

Feedback Control of Growth Rate and Surface Roughness in Thin Film Growth

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A method for multivariable feedback control of surface roughness and growth rate in thin film growth using kinetic Monte-Carlo (MC) models is proposed. The method is applied to the process of thin film growth in a stagnation flow geometry including atom adsorption, desorption, and surface migration as the three processes that shape film microstructure and determine film growth rate. A multiscale model involving coupled partial differential equations to model the gas phase and a kinetic MC simulator, based on a large lattice, for the modeling of film growth, is used to simulate the process. A roughness and growth rate estimator constructed allowed computing estimates of the surface roughness and growth rate at a time-scale comparable to the real-time evolution of the process. The estimator involves kinetic MC simulators based on small lattice models, adaptive filters used to reduce stochastic fluctuations of the outputs of the small lattice MC simulator outputs, and measurement error compensators used to reduce the errors between the estimates and measurements. The interactions between the inputs (substrate temperature and inlet precursor mole fraction) and outputs (growth rate and surface roughness) in the closed-loop system are studied and found to be significant. A multivariable feedback controller, which uses the state estimator and explicitly compensates for the effect of input/output interactions, is designed to simultaneously regulate the growth rate and surface roughness by manipulating substrate temperature and inlet precursor mole fraction. Application of the estimator/controller structure to the multiscale process model demonstrates successful regulation of the surface roughness and growth rate to the desired set point values. This approach is shown to be superior to control of the growth rate and surface roughness using two independent feedback control loops.

Introduction

Deposition of thin films from gas-phase precursors has great industrial importance. The modern integrated circuit technology depends strongly on the uniformity and microstructure of deposited thin films (Granneman, 1993). Due to the increasingly stringent requirements on the quality of such films including uniformity, composition, and microstructure, and the desire to improve productivity by increasing wafer dimensions and reducing product variability, real-time feedback control of thin film deposition becomes important. These trends have motivated significant research efforts on feedback control of film deposition processes with emphasis on control of film spatial uniformity in rapid thermal chemi-

cal vapor deposition (for example, Baker and Christofides, 1999; Theodoropoulou et al., 1999) and plasma-enhanced chemical vapor deposition (for example, Armaou and Christofides, 1999). From a control point of view, film spatial uniformity control is a distributed control problem that can be addressed on the basis of continuum type transport-reaction models by using controller design methods for nonlinear parabolic partial differential equations (PDEs) (see Christofides (2001) for results and references on this problem).

In addition to achieving spatially uniform deposition of thin films, one would like to control film properties such as microstructure and composition that characterize film quality. This is motivated by the strong dependence of the electrical and mechanical properties of thin films on their microstruc-

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ture and composition (see, for example, Akiyama et al., 2002; Chang et al., 2001; Lee et al., 1999). A typical example of thin film growth where feedback control of film surface roughness could be useful is the deposition of thin films of materials with high dielectric constant (such as ZrO_2); such materials are expected to replace SiO_2 thin films to achieve higher performance and lower static-power operation in complementary metal oxide semiconductor (CMOS) devices (Lee et al., 1999). In this application, a rough ZrO_2 film surface would lead to silicidation of the interface between the ZrO_2 film and the polysilicon gate during rapid thermal annealing (Busch et al., 2002), which, in turn, would lead to an interfacial layer with low dielectric constant which would reduce the effective capacities of the gate dielectrics (Wallace and Wilk, 2002). Therefore, good control of surface roughness in the deposition of the ZrO_2 thin film is needed to achieve a smoother film surface with fewer reaction sites which would result in a suppression of the interfacial reactions. The study of feedback control of surface roughness is also motivated by the possibility to obtain roughness measurements in real-time using scanning tunneling microscopy (Voigtländer, 2001), spectroscopic ellipsometry techniques (Zapien et al., 2001), or by a combination of on-line measurements techniques for measuring gas-phase compositions with off-line measurement techniques for measuring surface roughness. An implementation of the latter approach was recently reported in Ni et al. (2003), where it was used to measure carbon composition of thin films in plasma-enhanced chemical vapor deposition using a combination of optical emission spectroscopy (OES) and X-ray photoelectron spectroscopy (XPS).

While deposition uniformity control can be accomplished on the basis of continuum type distributed models, precise control of film properties requires models that predict how the film state (microscopic scale) is affected by changes in the controllable process parameters (macroscopic scale). The desire to understand and control the microstructure of thin films has motivated extensive research on fundamental mathematical models describing thin film growth and its interactions with the surrounding gas. Thin film deposition is a typical process including multiple time and length scales, and this has motivated the use of multiscale models to obtain modeling descriptions which can be simulated with reasonable computing power. Specifically, one approach to model thin film growth is to model the gas-phase phenomena using a set of partial differential equations derived from mass, momentum, and energy balances, and to model the film growth using the kinetic Monte-Carlo (MC) simulation method with appropriate lattice configurations (see, for example, Lam and Vlachos, 2001; Vlachos, 1997). The two models are coupled through the film surface boundary conditions. Other approaches have been also developed to study the growth of thin films including the level set method (Chen et al., 2001). While multiscale modeling provides a computationally attractive alternative with respect to direct modeling of the entire thin film growth process using a molecular model, it still leads to dynamic models that cannot be solved fast enough for real-time estimation and control purposes.

Mathematically, kinetic MC simulation methods provide a numerical solution to the master equation (Kang and Weinberg, 1992), which is a stochastic partial differential equation describing the evolution of the probability that the thin film is at a certain microconfiguration. As an alternative with re-

spect to closed-form process models (such as partial differential equation models), which are not available for describing the evolution of microstructure of thin films, the kinetic MC simulation method can be used to predict average properties of the thin film (which are of interest from a control point of view, for example, surface roughness), by explicitly accounting for the microprocesses that directly shape thin film microstructure. As the lattice size used to perform the kinetic MC simulation increases, the average thin film properties, which are computed from the kinetic MC simulation, converge to the values obtained from the solution of the master equation. The accuracy of solutions of the kinetic MC simulation method depends on the size of the lattice used in the simulation which, in turn, determines the computational requirements of the simulation. The computational requirements for a solution with reasonable computing power make impossible the direct use of kinetic MC simulators in an on-line feedback control scheme. Motivated by this, recent research efforts have focused on the development of order reduction techniques for the master equation (Gallivan and Murray, 2003). Other approaches have also been developed to identify linear models from outputs of kinetic MC simulators and perform controller design by using linear optimal control theory (Armaou et al., 2002).

Despite these methodological advances and the development of techniques for real-time roughness measurements using real-space imaging methods (such as scanning tunneling microscope (STM) (Curtis et al., 1997; Voigtländer, 2001)), there is no systematic framework for the design of feedback control systems that integrates real-time measurements, multiscale models, and feedback control algorithms to shape film microstructure in real time. One of the main obstacles for real-time feedback control is the difficulty to obtain real-time measurements of film properties (such as surface roughness) at a frequency that is large enough to fully capture the dynamics of the growth process. This limits the direct use of real-time roughness measurement techniques for feedback control and motivates the design of model-based roughness estimators. In a previous work (Lou and Christofides, 2003), we proposed a systematic method for the design of roughness estimators which consist of kinetic Monte Carlo simulators based on small lattice models, an adaptive filter for noise reduction, and a measurement error compensator. Based on the estimator, a single-loop feedback control system was developed and applied to the multiscale model of a thin film growth process to regulate surface roughness.

This work focuses on multivariable feedback control of surface roughness and growth rate in thin film growth. The objective is to understand the nature of this multivariable control problem and to develop a systematic method for the design of a multivariable feedback control system, which can be implemented in real time. While the proposed method for estimation/control using kinetic Monte-Carlo simulators is applied to the process of thin film growth in a stagnation flow geometry, the methodological approach of this work is applicable to other thin film growth processes.

Initially, the thin film growth process is introduced and a multiscale model is presented. An estimator is then constructed that allows computing estimates of the surface roughness and growth rate at a time-scale comparable to the real-time evolution of the process. The multivariable control problem is then studied and a multivariable feedback control

system, which directly uses the estimator and compensates for the effect of multivariable input/output interactions, is designed to simultaneously regulate the growth rate and surface roughness by manipulating substrate temperature and inlet precursor concentration. Finally, the proposed control system is applied to the multiscale process model and successful regulation of the surface roughness and growth rate to the desired set point values is demonstrated through computer simulations.

Thin Film Growth Process

We consider the growth of a thin film from a fluid in a vertical, stagnation flow geometry (Gadgil, 1993). The process is shown in Figure 1. In this geometry, inlet fluid flow forms a uniform boundary layer adjacent to the surface of the substrate and precursor atoms diffuse through the boundary layer, reach at the surface, and adsorb onto the surface. Subsequently, adsorbed atoms may desorb to the gas phase or migrate on the surface.

From a modeling point of view, the major challenge is the integration of the wide range of length and time scales that are encountered in this thin film growth process (Vlachos, 1997). Specifically, in the gas phase, the processes of heat/mass transport can be adequately modeled under the hypothesis of continuum, thereby leading to PDE models for chamber temperature and species concentration. However, when the microstructure of the surface is studied, microscopic events such as atom adsorption, desorption, and migration have to be considered, and the length scale of interest reduces dramatically to the order of several atoms. In such a small length scale, the continuum hypothesis is no longer valid and deterministic PDEs cannot be used to describe the microscopic phenomena. Microscopic modeling techniques, such as Monte-Carlo simulation or molecular dynamics, should be employed to model the evolution of surface microstructure.

