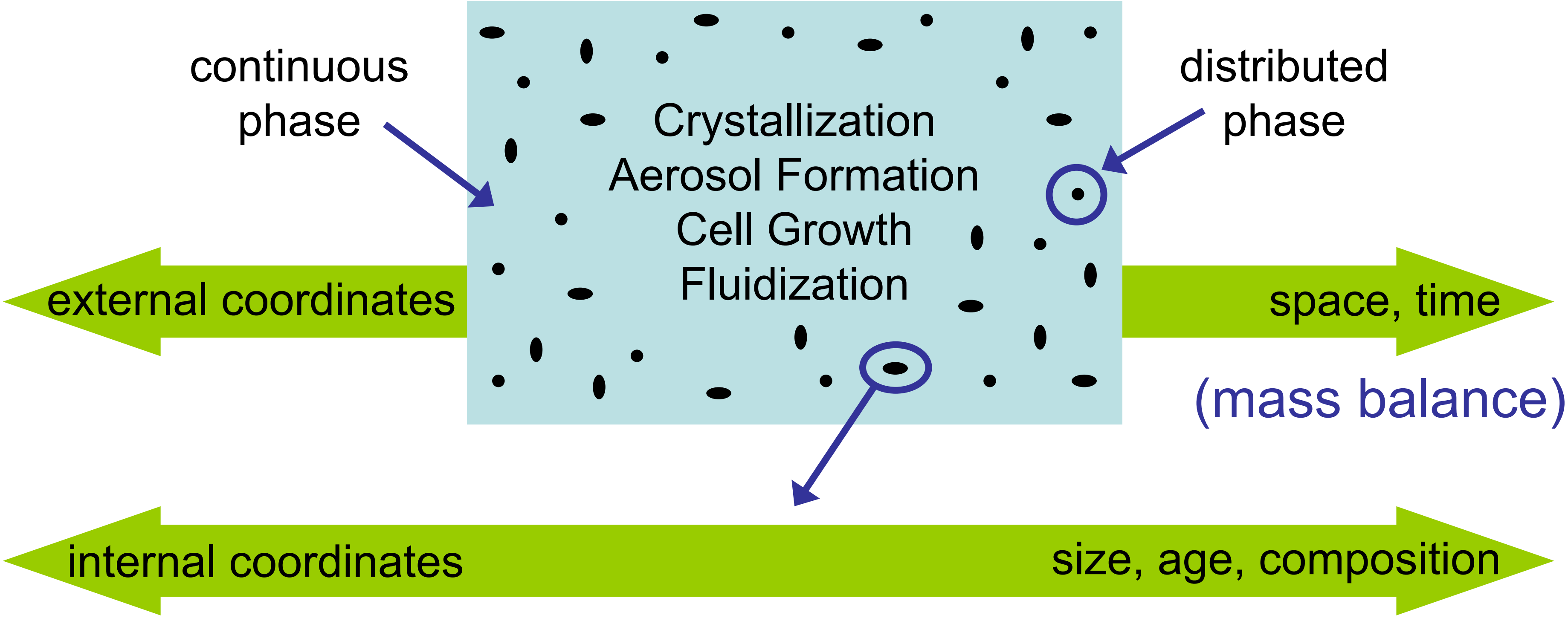
Dense Phase

SiH_4 Decomposition

Particle Growth

Size Distribution

Modeling Particulate Processes



continuous phase

Crystallization
Aerosol Formation
Cell Growth
Fluidization

distributed phase

external coordinates — space, time

(mass balance)

internal coordinates — size, age, composition

Population Balance

$$V\left(\frac{\partial \boxed{n}}{\partial t} + \underbrace{\nabla\cdot \boldsymbol{v_i} n}_{\text{internal flux}} + \underbrace{D - B}_{\text{death and birth terms}}\right) + n\frac{dV}{dt} = -\sum_k \underbrace{Q_k n_k}_{\text{external flow terms}}$$

density distribution

Overall Reaction and Gas Phase

Thermal Decomposition of Silane: $\mathrm{SiH}_{4(g)} \rightarrow \mathrm{Si}_{(s)} + 2\mathrm{H}_{2(g)}$

Ideal operation: dense zone is a CSTR

- Gas phase balance equations (SiH_4, H_2)

$$\frac{d(V_g \cdot C_{so})}{dt} = F_{in} \cdot C_{si} - F_{out} \cdot C_{so} - (R + loss_{hom}) \cdot V_g$$

$$\frac{d(V_g \cdot C_{ho})}{dt} = -F_{out} \cdot C_{ho} + 2 \cdot (R + loss_{hom}) \cdot V_g$$

