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Model predictive control of a CVD reactor for production of polysilicon rods

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Production of polysilicon plays a key role in the development of hi-tech and renewable energy industry. Massive production is obtained by chemical vapor deposition (CVD) in semi-batch reactors, traditionally called Siemens reactors, where silicon rods are grown. Following recent increase in market demand for polysilicon, a fine process control on industrial processes for improving production yield and reducing energy consumption is required. In this work, a technique for a real-time model-based predictive control applied to a laboratory-scale Siemens reactor is presented; a lumped model is used for describing the CVD process. Discussion is based on numerical results.

1. Introduction

Polysilicon, or polycrystalline silicon, is the most used raw material as semi-conductor for industrial manufacturing of photovoltaic cells and, after refinement, of electronic devices. In the last decade, polysilicon market demand has boosted (Pizzini and Bulk, 2009) ; consequently, a rising need for optimizing production plants has occurred. Well-run processes for massive production of polysilicon are based on chemical vapor deposition of silicon from a gaseous mixture of silanes. Deposition of solid silicon is traditionally obtained in reactors where rods made in high-purity silicon are used as seed; due to low vapor pressure of silanes, operating conditions are characterized by low pressure and high temperatures. Such reactors are commonly known as Siemens reactors, due to the first industrial application. Reduction of silanes to silicon occurs on the surface of rods; as this is thermodynamics and kinetics controlled process, high temperatures are required. Accordingly, seed rods are heated by Joule effect that is by a flowing electrical current, as this allows a fine temperature control. Rods are progressively grown-up until a critical diameter is reached, then the CVD process is shut-down. Due to diameter growth, rod surface raises; as a result, silicon deposition rate and temperature dispersion raise as well. For keeping rod surface at constant

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temperature, Joule effect must be increased; consequently, excessive temperatures can occur inside rods, with subsequent local melting. From an industrial process standpoint, the Siemens reactor is of semi-batch type. Gaseous precursors and reaction products are continuously fed and discharged, respectively; on the other hand, as rods reach their critical diameter the reactor is shut-down, and grown rods replaced by fresh ones. Nevertheless, from chemical reactor engineering standpoint, the reactor can be considered a continuous stirred system, as the growth process is longer than the replacement operation and equilibrium conditions set-up in vapor phase. Polysilicon CVD production is a high energy consuming process; fine control on purity of final product and on quantitative yield of semi-batch campaign is essential. Consequently, effects due to any operating disturbance or set-point trajectory transition occurring during the CVD process must be prevented by a fast-response process control. For such purpose, a nonlinear model predictive control (NMPC) technique can be effectively adopted. Numerical application of this technique to a pilot Siemens reactor is discussed.

2. Differential-algebraic model

A typical Siemens reactor is constituted of a bell-shaped vessel containing U-rods (see Fig 1), arranged in a symmetrical configuration. Silanes and reaction products are respectively fed and discharged from the bottom; H2 is used as carrier fluid. Vessel walls are usually kept in adiabatic conditions; the vessel is of flanged type so to be removed for replacement of rods. Operating conditions are high temperature (>500 °C) and low pressure; fluid-dynamics regime of gaseous streams is of laminar type.

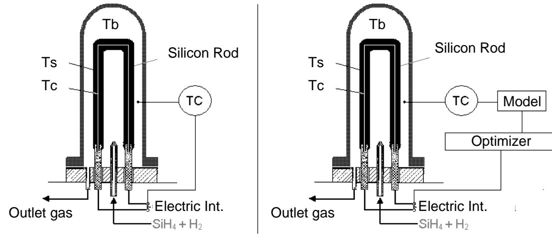

Figure 1: Conventional control and NMPC (right) arrangement for Siemens reactor

The deposition process occurs by both vapor-phase and surface reactions. Gaseous silanes undergo to vapor-phase thermal decomposition (Eq. 1); gaseous chemical species adsorb to and desorb from rods surface, while solid silicon is deposited (Eq. 1 and Eq. 2). Rods themselves work as a catalyst; the whole mechanism is typical of gas-solid catalytic reactions. Complete kinetic scheme is constituted of tens of reactions Weerts et al., 1998); however, for engineering mass and heat balances a lumped scheme can be used (Maso et al., 2000). Considering SiH_4 as the main precursor, the CVD process develops by following macro-reactions:

$$SiH_4 \rightarrow SiH_2 + H_2 \quad (1)$$

$$SiH_2 + \text{surface} \rightarrow Si_{(s)} + H_2 \quad (2)$$

$$SiH_4 + \text{surface} \rightarrow Si_{(s)} + 2H_2 \quad (3)$$

where "surface" term indicates that silanes are adsorbed on rods surface, which acts as catalyst. During normal operations, continuous stirred conditions can be assumed in the reactor; temperature and mass gradients are concentrated in a small vapor-phase zone surrounding the rods. This corresponds to the boundary layer typical of heterogeneous catalytic reactions. Along the production campaign, rods gradually grow under CVD, therefore active surface area, chemical kinetic rate, and electrical heating are variable. The system is intrinsically unsteady-state. The reactor is assumed perfectly stirred; thus, uniform temperature and chemical species concentration are considered in vapor-phase. As per boundary layer theory, mass and heat transport between vapor and solid-phase is expressed by means of effective transport coefficients. Resulting balance equations, where superscripts V and S stay for vapor-phase and rod surface respectively, are:

$$\frac{dC^V_{SiH_4}}{dt} = \frac{\dot{F}_{IN}}{V} x^{IN}_{SiH_4} - \frac{\dot{W}_{OUT}}{V} C^V_{SiH_4} - h_{m,SiH_4} \frac{A}{V}\left(C^V_{SiH_4} - C^S_{SiH_4}\right) - k_1 C^V_{SiH_4} \quad (4)$$

$$\frac{dC^S_{SiH_4}}{dt} = h_{m,SiH_4} \frac{A}{V}\left(C^V_{SiH_4} - C^S_{SiH_4}\right) - k_3 C^S_{SiH_4} \frac{A}{V} \quad (5)$$

$$\frac{dC^V_{SiH_2}}{dt} = \frac{\dot{F}_{IN}}{V} x^{IN}_{SiH_2} - \frac{\dot{W}_{OUT}}{V} C^V_{SiH_2} - h_{m,SiH_2} \frac{A}{V}\left(C^V_{SiH_2} - C^S_{SiH_2}\right) + k_1 C^V_{SiH_4} \quad (6)$$

$$\frac{dC^S_{SiH_2}}{dt} = h_{m,SiH_2} \frac{A}{V}\left(C^V_{SiH_2} - C^S_{SiH_2}\right) - k_2 C^S_{SiH_2} \frac{A}{V} \quad (7)$$

$$\frac{dC^V_{H_2}}{dt} = \frac{\dot{F}_{IN}}{V} x^{IN}_{H_2} - \frac{\dot{W}_{OUT}}{V} C^V_{H_2} - h_{m,H_2} \frac{A}{V}\left(C^V_{H_2} - C^S_{H_2}\right) + k_1 C^V_{SiH_4} \quad (8)$$

$$\frac{dC^S_{H_2}}{dt} = h_{m,H_2} \frac{A}{V}\left(C^V_{H_2} - C^S_{H_2}\right) + k_2 C^S_{SiH_2} \frac{A}{V} + 2k_3 C^S_{SiH_4} \frac{A}{V} \quad (9)$$

$$\frac{dC^S_{Si}}{dt} = k_2 C^S_{SiH_2} \frac{A}{V} + k_3 C^S_{SiH_4} \frac{A}{V} \quad (10)$$

$$\begin{aligned} \sum_{i=1}^{NC} C_i V \left(A_{Cp,i} + 2B_{Cp,i} T\right) \frac{dT}{dt} &= \dot{F}_{IN} \sum_{i=1}^{NC} x_i^{IN} \left(\tilde{H}_{f,i}\left(T_{rif}\right) + \int_{T_{rif}}^{T_{IN}} \tilde{C}p_i dt\right) + \\ &- \dot{W}_{OUT} \sum_{i=1}^{NC} C_i^V \left(\tilde{H}_{f,i}\left(T_{rif}\right) + \int_{T_{rif}}^{T} \tilde{C}p_i dt\right) + \sum_{j=1}^{NRV} r_i V \left(-\Delta\tilde{H}_{R,j}\left(T\right)\right) + \\ &+ \sum_{k=1}^{NRS} r_i A\left(-\Delta\tilde{H}_{R,k}\left(T_S\right)\right) + h_T A\left(T_S - T\right) + \sigma A\left(T_S^4 - T^4\right) \end{aligned} \quad (11)$$

with $\dot{F}_{IN}$ inlet molar flowrate [mol/s]; $\dot{W}_{OUT}$ outlet volumetric flowrate [cm^3/s]; x_i^{IN} inlet molar fraction; C_i^B vapor-phase concentration [mol/cm^3]; C_i^S concentration on rod surface [mol/cm^3]; V vapor-phase reaction volume [m^3]; A rods surface [m^2]; k_1 vapor-phase kinetics constant [1/s]; k_2 and k_3 surface kinetics constants [m/s]; $h_{m,i}$ mass transfer coefficient [m/s]; h_T heat transfer coefficient [cal/m^2/s/K]; σ Stefan-Boltzman constant [cal/m^2/s/K^4]; $NRS = 2$ number of surface reactions; $NRV = 1$ number of vapor-phase reactions; $A_{Cp,i}$ and $B_{Cp,i}$ coefficients for specific heat polynomial; $\Delta\tilde{H}_R$ molar reaction enthalpy [cal/mol]; $\tilde{H}_{f,i}$ formation enthalpy [cal/mol]; $\tilde{C}p_i$ specific heat

[cal/mol/K]; T and T_s vapor-phase and rod surface temperatures [K]. For this work, a laboratory-scale reactor was considered; geometrical and operating parameters used in the simulation study are reported in Table 1.