Although different modeling approaches are used to model the macroscopic and microscopic phenomena of the process, there are strong interactions between the macro- and microscale phenomena. For example, the concentration of the precursor in the inlet gas governs the rate of adsorption of atoms on the surface, which, in turn, influences the surface roughness and growth rate. On the other hand, the density of the adatoms on the surface affects the rate of desorption of atoms from the surface to the gas phase, which, in turn, influences the gas-phase concentration of the precursor. A multi-

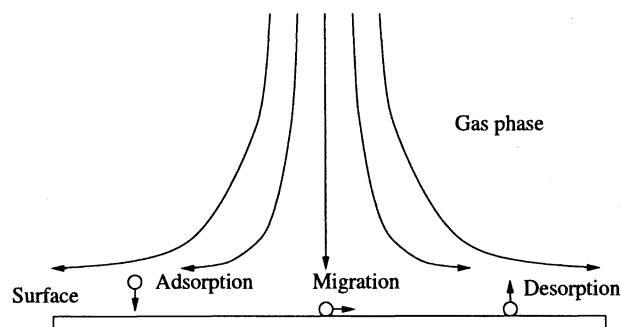


Figure 1. Thin film growth process.

scale model (Vlachos, 1997) is employed in this work to capture the evolution of both macroscopic and microscopic phenomena of the thin film growth process, as well as their interactions. A set of PDEs derived from the mass, momentum, and energy balances is used to describe the gas-phase dynamics. Kinetic MC simulation (Fichthorn and Weinberg, 1991) is employed to capture the evolution of surface microstructure and growth rate. Furthermore, the parameters of kinetic MC simulation, such as the temperature and precursor concentration, are provided by the solution of PDEs and the results from the kinetic MC simulation are used to determine the boundary conditions of the PDEs of the macroscopic model. Therefore, the macroscopic and microscopic models are coupled. In this article all the simulations are performed by coupling the kinetic MC simulation and the PDE model. In the remainder of this section, we provide the mathematical description of the multiscale process model.

Gas-phase model

Under the assumption of axisymmetric flow, the gas phase can be modeled through continuum type momentum, energy, and mass balances as follows (Lam and Vlachos, 2001)

$$\frac{\partial}{\partial \tau} \left(\frac{\partial f}{\partial \eta} \right) = \frac{\partial^3 f}{\partial \eta^3} + f \frac{\partial^2 f}{\partial \eta^2} + \frac{1}{2} \left[\frac{\rho_b}{\rho} - \left(\frac{\partial f}{\partial \eta} \right)^2 \right] \quad (1)$$

$$\frac{\partial T}{\partial \tau} = \frac{1}{P_r} \frac{\partial^2 T}{\partial \eta^2} + f \frac{\partial T}{\partial \eta} \quad (2)$$

$$\frac{\partial y_i}{\partial \tau} = \frac{1}{Sc_j} \frac{\partial^2 y_i}{\partial \eta^2} + f \frac{\partial y_i}{\partial \eta} \quad (3)$$

The following boundary conditions are used for $\eta \rightarrow \infty$

$$T = T_{\text{bulk}}$$

$$\frac{\partial f}{\partial \eta} = 1$$

$$y_j = y_{jb}, j = 1, \dots, N_g \quad (4)$$

and for $\eta \rightarrow 0$ (surface)

$$T = T_{\text{surface}}$$

$$f = 0$$

$$\frac{\partial f}{\partial \eta} = 0$$

$$\frac{\partial y_j}{\partial \eta} = 0, \text{ for } j \neq \text{growing species}$$

$$\frac{\partial y_{\text{growing}}}{\partial \eta} = \frac{Sc_{\text{growing}}(R_a - R_d)}{\sqrt{2a\mu_b\rho_b}} \quad (5)$$

where f is the dimensionless stream function, R_a and R_d are the rates of adsorption and desorption on the surface of the growing film, respectively, η is the dimensionless distance to

the surface, ρ is the density of the mixture, P_r is the Prandtl number, y_j and Sc_j are the mole fraction and Schmidt number of the species j , respectively, μ_b and ρ_b are the viscosity and the density at the bulk, respectively, a is the hydrodynamic strain rate, and $\tau = 2at$ is the dimensionless time.

Although the macroscopic model describes the spatio-temporal evolution of the precursor concentration and temperature (which influence the configuration of the growing surface), no direct information of the surface microstructure is available from the macroscopic model. Furthermore, the growth rate and the boundary conditions for the mass-transfer equation of the growing species depend on the rates of adsorption and desorption, which strongly depend on the surface microconfiguration. Therefore, a microscopic model is necessary to model the microstructure of the surface and to determine the boundary conditions of the mass-transfer equation.

Film growth model

The thin film growth problem considered in this work involves three distinct processes: the adsorption of atoms from the gas phase to the surface, the desorption of atoms from the surface to the gas phase, and the migration of atoms on the surface. The statistical properties of these processes can be studied by dividing a duration t into n identical time intervals of length δ . When $n \rightarrow \infty$, δ is small enough so that each time interval of length δ contains one event at the most. The average rate can be then defined as (Fichthorn and Weinberg, 1991)

$$r = \lim_{\delta \rightarrow 0} \frac{n_\delta}{t} \quad (6)$$

when n_δ is the number of time intervals containing events. Therefore, the probability that n_e events will occur in the time interval $[0, t]$ is

$$P_{n_e} = \binom{n}{n_e} (r\delta)^{n_e} (1 - r\delta)^{n - n_e} \quad (7)$$

where n is the number of intervals in time duration t and $\binom{n}{n_e} = n! / [n_e!(n - n_e)!]$. When $n \rightarrow \infty$

$$P_{n_e} = \frac{rt^{n_e}}{n_e!} e^{-rt} \quad (8)$$

which is a Poisson distribution (Van Kampen, 1992). Equation 8 can be readily applied to the adsorption, desorption, and migration processes involved in the thin film growth process. Due to the fact that the ensemble of independent Poisson processes is also a Poisson process, the thin film growth process is a Poisson process.

Due to the stochastic nature of the process, the probability that the surface is in a possible configuration α is described by the so-called master equation (Van Kampen, 1992)

$$\frac{dP_\alpha}{dt} = \sum_\beta (W_{\alpha\beta} P_\beta - W_{\beta\alpha} P_\alpha) \quad (9)$$

where P_α is the probability of the surface being in configuration α and $W_{\alpha\beta}$ is the transition probability rate of the surface going from configuration β to configuration α , which can be computed from the probability distribution of adsorption, desorption, and migration. It is hard to write down the explicit form of Eq. 9 because the number of the possible states is extremely large for most systems of realistic size. For example, for a system with 10×10 sites and a maximum height of 1, the number of configurations is $2^{100} \approx 10^{30}$. This makes impossible the direct solution of Eq. 9 for any system of meaningful size, using numerical methods for integration of ordinary differential equations (such as Runge-Kutta).

Monte Carlo methods provide an alternative approach to solve the master equation numerically. To capture the dynamic properties of the system, the MC algorithms must be able to satisfy the detailed balance criterion, appropriately calculate the lifetime of each MC event, and guarantee the independence of events by using an appropriate random number generator (Fichthorn and Weinberg, 1991). The way to satisfy detailed balance criterion differs when different algorithms are used. In general, there are two groups of MC algorithms which have been developed to simulate processes governed by the master equation: (a) the null-event algorithm (Ziff et al., 1986), and (b) the kinetic MC method (for example, Vlachos et al., 1993). When the null-event algorithm is used, which tries to execute MC events on randomly selected sites with certain probabilities of success, the probabilities of successful trials should be appropriately constructed to satisfy the detailed balance criterion. On the other hand, if the kinetic MC method is used, which selects the MC event before the selection of the site on which the MC event is going to be executed, the detailed balance criterion is satisfied by appropriately constructing the probabilities of the different kinds of MC events to be selected. Upon a successful MC event, the time passed during the event is computed based on the total rates of all the microprocesses (Vlachos, 1997). In this way, the dynamic properties of the system can be captured. Since the kinetic MC method is computationally more efficient than the null-event algorithm, it is used to simulate the surface processes in the thin film growth process in this study (see Reese et al. (2001) for a detailed discussion on comparison of computational efficiency of the kinetic MC method to the null-event algorithm).

To run the kinetic Monte-Carlo simulation for the evolution of the surface microstructure and growth rate, an $N \times N$ lattice is initially constructed. The size of the lattice (or the value of N) is determined based on the desired accuracy and the requirement to capture the main phenomena occurring on the surface. Generally, a larger lattice can achieve higher accuracy (in terms of average properties and size of stochastic fluctuations), but requires a larger solution time. When the lattice is ready, microscopic events are executed on the lattice based on the probabilities of the individual processes. To simplify the development, only first-nearest-neighbor interactions are considered, the solid-on-solid approximation of a simple cubic lattice is made, the (001) surface is studied, and periodic boundary conditions are used. Furthermore, although it has been shown that the barrier for a surface atom diffusion down a step and that for diffusion on a flat surface are not, in general, the same (Amar and

Family, 1996; Stumpf and Scheffler, 1994), the magnitude and even the sign of the difference are not generally known. In this work, we assume that the barrier for diffusion down a step and that for diffusion on a flat surface are the same.

In this study, we consider multilayer growth and assume that all the sites are available for adsorption at all times, and, thus, the adsorption rate is taken to be site independent. For an ideal gas, the adsorption rate at a site on the surface is given by the kinetic theory (Lam and Vlachos, 2001)

$$r_a = \frac{s_0 P y_{\text{growing}}}{\sqrt{2\pi m k T C_{\text{tot}}}} \quad (10)$$

where s_0 is the sticking coefficient, k is the Boltzmann constant, P is the chamber pressure, y_{growing} is the mole fraction of precursor on the surface of the growing film, C_{tot} is the concentration of sites on the surface, m is the molecular weight of the precursor, and T is the gas-phase temperature above the surface.