- Reaction rate defined by Lai et al. (1986)

$$R_{het} = 2.79 \times 10^8 exp(-19530/T)C_s$$

$$R_{hom} = 2 \times 10^{13} exp(-26000/T)C_s$$

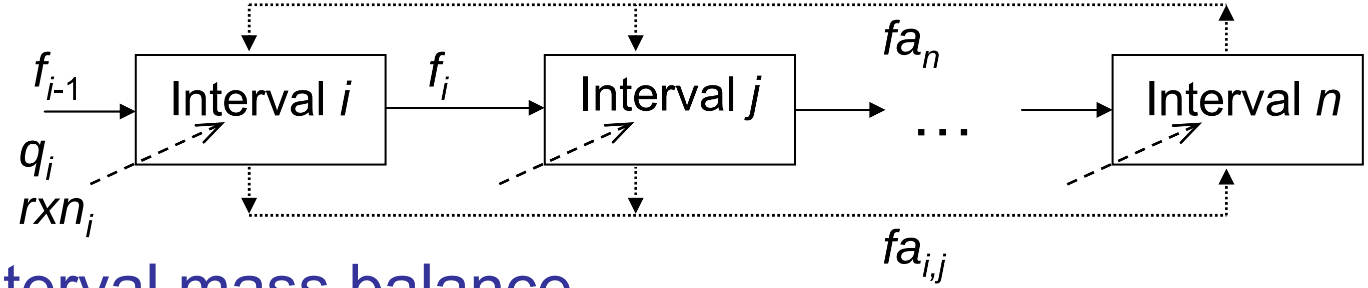
- Accounting for loss through entrainment → Total reaction:

$$R = R_{het} + (1 - \eta) R_{hom}$$

η is fraction of product (powder) lost

Behavior along external coordinate axes: position and time

Solid Phase



Size interval mass balance

$$\frac{dM_i}{dt} = f_{i-1} + rxn_i - f_i + fa_i^{IN} - fa_i^{OUT} + \sum_i q_i$$

Assume

$$rxn_i = R \cdot \frac{A_i}{\sum_i A_i}$$

$$fa_{i,j} = k_{i,j} C_i C_j \Rightarrow$$

$$fa_i^{OUT} = \sum_j \left(fa_{i,j} \cdot \frac{m_i}{m_i + m_j} \right)$$

$$fa_i^{IN} = \sum_j \sum_k fa_{j,k}, \text{ for}$$

$$m_i^{LB} \leq (m_j + m_k) < m_{i+1}^{LB}$$

Derive f_i assuming continuous number:

$$N_i = \frac{M_i}{m_i} \Rightarrow \frac{dN_i}{dt} = \frac{1}{m_i} \cdot \frac{dM_i}{dt}$$

can obtain:

$$f_i = (rxn_i + aterm_i) \cdot \frac{m_{i+1}}{m_{i+1} - m_i}$$

$$aterm_i = fa_i^{IN} - m_i N_i$$

Behavior along internal coordinate axis: size

Solution Strategy for Discrete Model

Ordinary differential equations for mass in gas and solid phases

+

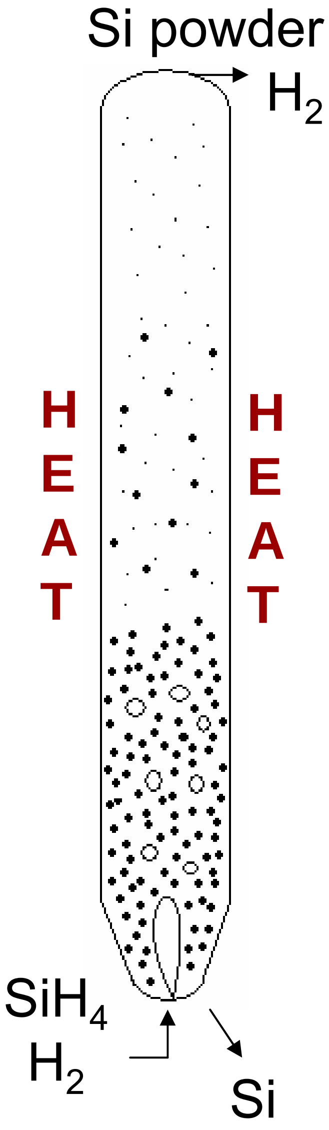
Algebraic constitutive equations

↓

DAE system solved by MATLAB's ode15s

Adjustable Parameters		*Example Values*
η	Fraction of powder lost	$0 - 1$
$k_{i,j}$	Aggregation proportionality constant	$0 - 10^{-8}$ (size dependent)