Table 1 – Geometrical and operating parameters

Parameter	Value	Units	Parameter	Value	Units
V	480.	cm^3	$\Delta H°_{f,H2}$	0.	cal/mol
A	120.	cm^2	$\Delta H°_{f,SiH4}$	8100.	cal/mol
V	10.	cm/s	$\Delta H°_{f,SiH2}$	64590.	cal/mol
x_{H2}^{IN}	0.999	%	$\Delta H°_{f,Si}$	0.	cal/mol
x_{SiH4}^{IN}	0.001	%	$S°_{f,H2}$	31.2	cal/mol/K
x_{SiH2}^{IN}	0.000	%	$S°_{f,SiH4}$	48.8	cal/mol/K
T^{IN}	373.15	K	$S°_{f,SiH2}$	49.5	cal/mol/K
T_S	1100.	K	$S°_{f,Si}$	4.5	cal/mol/K
$Nu_M = Nu_T$	3.66	-	ρ_{Si}	0.19	mol/cm^3

Parameter	Value	Units
D_{H2}	$1.276 \cdot (T[K] / 300)^{1.68}$	cm^2/s
D_{SiH4}	$0.58 \cdot (T[K] / 300)^{1.8}$	cm^2/s
D_{SiH2}	$0.59 \cdot (T[K] / 300)^{1.8}$	cm^2/s
Cp_{H2}	$6.5 + 0.0016 \cdot T[K]$	cal/mol/K
Cp_{SiH4}	$4.9 + 0.018 \cdot T[K]$	cal/mol/K
Cp_{SiH2}	$6. + 0.001 \cdot T[K]$	cal/mol/K
Cp_{Si}	5.4	cal/mol/K
k_V	$2.7 \cdot 10^{14} \cdot \exp(-57000/R/T[K])$	1/s
$k_{S,1}$; $k_{S,2}$	$1.9 \cdot 10^4$; 5.4	cm/s
k_T	$1.3 \cdot 10^{-4} + 3.45 \cdot 10^{-4} \cdot T[K]/300 - 2.9 \cdot 10^{-5} \cdot (T[K]/300)^2$	cal/cm/s/K

High surface temperature is obtained by Joule effect. Significant electrical energy is fluxed along rods; heat is generated in the solid bulk and is dissipated on the surface due to convective exchange, radiation and surface reactions. Due to thermal resistance, a parabolic thermal profile takes place in the rod, which maximum temperature is located in the center (Bird et al., 2007); larger the rod diameter, larger the difference in temperature between center and surface. When rod temperature approaches melting point, rod gets structurally weak and crystalline structure is affected also; accordingly, the silicon rod has reached its critical size. Heat balance on the rod surface reads:

$$\left(mCp_{Si}\right)\frac{dT_S}{dt} = -h_T A\left(T_S - T\right) - \sigma A\left(T_S^4 - T^4\right) + \frac{\lambda_T}{R}\left(T_C - T_S\right) \quad (12)$$

$$T_C = T_S + \left(\frac{I^2}{\lambda_e}\right)\frac{R^2}{4\lambda_T} \quad (13)$$

where m is the molar mass of rod [mol], T_c is the temperature in the center of rod [K], λ_e and λ_T are respectively the electrical [1/Ω/m] and thermal [cal/m/s/K] conductivity of rod, I is the current intensity [A]. Eq. (13) includes Joule effect. Radius and surface of the rod, R and A , change under chemical vapor deposition; for a rod of initial surface A_0 and length L, quantities are linked by:

$$A = A_0 + 2\pi LR\left(t\right) \quad (14)$$

When CVD process is conducted at constant current intensity I, we noted in simulations that rod, surface, and vapor-phase temperatures significantly decrease as the heat

generated by Joule effect is progressively distributed on a wider rod surface. Consequently, it is necessary to regulate *I* for keeping the desired *Tb*.