The rate of desorption at a site on the surface depends on the local activation energy. Under the consideration of only first nearest neighbor interactions, the desorption rate of an atom from the surface with n first nearest neighbors is

$$r_d(n) = \nu_0 \exp\left(-\frac{nE}{kT}\right) \quad (11)$$

where E is the energy associated with a single bond on the surface and ν_0 is the frequency of events, which is determined by the following expression

$$\nu_0 = k_{d0} \exp\left(-\frac{E_d}{kT}\right) \quad (12)$$

where k_{d0} is the event frequency constant and E_d is the energy associated with desorption.

Surface migration is modeled as desorption followed by re-adsorption (Gilmer and Bennema, 1972), and the migration rate per surface site is given by

$$r_m(n) = \nu_0 A \exp\left(-\frac{nE}{kT}\right) \quad (13)$$

The parameter A is associated with the energy difference that an atom on a flat surface has to overcome in jumping from one lattice site to an adjacent one and is given by

$$A = \exp\left(\frac{E_d - E_m}{kT}\right) \quad (14)$$

where E_m is the energy associated with migration.

The parameters in Eqs. 10–14 can be determined either by experiments or by theoretical computations. Parameters of the process studied in this work are shown in Table 1. When the lattice is set and the rates of the three events are determined based on the corresponding rate expressions, a kinetic Monte-Carlo simulation is executed following the algorithm reported in Vlachos (1997). First, the surface atoms are

Table 1. Process Parameters

Sticking coefficient	s_0	0.1	
Precursor molecular weight	m	4.65×10^{-26}	kg
Surf. site concentration	C_{tot}	10^{19}	sites/m ²
Event freq. const	k_{d0}	10^{13}	s ⁻¹
Bond energy	E	7.14×10^4	J/mol
Desorption energy	E_d	7.14×10^4	J/mol
Migration energy	E_m	4.28×10^4	J/mol
Chamber pressure	P	10^5	Pa
Strain rate	a	5	s ⁻¹

grouped into five classes based on the number of side neighbors (such as surface atoms have 0, 1, 2, 3 and 4 side neighbors); in each class, the atoms have the same desorption and migration rates (the adsorption rate is site independent). Then, a random number is generated to select an event to be run based on the rates; if the event is desorption or migration, the class in which the event will happen is also selected. After that, a second random number is generated to select the site where the event will be executed; if the event is adsorption, the site is randomly picked from sites in the entire lattice; if the event is desorption or migration, the site is randomly picked from the list of the sites in the selected class. After the site is selected, the MC event is executed. If the event is adsorption or desorption, it is executed by adding or removing one atom on the selected site; if the event is migration, a third random number is generated to randomly pick a neighboring site that has a lower height (target site), and to move the atom from the original site to the target site. Upon an executed event, a time increment dt is computed by (Lam and Vlachos, 2001)

$$dt = \frac{-\ln \xi}{r_a \times N_T + \nu_0(1 + A) \sum_{m=1}^5 N_m \exp\left(\frac{-mE}{kT}\right)} \quad (15)$$

where ξ is a random number in the (0, 1) interval, N_T is the total number of sites on the lattice, and N_m is the number of atoms that have m side-neighbors on the surface. After an MC event is executed, the five classes are updated and the next step of simulation can be performed. This algorithm guarantees that every trial is successful, satisfies the detailed balance criterion, guarantees the independence of events, and is efficient compared to traditional null event algorithms (Reese et al., 2001).

Real-Time Estimator for Surface Roughness and Growth Rate

Surface roughness and growth rate are properties of interest from a control point of view since they directly influence device properties and productivity. To be able to achieve good control of surface roughness and growth rate, it is important to develop an estimator that can provide estimates of these variables in real time. In this section, we develop such an estimator by following a methodology that was proposed in Lou and Christofides (2003).

The roughness r is represented by the number of broken bonds on the surface and is computed according to the fol-

lowing expression (Raimondeau and Vlachos, 2000)

$$r = \frac{\sum (|h_{i+1,j} - h_{i,j}| + |h_{i-1,j} - h_{i,j}| + |h_{i,j+1} - h_{i,j}| + |h_{i,j-1} - h_{i,j}|)}{2 \times N \times N} + 1 \quad (16)$$

where N is the size of the lattice and $h_{i,j}$ is the number of atoms at site (i, j) .

The growth rate gr (units monolayer/second, ML/s) determines how fast the thin film grows. In this study, the growth rate is defined as the difference between the rate of adsorption and the rate of desorption on the surface, and is given by the following expression

$$gr = \frac{\sum_{i,j} r_{ai,j} - r_{di,j}}{N \times N} \quad (17)$$

where $r_{ai,j}$ and $r_{di,j}$ (units s^{-1}) are the rates of adsorption and desorption at the site (i, j) of the surface, respectively, and N is the size of the lattice. The rate $r_{di,j}$ depends on the number of nearest neighbors at site (i, j) and is obtained from the kinetic MC simulations.

In the kinetic MC simulation, the size of the lattice influences the accuracy of the result and the computational demand. In previous work (Lou and Christofides, 2003), we discussed the dependence of simulation results on the lattice size used in the MC simulations. The simulation results show that when the lattice size is sufficiently large, the roughness profiles obtained from kinetic MC simulations are independent of the lattice size. To implement real-time feedback control based on a model that captures the evolutions of surface roughness and growth rate, the size of the lattice has to be selected to make the model solution time comparable to the process real-time evolution, while capturing the dominant phenomena occurring on the surface. In our simulations, when the size of the lattice is reduced to 30×30 , the solution time of the kinetic MC simulation is comparable to the real-time process evolution and the average values of the surface roughness and growth rate approximate well the average val-

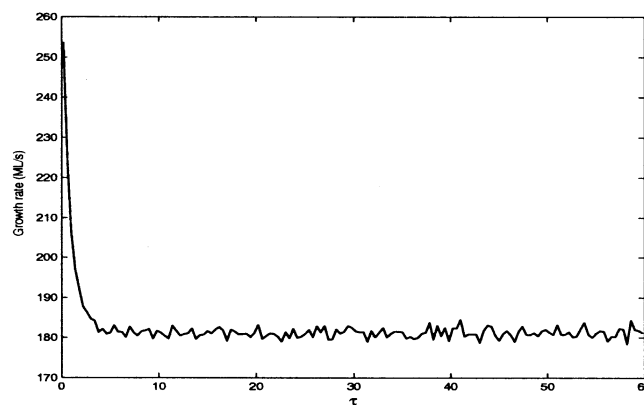


Figure 3. Growth rate from the computation of the average of six independent kinetic MC simulations, which utilize a 20×20 lattice.

ues of these variables, which are obtained by running the kinetic MC simulation on a 120×120 lattice (this is a sufficiently large lattice to ensure simulation results which are independent of the lattice size; see the discussion at the end of remark 1). However, the outputs from a kinetic MC simulation using a 30×30 lattice contain significant stochastic fluctuations, and, thus, they cannot be directly used for feedback control (such an approach would result in significant fluctuations of the control action, which could perturb unmodeled (fast) process dynamics and should be avoided).

The fluctuations on the values of the outputs (that is, surface roughness and growth rate) obtained from the kinetic MC simulation using the 30×30 lattice can be reduced by independently running several small lattice kinetic MC simulations with the same parameters and averaging the outputs of the different runs. Figure 2 shows the growth rate obtained from a kinetic MC simulation, which uses 20×20 lattice. Figure 3 shows the growth rate obtained from the computation of the average of six independent kinetic MC simulations which utilize a 20×20 lattice. These results show that when the outputs from multiple kinetic MC simulations that use small lattices are averaged, growth rate fluctuations can be significantly reduced. Figure 4 shows the surface rough-

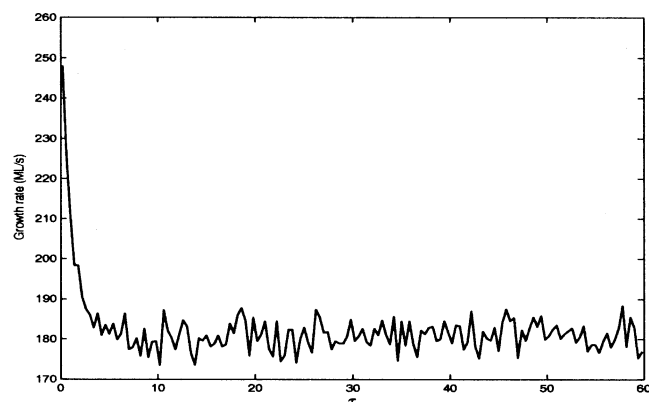


Figure 2. Growth rate from a kinetic MC simulation, which uses a 20×20 lattice.

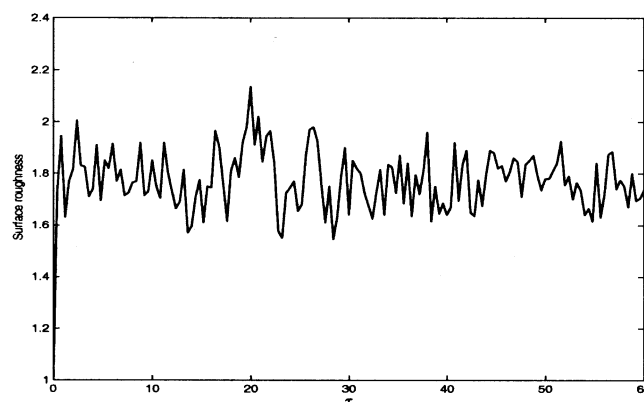


Figure 4. Surface roughness from a kinetic MC simulation, which uses a 20×20 lattice.