Experimental Data for Validation



Operation of pilot scale reactor

Load known initial distribution

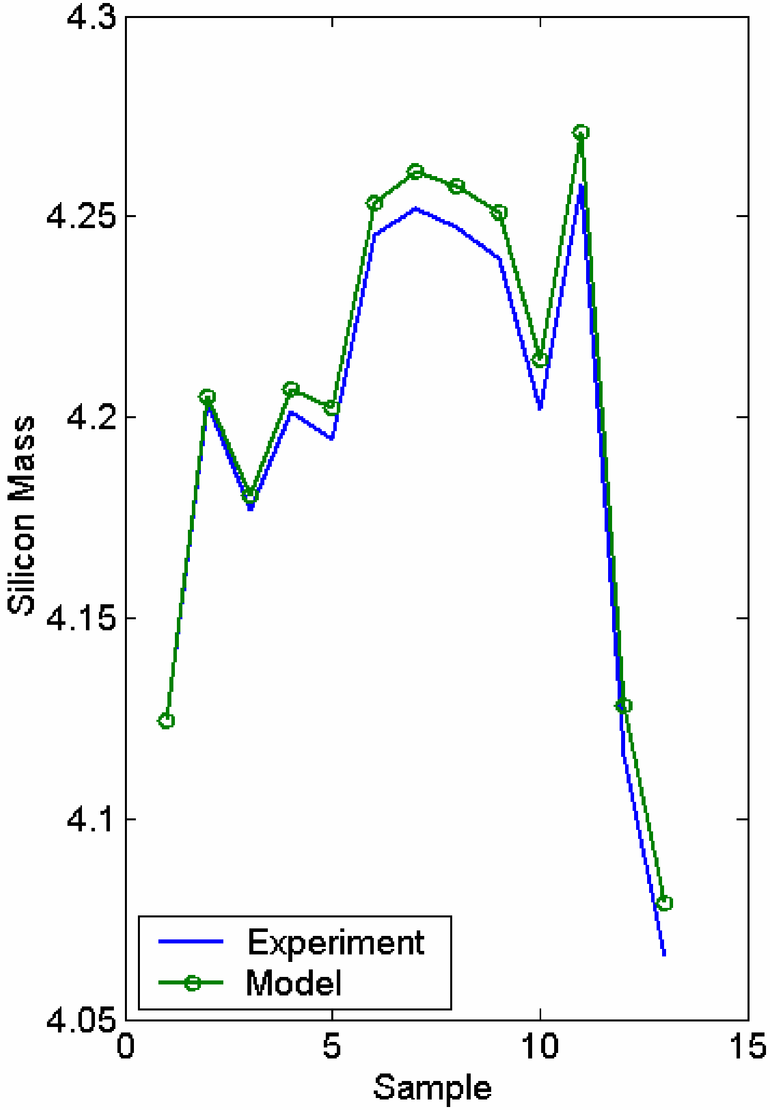
– sieves used to measure distribution

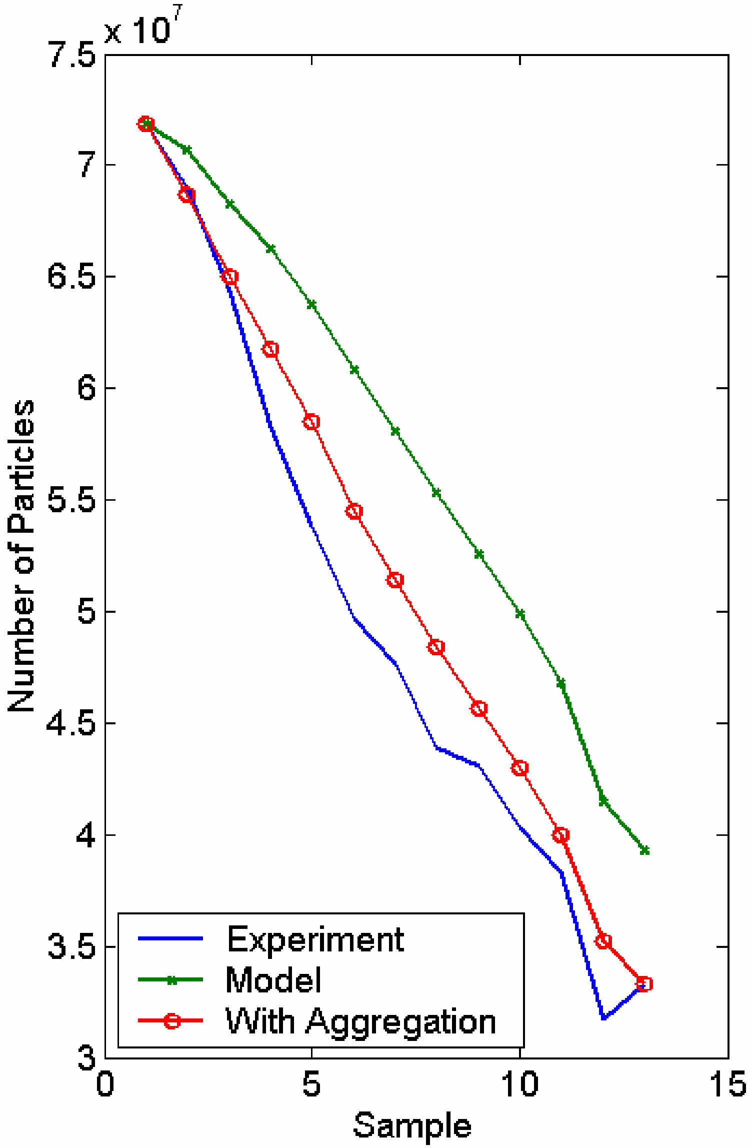
Continuous feed to fluidized bed