3. Essentials of nonlinear model predictive control

Need for a fine process control arises from high production costs of polysilicon. The Siemens process is very energy demanding, consequently an increase in process performances and a fast control on operating disturbances lead to a significant income. Operating disturbances may engender a prolongation of scheduled campaign, a reduced yield, a considerable recycle of unreacted precursors and an improper rod temperature. This last parameter is the most critical as it determines surface kinetics and structural soundness of the rod. Operating disturbances normally happen during start-up and shut-down phase; disturbances may occur when external items, like exchangers or pumps, undergo transients. Potential, critical sources of trouble are the electrical circuit and the external perturbation caused by electro-magnetic effects of neighbor reactors. Practically, during operations variations may occur on energy supply, magnetic field, reactants concentration, etc. Since it is not possible to directly measure the temperature of the rod, *Tp* or *Tc*, traditional (non-model-based) control is particularly rough. As high temperatures are detected on rods surface, without regard to causes, reactor is shut-down and rods are checked. However, surface hot spots may be due to operating disturbances rather than critical diameter of rods; therefore, shut-down may realize to be an additional cost. In order to prevent effects due to disturbances and to perform a control of rods temperature, it is necessary to adopt an advanced process control technique. This can be accomplished by means of a nonlinear model predictive control (NMPC) of the reactor. The control technique consists in solving repeatedly a nonlinear programming problem (NLP) where optimization constraints are differential and algebraic equations of the convolution model (Rawling et al., 2000; Done et al; 2010):

$$\min_{u(k)\ldots\ldots u(k+h_c-1)} \left\{ \sum_{j=k+1}^{k+h_p} \omega_j \left(y_j - y_j^{set}\right)^2 + \sum_{l=k}^{k+h_p-1} \omega_{1,l} \left(u_l - u_l^{tar}\right)^2 + \sum_{l=k}^{k+h_p-1} \omega_{2,l} \left(u_l^i - u_l^{i-1}\right)^2 \right\}$$
$$\text{s.t.:} \begin{cases} y_j^{\min} \le y_j \le y_j^{\max};\; u_l^{\min} \le u_l \le u_l^{\max};\; \Delta u_l^{i,\min} \le \Delta u_l^i = u_l^i - u_l^{i-1} \le \Delta u_l^{i,\max} \\ \mathit{Convolution\ Model} \end{cases} \quad (15)$$

where $y_j - y_j^{set}$ is the deviation between the $j-th$ controlled variable and its setpoint; $u_l - u_l^{tar}$ is the deviation between the $l-th$ manipulated variable and its target; $u_l^i - u_l^{i-1}$ is the incremental variation between $i-th$ and $(i-1)-th$ time intervals of the $l-th$ manipulated variable; coefficients of diagonal semi-positive definite matrices $\boldsymbol{\omega}$ are the weighting factors; HP is the prediction horizon; HC is the control horizon; min and max superscripts are lower and upper bounds for manipulated and controlled variables.

4. Numerical results

Figure 1 shows two control schemes for Siemens reactor; the traditional one consists of a control loop where bulk temperature *Tb* is regulated by acting on electrical intensity *I*. In the model, both conventional and predictive control techniques are implemented for keeping *Tb* = 1066 K (to have *Tp* = 1100 K) by acting on *I*. Tuning parameters for

conventional control are: proportional gain $k_C=10^{-5}$ and time integral; NMPC is set using HP = 600 s and HC = 60 s. Simulation results are shown in Figure 2; use of conventional control leads to long oscillations and dangerous system instabilities, where peaks of temperature approach silicon melting temperature. Conversely, NMPC is fast to dampen any perturbation and able to drive start-up along an optimal trajectory.

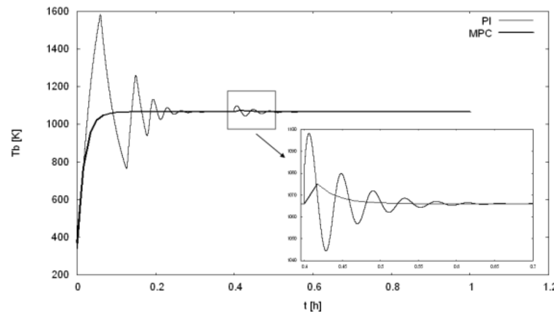

Figure 2: Comparison of conventional control and NMPC for servo (start-up phase of Siemens reactor) and regulation (feed flow perturbation) problems

5. Conclusions

An advanced process control technique for the Siemens reactor has been discussed according to a numerical approach. Process control is applied on silicon rod temperature, which represents the most critical operating parameter, by acting on electrical current supplied to rod. The simulation study is based on a lumped chemical kinetics scheme and reactor model; continuous and perfectly stirred conditions are assumed. Heat and mass balances have been given both for vapor-phase and solid rods.
Numerical results show that benefits can be obtained by applying a nonlinear model predictive control instead of a conventional control. Instabilities in controlling rod temperature are eliminated and false shutdowns may be avoided. As polysilicon production by Siemens process is highly energy consuming, application of advanced control techniques to such reactors is expected to be extensively developed in the future.

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