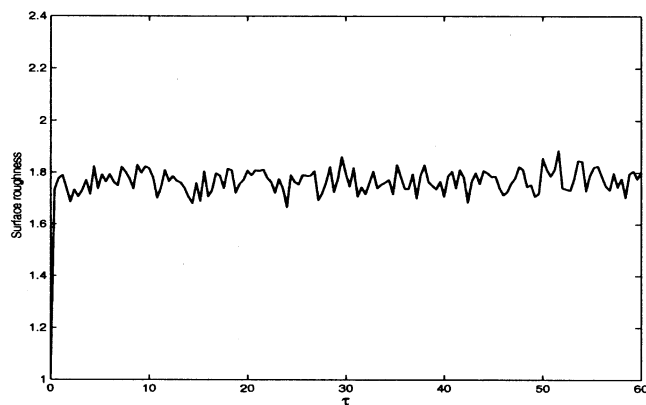


Figure 5. Surface roughness from the computation of the average of six independent kinetic MC simulations, which utilize a 20×20 lattice.

ness obtained from a MC simulation which uses a 20×20 lattice. Figure 5 shows the surface roughness obtained from the computation of the average of six independent kinetic MC simulations which utilize a 20×20 lattice. Again, fewer fluctuations are observed in the roughness profile obtained by averaging the outputs from multiple small lattice kinetic MC simulations.

However, by increasing the number of kinetic MC simulations which run simultaneously, the computational time is also increasing (unless each simulation can run in a different processor which is possible because there is no interaction between the six simulations). For the results shown in Figures 3 and 5, the computational time is six times as much as that needed to perform one MC simulation run (Figures 2 and 4) and it is approximately equal to the computational time needed to run a MC simulation on a 30×30 lattice. Figures 6 and 7 show growth rate and roughness profiles obtained from a MC simulation which uses a 30×30 lattice. Comparing the simulation results shown in Figures 3 and 6 and Figures 5 and 7, we find that the roughness and growth rate obtained by averaging six independent kinetic MC simulations, which use a 20×20 lattice model, contain less fluctuations than those obtained from a kinetic MC simulation which uses a

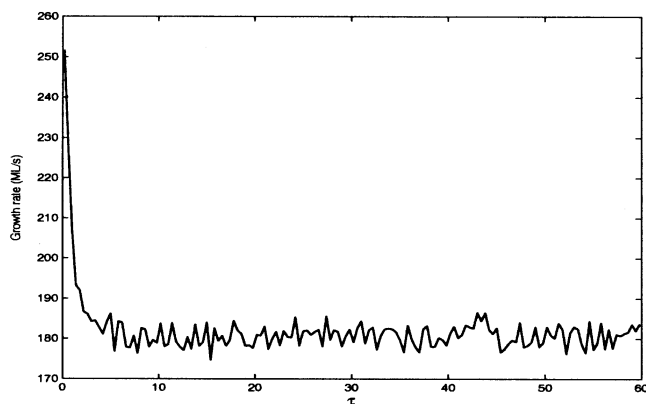


Figure 6. Growth rate from a kinetic MC simulation, which uses a 30×30 lattice.

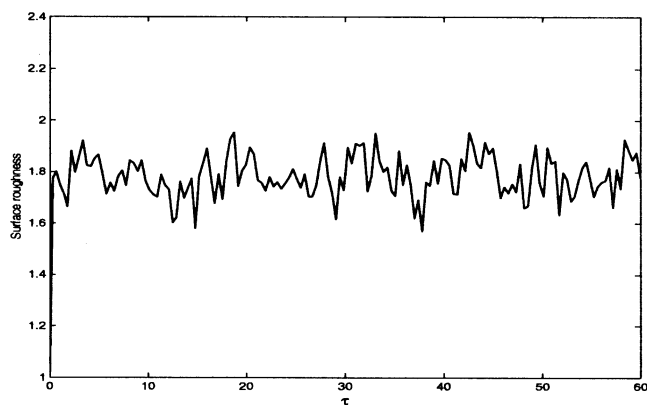


Figure 7. Surface roughness from a kinetic MC simulation, which uses a 30×30 lattice.

30×30 lattice model. This demonstrates that, for a fixed computational time, the use of a kinetic MC simulator based on multiple small lattice models would yield growth rate and surface roughness profiles with fewer fluctuations compared to a kinetic MC simulator, which uses a single lattice with larger size.

The predicted profiles of surface roughness and growth rate, which are obtained from kinetic MC simulation based on multiple small lattice models, still contain stochastic fluctuations and are not robust (due to the open-loop nature of the calculation) with respect to disturbances and variations in process parameters. To alleviate these problems, we follow the approach proposed in Lou and Christofides (2003) and combine the small lattice kinetic MC simulator with an adaptive filter to reduce the stochastic fluctuations on the surface roughness and growth rate profiles, and a measurement error compensator to improve the estimates of these variables using on-line measurements. We use the same adaptive filter and measurement error compensator structure for both the surface roughness and the growth rate. To simplify the notation of the mathematical formulas, we only present the general structure of the adaptive filter and of the measurement error compensator. Specifically, the adaptive filter is a second-order dynamical system with the following state-space representation

$$\begin{aligned} \frac{d\hat{y}_r}{d\tau} &= y_1 \\ \frac{dy_1}{d\tau} &= \frac{K}{\tau_I} (y_r - \hat{y}_r) - \frac{1}{\tau_I} y_1 \end{aligned} \quad (18)$$

where y_r is the output of the kinetic MC simulation based on multiple small lattice models, $\hat{y}_r$ is the filter output, K is the filter gain, and τ_I is the time constant. To accelerate the response of the filter and avoid large overshoot, $\tau_I = 0.5/K$. To achieve both fast tracking of the dynamics of the outputs and efficient noise rejection, the gain of the filter is adaptively adjusted according to the following law

$$K(\tau) = K_0 \frac{\left| \int_{\tau-\Delta\tau}^{\tau} y_r(t) dt - \int_{\tau-2\Delta\tau}^{\tau-\Delta\tau} y_r(t) dt \right|}{\Delta\tau^2} + K_s \quad (19)$$

where K_0 is a constant, K_s is the steady-state gain for the adaptive filter, and $\Delta\tau$ is the dimensionless time interval between two updates of K . Although a better tracking performance is expected when a small $\Delta\tau$ is used, a very small $\Delta\tau$ will introduce the effect of fluctuations on the filter gain and should be avoided.

The measurement error compensator uses the available on-line measurements (in the numerical simulations the values of the surface roughness and growth rate obtained from the large lattice model are used) to produce improved estimates of the surface roughness and growth rate. The state-space representation of the measurement error compensator is

$$\frac{de}{d\tau} = K_e [y_h(\tau_{m_i}) - \hat{y}(\tau_{m_i})]; \quad \tau_{m_i} < \tau \leq \tau_{m_{i+1}}; i = 1, 2, \dots \quad (20)$$

and the final roughness estimates are computed by

$$\hat{y} = \hat{y}_r + e \quad (21)$$

In the above equations, K_e is the compensator gain, e is the estimated model error, which is used to compensate the model output, $\hat{y}$ is the roughness estimates, $\hat{y}_r$ is the filtered output from a kinetic MC simulator which uses a small lattice and y_h is the output of a kinetic MC simulator which uses the large lattice (in an experimental setup y_h could be obtained from the measurement sensor). Since the roughness measurements are only available at discrete points in time $\tau_m = [\tau_{m_1}, \tau_{m_2}, \dots]$, the righthand side of Eq. 20 is computed at the time that a roughness measurement is available and is kept in this value in the time interval between two available roughness measurements.

The combination of the adaptive filter and the measurement error compensator functions as an estimator, which is capable of accurately predicting the evolution of surface roughness and growth rate during the thin film growth by using measurements of the precursor concentration above the substrate. In this work, we assume that measurements of pre-

cursor concentration above the substrate are available; when such measurements are not available, a state estimator can be constructed on the basis of the PDE model that describes the gas-phase species concentrations and the temperature to obtain estimates of this quantity (see Christofides (2001) for estimator design methods for PDE systems). Figures 8 and 9 show the growth rate and surface roughness profiles computed by the estimator, which uses a kinetic MC simulator based on six 20×20 lattice models (solid lines); they are compared with the growth rate and surface roughness profiles obtained from a kinetic MC simulator, which uses a 120×120 lattice model. The results clearly show that the developed estimator can accurately predict the evolution of growth rate and surface roughness. Note also that the developed estimator can be used for real-time feedback control, since the computational time needed to run kinetic MC simulation based on six 20×20 lattice models is comparable to the real-time process evolution.