Withdraw samples regularly

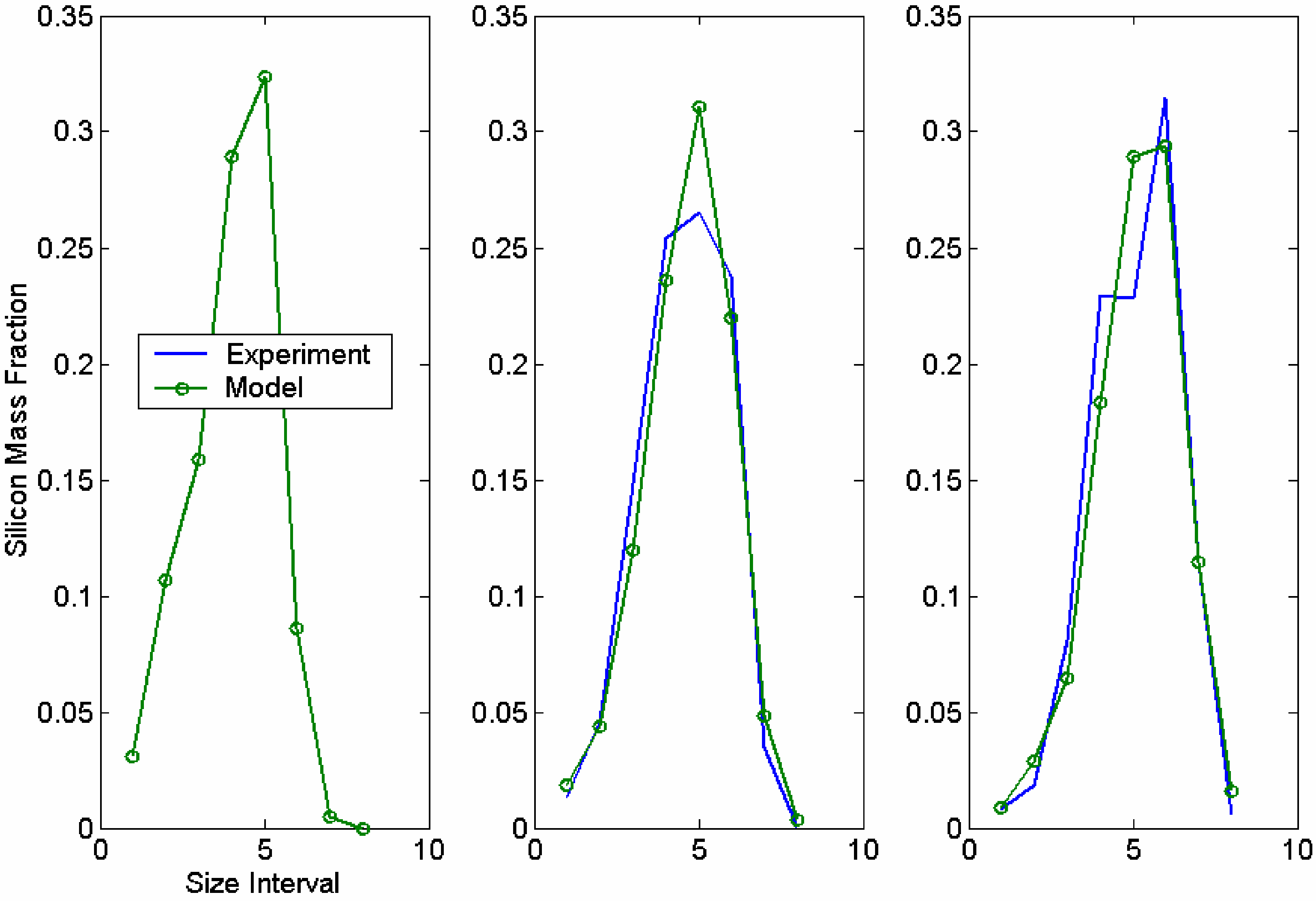
~8-12 samples per run

Silicon in Reactor



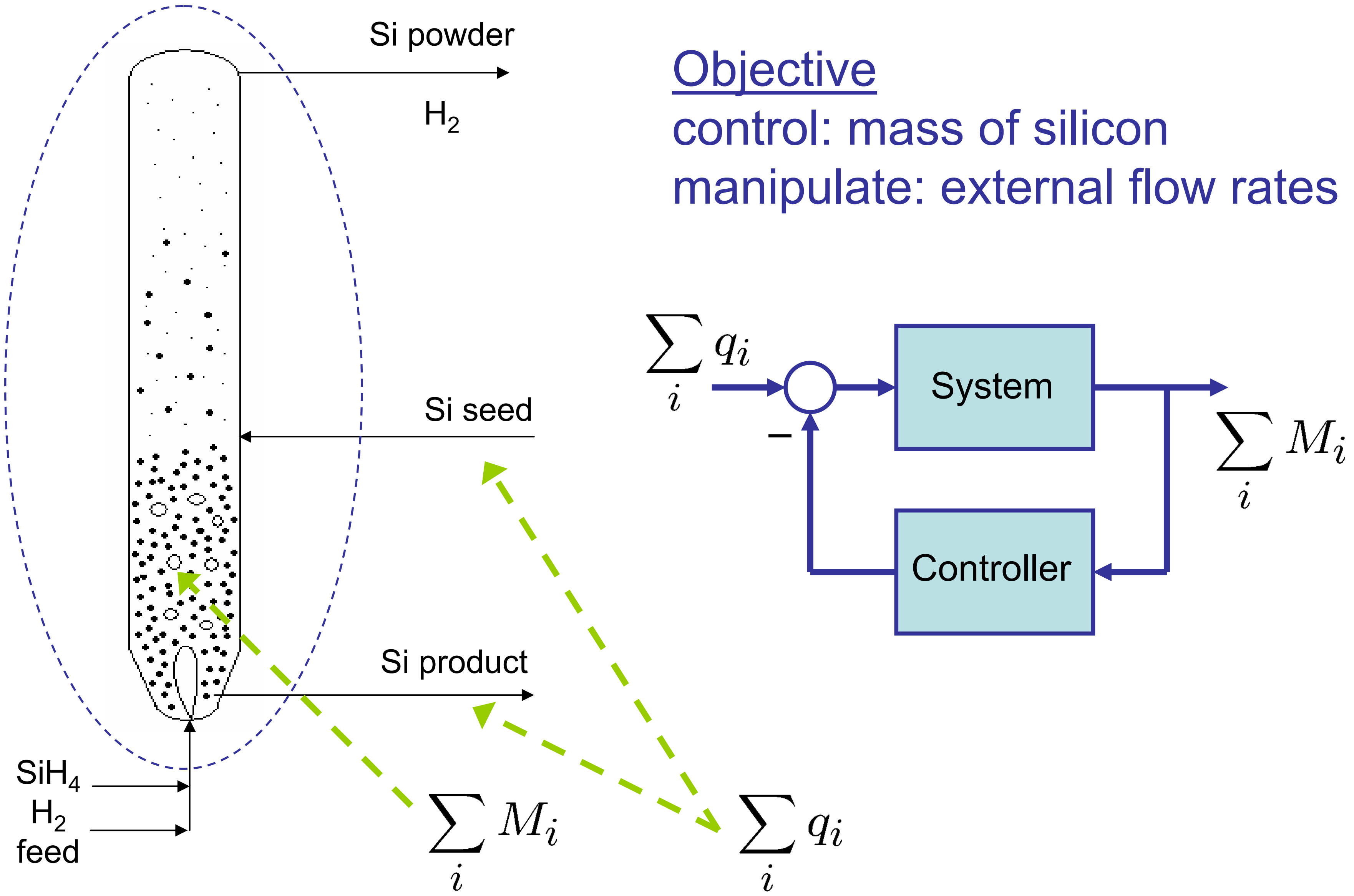


Particle Size Distribution



Controlling Continuous Operation

Objective
control: mass of silicon
manipulate: external flow rates



Inventory Control of Population Balance

$$\frac{dM_i}{dt} = \underbrace{f_{i-1} + rxn_i - f_i + fa_i^{IN} - fa_i^{OUT}}_{g_i} + \sum_i q_i$$

Apply inventory control to system:

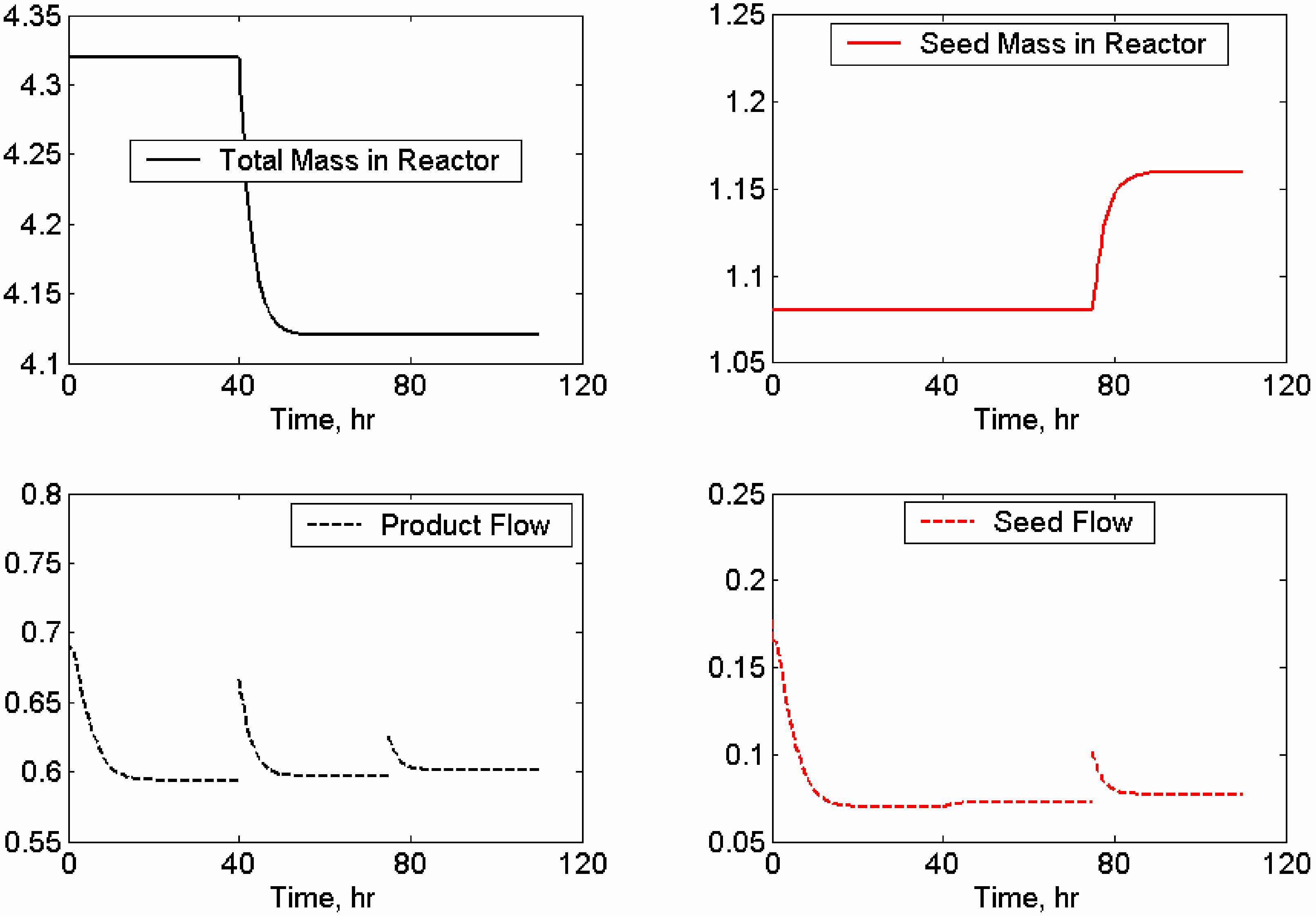
$$\sum_i \frac{dM_i}{dt} = \sum_i g_i + \sum_i q_i = -K\left(\sum_i M_i - M^*\right)$$

$$\Rightarrow \sum_i q_i = -\sum_i g_i - K\left(\sum_i M_i - M^*\right)$$

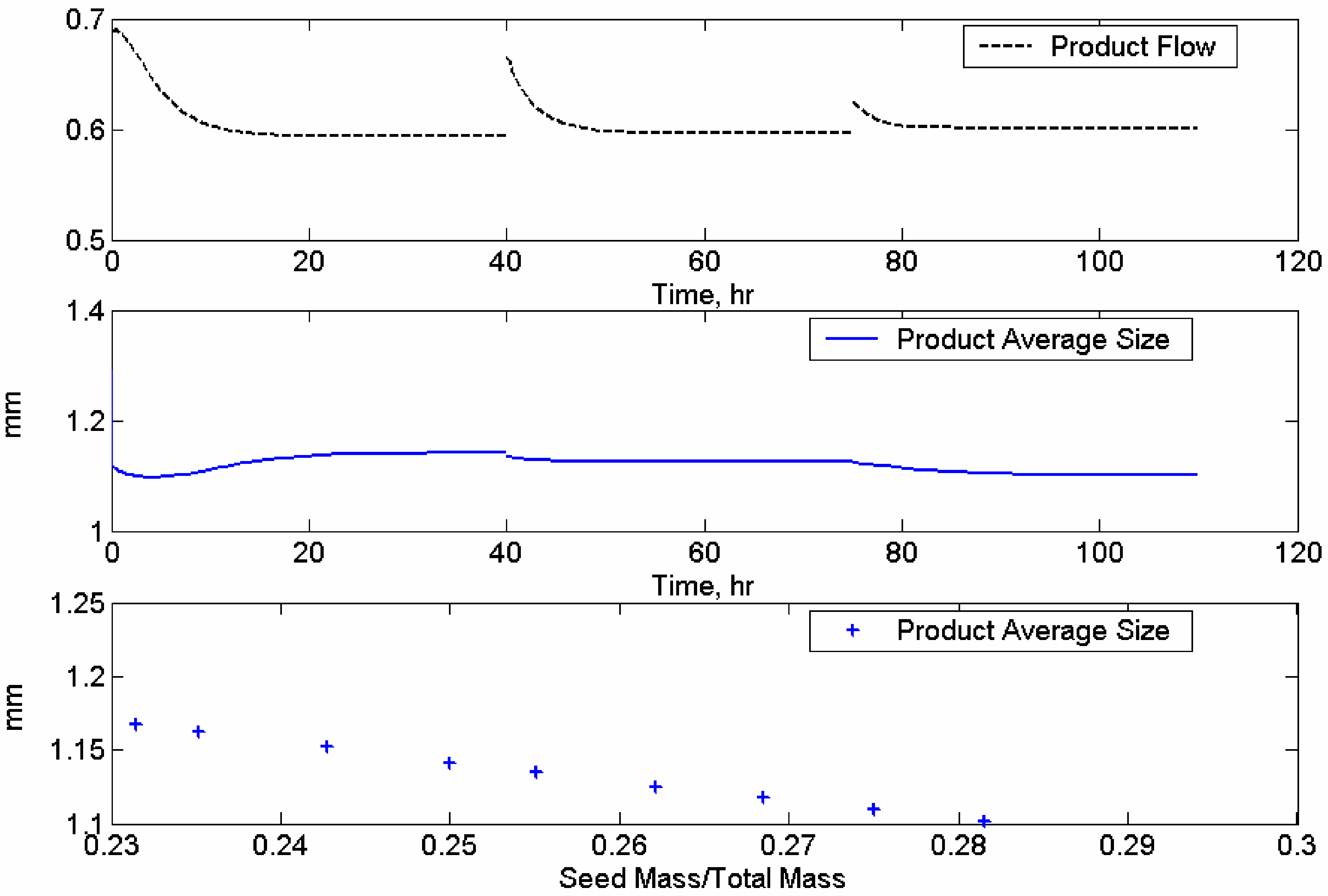
Constant mass in reactor: $product = -\sum_{i=1}^{N} g_i - K\left(\sum_{i=1}^{N} M_i - M^*\right)$

Constant seed mass: $seed = -\sum_{i=1}^{I_s} g_i - K\left(\sum_{i=1}^{I_s} M_i - M_s^*\right)$