Remark 1. Referring to the selection of the lattice size, it is important to point out that while kinetic MC simulation, based on multiple 20×20 lattice models, can adequately capture the evolution of surface roughness and growth rate in the specific thin film growth problem under consideration, the dimension of the small lattice in general should be chosen so that the interactions between the surface atoms are adequately captured, and also that it is large enough to describe all the spatio-temporal phenomena occurring on the surface (such as cluster formation). Furthermore, the small lattice should be chosen to provide accurate estimates of the desired properties to be controlled. For example, in the case of surface roughness, this quantity is defined as the average number of broken bonds for all surface atoms and the microscopic unit involved is an individual atom. When a 20×20 small lattice is used, the computation of surface roughness involves hundreds of surface atoms, which is adequate to obtain the expected value. However, when the property of interest is, for example, step density, a larger lattice is needed to obtain a convergent average value from the kinetic MC simulation. At this point, it is important to note that the pro-

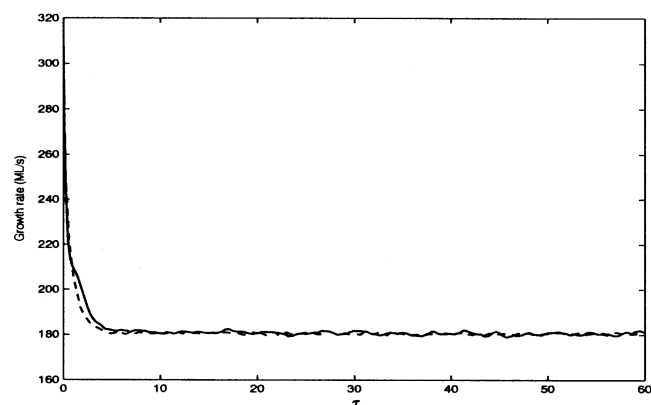


Figure 8. Growth rate profiles from the estimator (solid line) and from a kinetic MC simulation, which uses a 120×120 lattice model (dashed line).

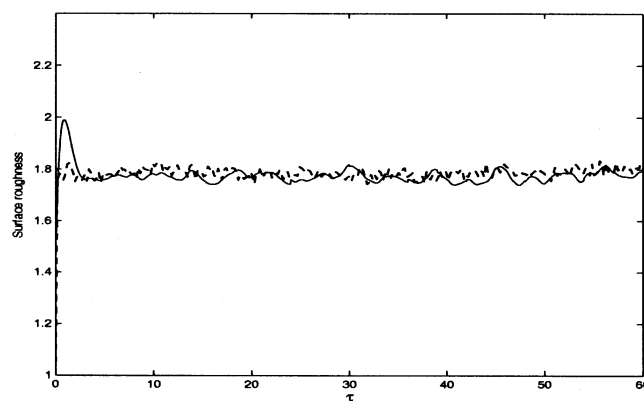


Figure 9. Surface roughness profiles from the estimator (solid line) and from a kinetic MC simulation, which uses a 120×120 lattice model (dashed line).

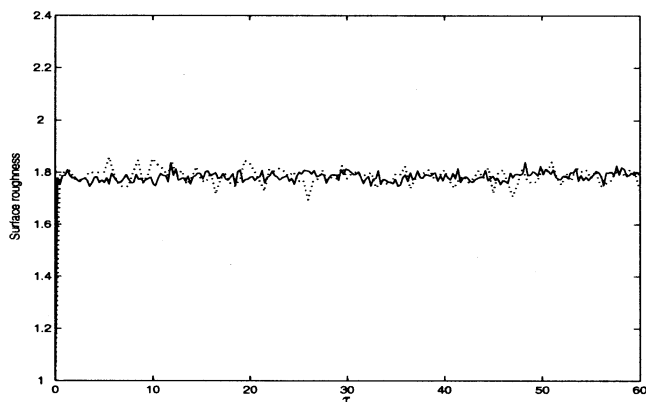


Figure 10. Comparison of surface roughness profiles from a kinetic MC simulator, which uses an 80×80 lattice (dotted line), and that from a kinetic MC simulator, which uses a 120×120 lattice (solid line).

posed reduction of lattice size can be viewed as an alternative (and quite intuitive) way to perform order reduction of the master equation (see Gallivan and Murray, 2003) for reduction approaches based directly on the master equation). In this work, when the surface roughness and growth rate are considered in the process with parameters shown in Table 1, we found out that an 80×80 lattice is sufficient to capture the evolution of the process and that a further increase of the lattice size leads to no observable improvement in the accuracy of the simulation results; this is shown in Figures 10 and 11, which show comparisons of surface roughness profiles and growth rate profiles from a kinetic MC simulator, which uses an 80×80 lattice, and those from a kinetic MC simulator, which uses a 120×120 lattice. Therefore, in the remainder of this work, we use a kinetic MC simulator, which uses an 80×80 lattice to describe the evolution of the thin film growth under open-loop and closed-loop conditions.

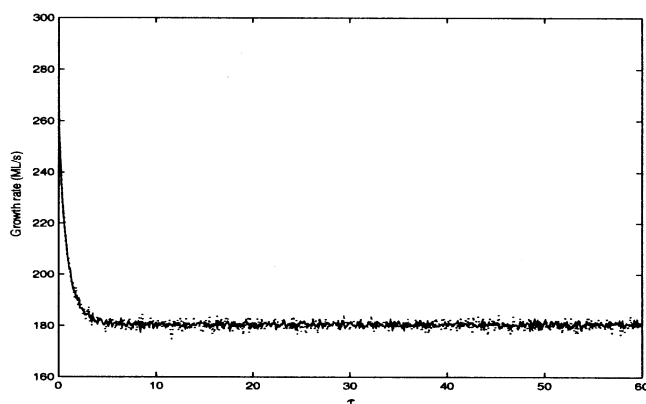


Figure 11. Comparison of growth rate profiles from a kinetic MC simulator, which uses an 80×80 lattice (dotted line), and that from a kinetic MC simulator, which uses a 120×120 lattice (solid line).

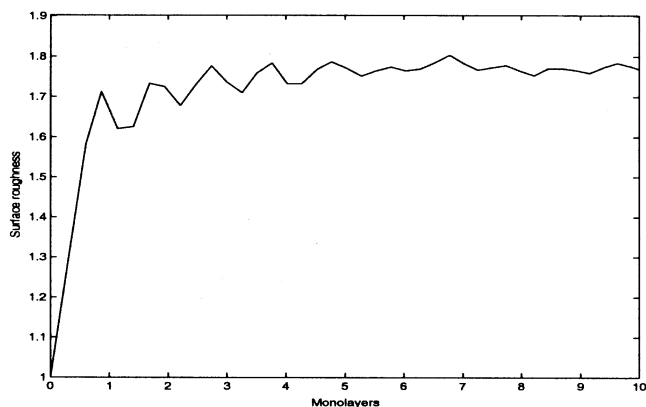


Figure 12. Evolution of surface roughness during the formation of the first 10 layers.

Remark 2. We note that the surface roughness during the formation of a single monolayer slightly fluctuates around the surface roughness value of the completely formed monolayer. This can be observed in Figure 12, which shows the evolution of surface roughness during the formation of the first 10 layers in the deposition from a kinetic MC simulator using an 80×80 lattice. However, this fluctuation is very small and does not influence the performance of the feedback control system. The same comment also applies to the evolution of the growth rate; Figure 13 shows the evolution of the growth rate during the formation of the first 200 layers from the same kinetic MC simulation.

Remark 3. It is important to note that, because available techniques for real-time measurement of film properties (such as surface roughness) do not provide measurements at a large enough frequency to fully capture the film growth dynamics, the use of a roughness estimator within a feedback control scheme is needed to predict film growth behavior at time instants in which on-line measurements are not available. In a recent study (Lou and Christofides, 2003), we demonstrated that feedback control, which exclusively relies on infrequent on-line measurements, leads to poor closed-loop perfor-

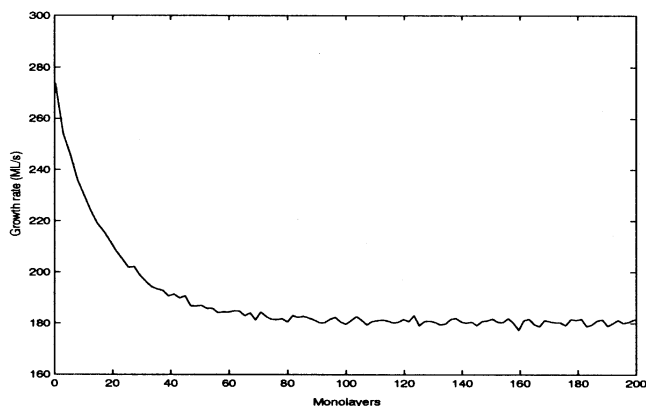


Figure 13. Evolution of growth rate during the formation of the first 200 layers.

mance (no matter what is the tuning of the controller) compared to feedback control, which employs our estimator to compute roughness estimates.

Feedback Control of Surface Roughness and Growth Rate

The efficient production of high-quality thin films requires that the surface roughness and growth rate are maintained at desired levels. Therefore, the objective of this section is to study the nature of the multivariable control problem and to develop a systematic method for the design of a multivariable feedback control system, which can be implemented in real time. The feedback control system makes use of the developed estimator, which provides a computationally feasible approach to predict the growth rate and surface roughness of the thin film in real time. Given the set of available manipulated inputs and the desired control objectives, the control problem is formulated as the one of regulating the surface roughness and growth rate by manipulating the substrate temperature and precursor mole fraction in inlet gases.

We will begin with a study of the feasibility of the control problem formulation, continue with an analysis and evaluation of the effect of input/output interactions on closed-loop performance, and close with the design and evaluation of the multivariable feedback control structure.

Feasibility of the control problem formulation

In this subsection, we establish that it is feasible to control surface roughness and growth rate by manipulating the substrate temperature and precursor mole fraction in inlet gases. To this end, we perform the following set of closed-loop simulations: (1) the growth rate is controlled by manipulating the precursor mole fraction in the inlet gas while the substrate temperature is kept constant; (2) the surface roughness is controlled by manipulating the substrate temperature, while the inlet precursor mole fraction is kept constant. The process parameters used in the simulations are shown in Table 1. In our calculations, since the precursor mole fraction is very small, we assume that the chamber pressure is independent of the precursor mole fraction. In each simulation, a single-loop controller/estimator structure of the type shown in Figure 14 is used to control the process. In this structure, the estimator, which includes multiple kinetic MC simulators

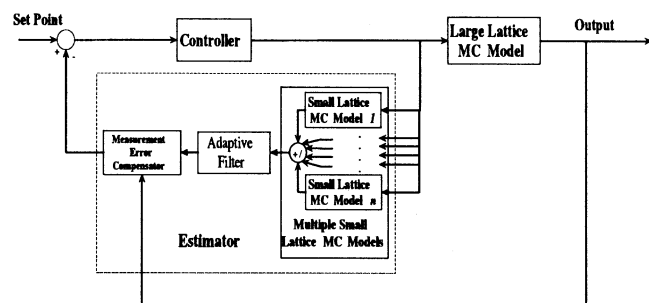


Figure 14. Estimator/controller structure using a kinetic MC simulator based on multiple small lattice models.