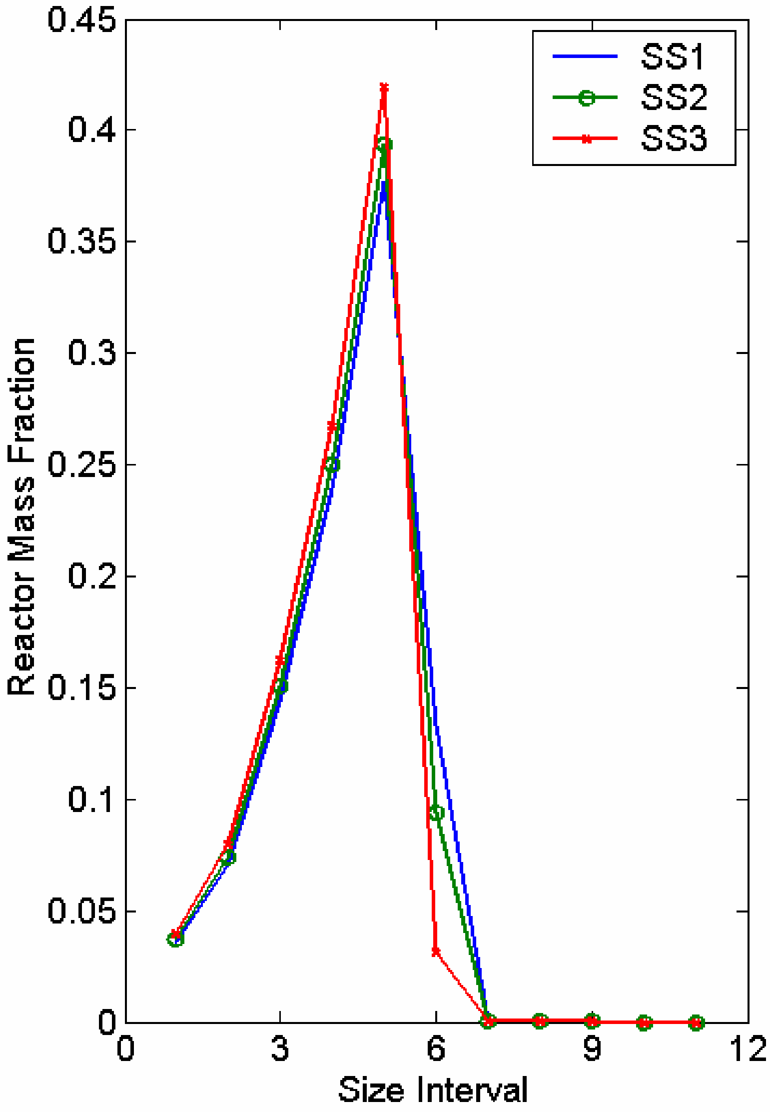
Response to Set Point Changes

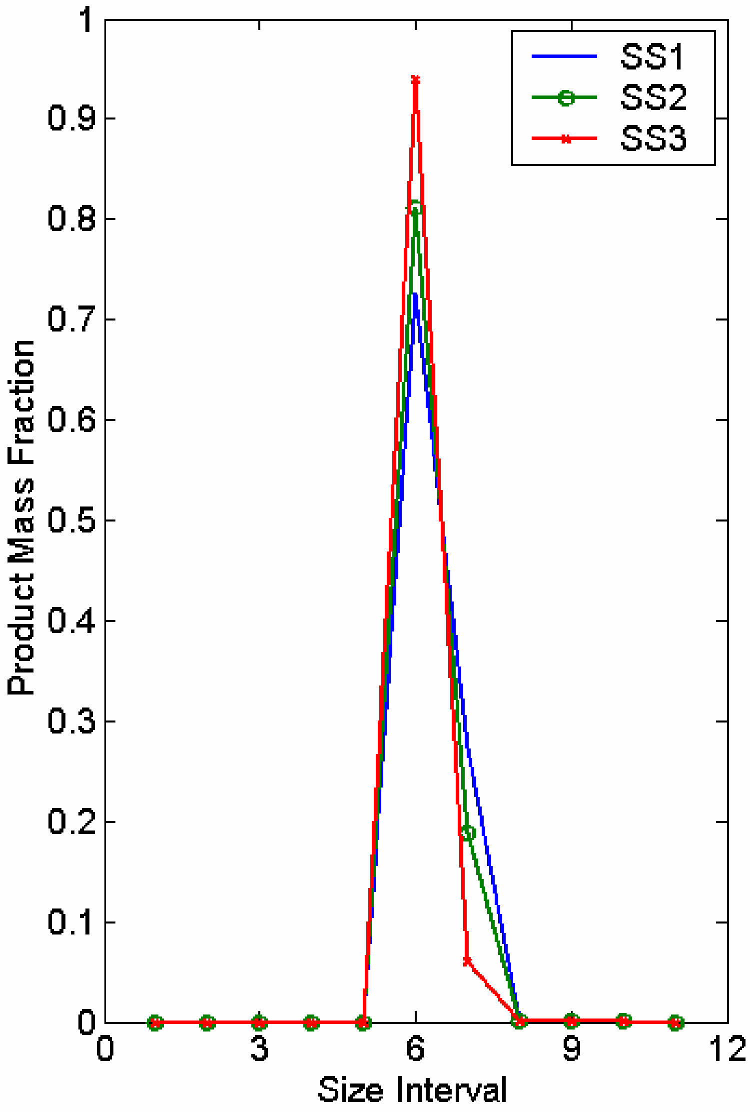


System Dynamics



Silicon Size Distribution





Summary

- Size interval mass balance predictions of particle distribution compare well with data
- Simulations of continuous operation and inventory control indicate that size is controllable
- Further investigate measurability and stability
- Application to other particulate processes or multi-scale modeling is significant

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