Table 2. Growth Rate Estimator and Controller Parameters

K_0	K_s	K_e	K_c	τ_c
0.5	1.0	0.08	2.0×10^{-9}	0.4

using small lattice models, an adaptive filter, and a measurement error compensator, is used to provide estimates of the controlled variable (surface roughness or growth rate) in a time-scale comparable to the real-time evolution of the process. The estimates are used in the controller to determine the control action. Since the models that describe the evolution of surface roughness and growth rate are not available in a closed form, a proportional integral (PI) controller is used to compute the control action

$$u(\tau) = K_c \left[(y_{\text{set}} - \hat{y}) + \frac{1}{\tau_c} \int_0^\tau (y_{\text{set}} - \hat{y}) dt \right] \quad (22)$$

where y_{set} is the set point of the output, $\hat{y}$ is the output of the estimator, K_c is the proportional gain, and τ_c is the integral time constant.

In the case of controlling the growth rate, the size of the small lattice is 20×20 and the outputs of six small lattice kinetic MC simulators are averaged within the estimator. A kinetic MC simulator based on an 80×80 lattice model is used to describe the evolution of the process. The time interval between two available measurements is taken to be 0.3 s, which is consistent with available techniques that can be used to measure growth rate in real time (Pickering, 2001). The substrate temperature is kept constant at 800 K and the initial inlet precursor mole fraction is 2.0×10^{-5} ; these values correspond to a growth rate of about 180 ML/s. The desired set point value for the growth rate is 220 ML/s. The parameters for the growth rate estimator and the PI controller used in this simulation are shown in Table 2. Figures 15 and 16 show the growth rate and the inlet precursor mole fraction under feedback control. The results clearly show that the growth rate can be successfully controlled to the desired set point by manipulating the precursor mole fraction.

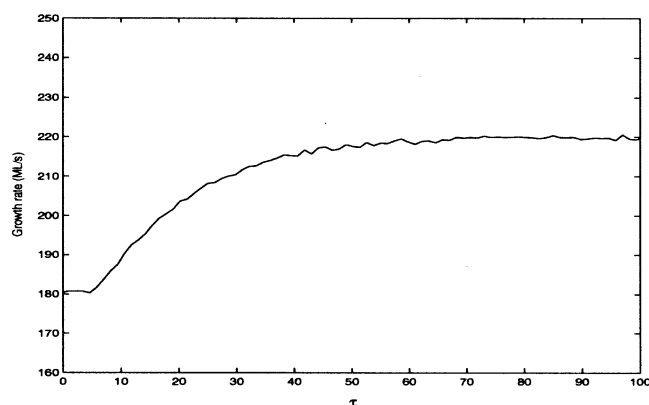


Figure 15. Closed-loop growth rate under single-loop feedback control using the estimator/controller structure of Figure 14.

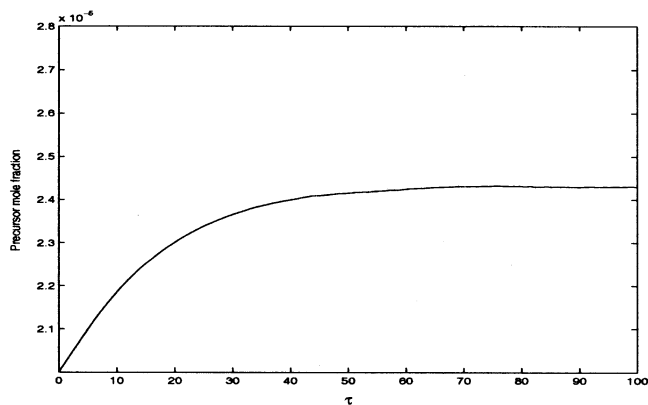


Figure 16. Inlet precursor mole fraction under single-loop feedback control using the estimator/controller structure of Figure 14.

In the second simulation run, the surface roughness is controlled by manipulating the substrate temperature while the inlet precursor mole fraction is kept constant. Figures 17 and 18 show the profiles of the surface roughness and substrate temperature in the closed-loop system. A kinetic MC simulator, which uses an 80×80 lattice model, is used to describe the evolution of the process. The inlet precursor mole fraction is kept constant 2.0×10^{-5} and the substrate temperature is initially 800 K. The desired set point value of surface roughness is 1.5. The parameters for the roughness estimator and controller are shown in Table 3. The controller successfully drives the surface roughness to the set point by manipulating the substrate temperature.

Effect of multivariable input/output interactions

The objective of this subsection is to understand the influence of multivariable input/output interactions on closed-loop performance and determine whether there is a need for the design and implementation of a multivariable controller, which compensates for the effect of such interactions. To this end, we consider the problem of simultaneous regulation of

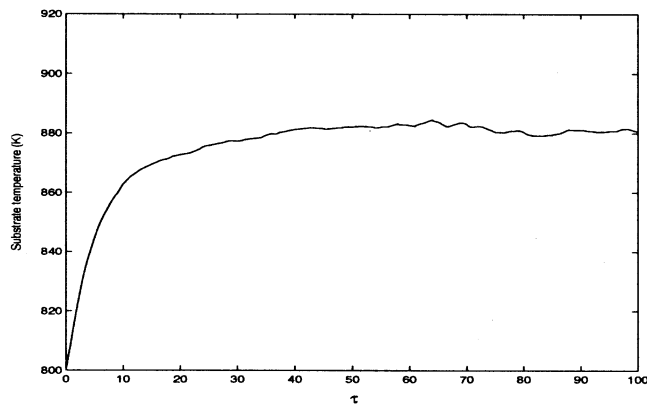


Figure 18. Substrate temperature under single-loop feedback control using the estimator/controller structure of Figure 14.

the growth rate and of the surface roughness and use a feedback control system that comprises of the estimator and two *single-loop* proportional integral controllers. Specifically, based on the results of the previous subsection, the input/output pairs are substrate temperature (T), surface roughness (r), and inlet precursor mole fraction (y), growth rate (gr).

A closed-loop system simulation is carried out to evaluate this approach to simultaneous control of growth rate and surface roughness. In this simulation, the outputs from six kinetic MC simulators running 20×20 lattice models are averaged within the estimator. A kinetic MC simulator which uses an 80×80 lattice model is used to describe the evolution of the process. The parameters for the estimator and the controllers are those listed in Tables 2 and 3.

Figures 19 and 20 show the surface roughness and substrate temperature. Comparing Figures 17 and 19, we find

Table 3. Roughness Estimator and Controller Parameters

K_0	K_s	K_e	K_c	τ_c
0.5	1.0	0.08	-15	0.3

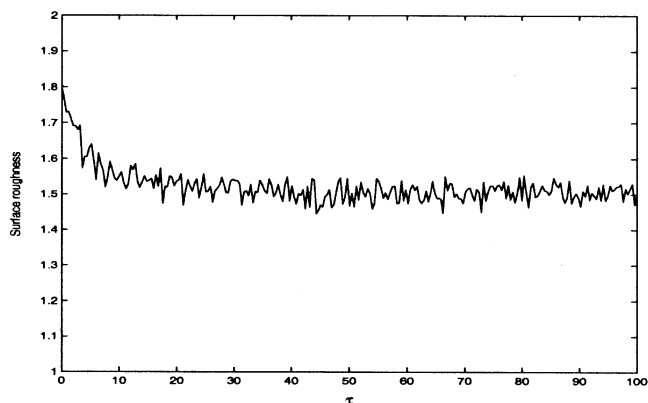


Figure 17. Closed-loop surface roughness under single-loop feedback control using the estimator/controller structure of Figure 14.

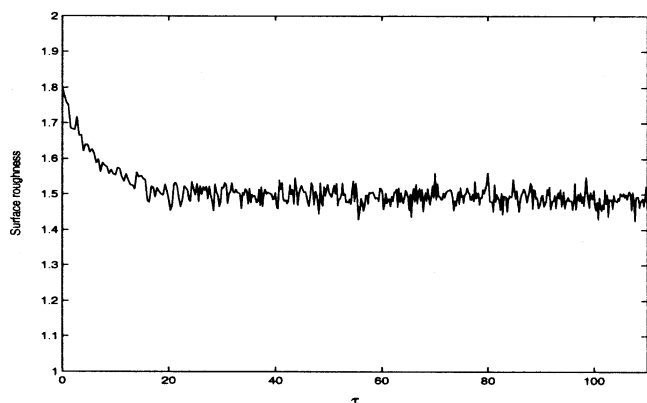


Figure 19. Closed-loop surface roughness under feedback control, which does not account for multivariable input/output interactions.

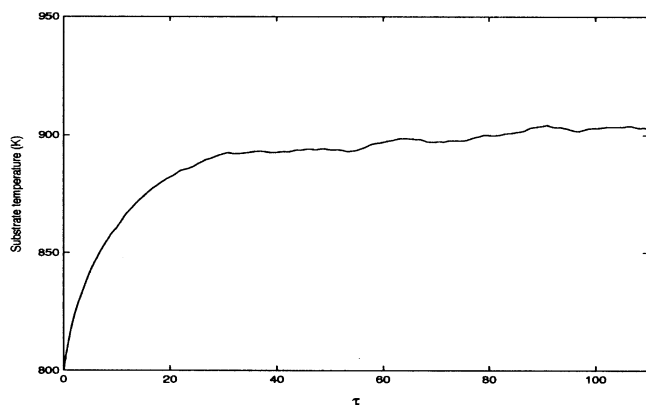


Figure 20. Substrate temperature using feedback control, which does not account for multivariable input/output interactions.

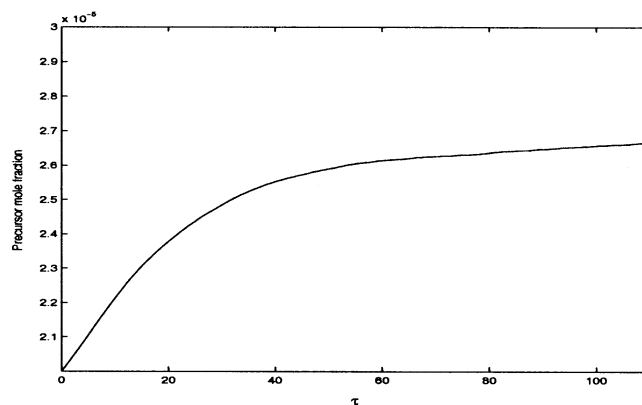


Figure 22. Precursor mole fraction using feedback control, which does not account for multivariable input/output interactions.

very similar roughness profiles, which means that multivariable input/output interactions do not influence the profile of the surface roughness under single-loop control.

Figures 21 and 22 show the profiles of the growth rate and inlet precursor mole fraction for the same simulation run. It is clear that the control system successfully drives the growth rate to the desired set point values. However, the transient response of the growth rate in the case of simultaneous growth rate and surface roughness control is slower compared to the case where the growth rate is the only controlled output and the structure of Figure 14 is used (Figure 23).

The reason for the slower transient response of the growth rate is the effect of multivariable input/output interactions (that is, the influence of the variation of substrate temperature on growth rate and the influence of the variation of inlet precursor mole fraction on surface roughness in the closed-loop system); these interactions need to be compensated for in order to speed up the growth rate response and improve closed-loop performance.

Multivariable feedback control structure

This subsection focuses on the design and evaluation of a multivariable feedback control structure. To this end, we need

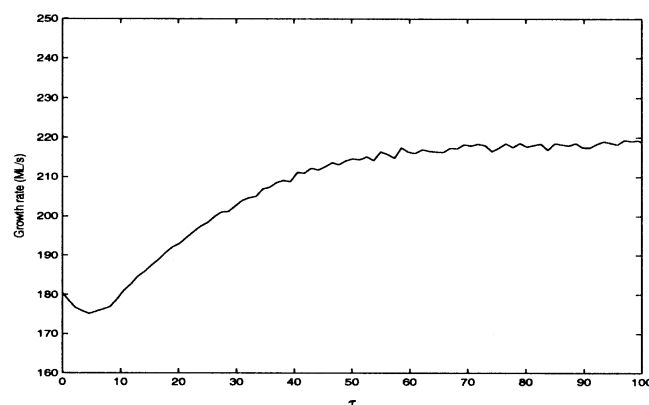


Figure 21. Closed-loop growth rate under feedback control, which does not account for multivariable input/output interactions.

to understand and model the interactions between inputs and outputs; two simulations are carried out to observe the changes of the surface roughness and growth rate for: (1) a step change on substrate temperature with constant inlet precursor mole fraction; (2) a step change on inlet precursor mole fractions with constant substrate temperature.

Figure 24 shows the growth rate and surface roughness when the substrate temperature is kept at 800 K and the inlet precursor mole fraction changes from 2.0×10^{-5} to 2.1×10^{-5} at $\tau = 10$. The results show that the growth rate increases from around 180 ML/S to 190 ML/S, but there is no observable change in the surface roughness, which means the change of the inlet precursor mole fraction has very little influence on the surface roughness.

The interactions between the substrate temperature and the surface roughness and growth rate are studied by keeping the inlet precursor mole fraction at 2.0×10^{-5} and increasing the substrate temperature from 800 K to 840 K at $\tau = 10$. Figure 25 shows the responses of growth rate and surface

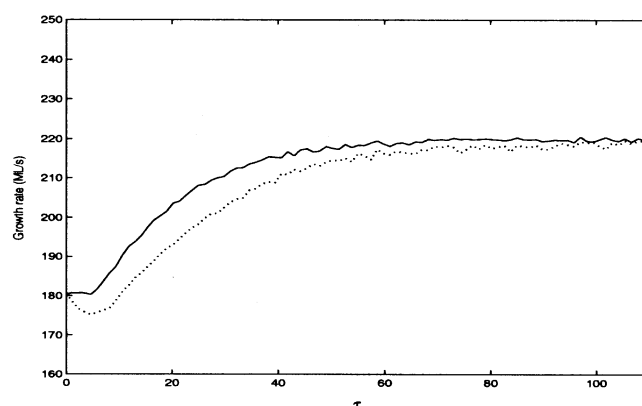


Figure 23. Comparison of closed-loop growth rate profiles.

(a) Growth rate is the only controlled output and the structure of Figure 14 is used (solid line); (b) simultaneous regulation of growth rate and surface roughness is considered and feedback control, which does not account for multivariable input/output interactions, is used (dotted line).

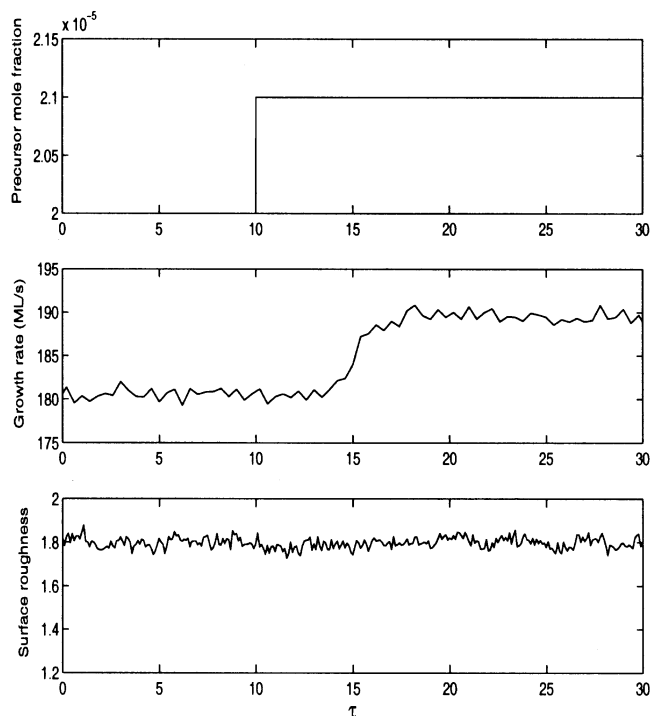


Figure 24. Growth rate (middle plot) and surface roughness (bottom plot) profiles for a step change in inlet precursor mole fraction (top plot) from 2.0×10^{-5} to 2.1×10^{-5} —substrate temperature is kept at 800 K.

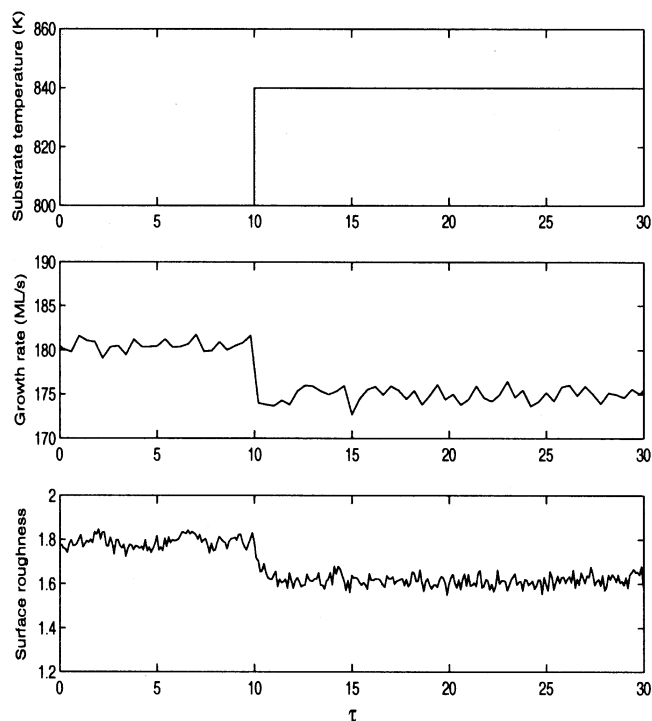


Figure 25. Growth rate (middle plot) and surface roughness (bottom plot) with a step change in substrate temperature (top plot) from 800 K to 840 K.

Inlet precursor mole fraction is 2.0×10^{-5} .

roughness to the substrate temperature change. The simulation results show that the growth rate drops from approximately 180 ML/s to 175 ML/s and the roughness drops from approximately 1.8 to 1.6, which indicates that variations on the substrate temperature influence both surface roughness and growth rate. It should be pointed out that this specific coupling pattern is valid only under the specific growth conditions used in this simulation.

To improve the closed-loop performance, a multivariable feedback control structure is developed, which explicitly compensates for the effect of the multivariable interactions occurring in the process. The controller structure is obtained by introducing a compensation block between the multiple single-loop controllers and the process. Because the interactions between inlet precursor mole fraction and surface roughness are not significant (as shown in Figure 24—bottom plot), only one compensator is needed in this example. The multivariable control system using the estimator/controller structure with interaction compensation is shown in Figure 26. $G_1(s)$ is the transfer function between the substrate temperature and the growth rate, and $G_2(s)$ is the transfer function between the inlet precursor mole fraction and the growth rate. Step tests are used to identify the expression and parameters of $G_1(s)$ and $G_2(s)$. Specifically, based on the simulation results shown in Figures 24 (middle plot) and Figure 25 (middle plot), G_1 and G_2 are taken to be constants with the following values $G_1(s) = 0.125$ and $G_2(s) = 1.0 \times 10^7$. The rest of the process and controller parameters used in the simulation are the same to those shown in Tables 2 and 3.

A closed-loop system simulation is performed to evaluate the effectiveness of the multivariable estimator/control structure with interaction compensation shown in Figure 26. The size of the small lattice is 20×20 and the outputs from six kinetic MC simulators based on 20×20 lattice models are averaged within the estimator. A kinetic MC simulator based on an 80×80 lattice model is used to describe the evolution of the film growth. The roughness set point value is 1.5 and the growth rate set point value is 220 ML/s. Initially, the substrate temperature is $T = 800$ K and the inlet precursor mole fraction is 2.0×10^{-5} ; these conditions correspond to a growth rate of about 180 ML/s and a surface roughness of about 1.8. The proposed multivariable control system (Figure 26) is applied to the multiscale process model to regulate the

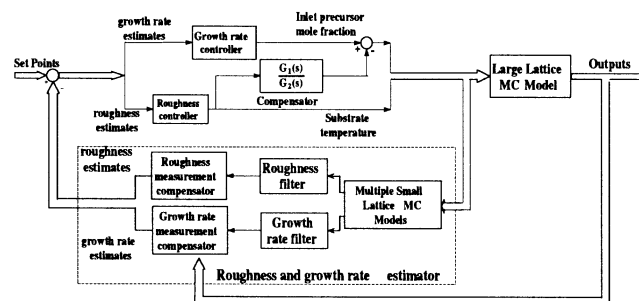


Figure 26. Multivariable feedback control system with interaction compensation.

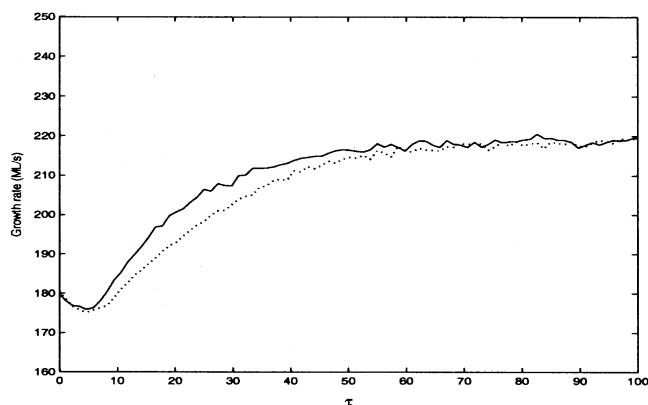


Figure 27. Comparison of the closed-loop growth rate under multivariable feedback control with interaction compensation (solid line) and under multiple single-loop control (dashed line).

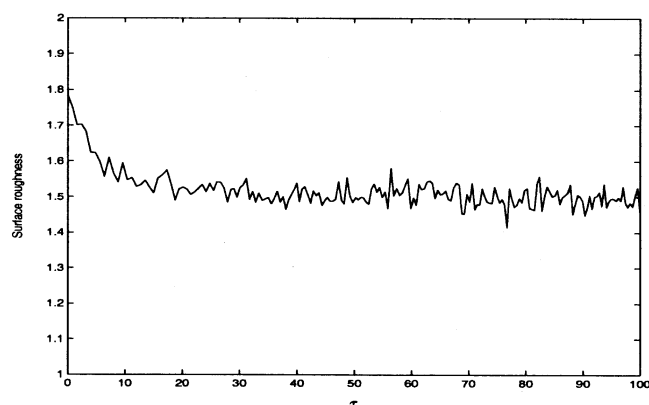


Figure 29. Closed-loop surface roughness under multivariable feedback control with interaction compensation.

growth rate and surface roughness to the desired set point values. Figure 27 shows the comparison of the growth rate profiles under multivariable feedback control with interaction compensation and under multiple single-loop control without interaction compensation. By using the interaction compensator, the growth rate converges to the desired set point value faster. Figure 28 shows the comparison of the inlet precursor mole fraction under multivariable control with interaction compensation and that obtained under multiple single-loop control. Figures 29 and 30 show the surface roughness under multivariable control with interaction compensation and the profile of the substrate temperature, respectively. The microstructure of the thin film at the beginning and at the end of the closed-loop system simulation run is shown in Figure 31 and Figure 32, respectively. These results show that the proposed multivariable control system with interaction compensation can simultaneously drive the growth rate and surface roughness to the desired set point values and improve closed-loop response.

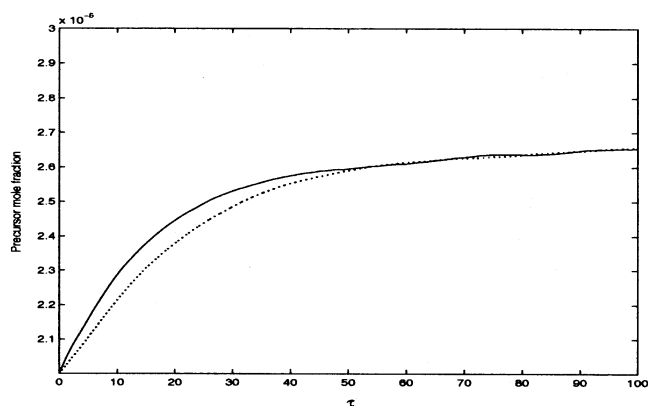


Figure 28. Comparison of inlet precursor mole fraction under multivariable feedback control with interaction compensation (solid line) and under multiple single-loop control (dashed line).

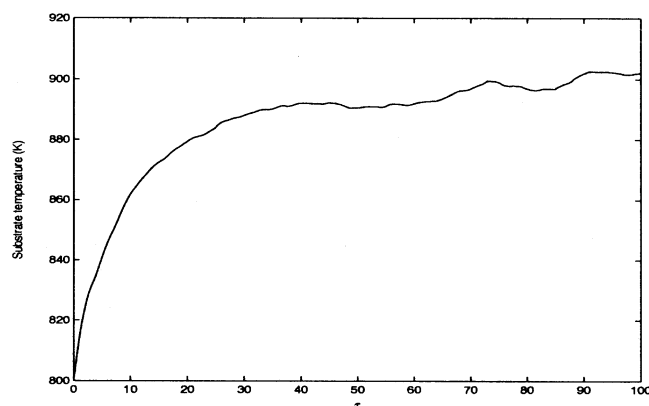


Figure 30. Substrate temperature under multivariable feedback control with interaction compensation.

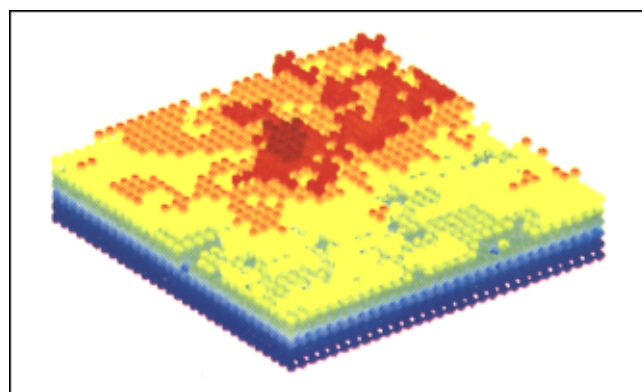


Figure 31. Surface microconfiguration at the beginning of the closed-loop simulation run (roughness = 1.8).

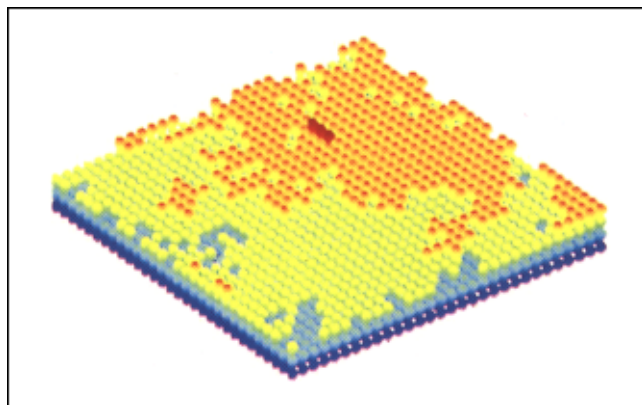


Figure 32. Surface microconfiguration at the end of the closed-loop simulation run (roughness = 1.5) under multivariable feedback control with interaction compensation.

Conclusions

This work focused on multivariable feedback control of surface roughness and growth rate in thin film growth in a stagnation flow geometry. The multivariable control problem was studied and a multivariable feedback control system, which can be implemented in real time, was proposed. The proposed control system uses an estimator that provides estimates of the surface roughness and growth rate at a time-scale comparable to the real-time evolution of the process, a multivariable interaction compensation block, and two proportional-integral feedback controllers. The control system was applied to the multiscale process model and was found to successfully regulate the surface roughness and growth rate to the desired set point values. While the proposed method for estimation/control using kinetic Monte-Carlo simulators was applied to the process of thin film growth in a stagnation flow geometry, the methodological approach of this work is applicable to other thin film growth processes.

Acknowledgments

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