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Multivariable Predictive Control of Thin Film Deposition Using a Stochastic PDE Model

Dong Ni and Panagiotis D. Christofides

Department of Chemical Engineering
University of California, Los Angeles, CA 90095-1592

Abstract— **In this work, we construct a 2-dimensional (2D) stochastic partial differential equation (PDE) model for a thin film deposition process and design a multivariable predictive controller based on the constructed model to control thin film thickness and surface roughness. We focus on a thin film deposition process governed by three microscopic processes including molecule adsorption, migration and desorption. A 2D linear stochastic PDE model is initially constructed following the methodology proposed in our previous work [18]. Then, a stochastic PDE model-based multivariable controller is designed using the constructed stochastic PDE model. The control problem is formulated as a predictive control problem, in which the constructed stochastic PDE model is used to predict both the thin film thickness and the surface roughness. Moreover, the controller design is performed based on a finite stochastic ordinary differential equation (ODE) approximation of the stochastic PDE model to achieve high computational efficiency. The model-based predictive controller is applied to the kinetic Monte-Carlo (kMC) simulation of the deposition process to simultaneously control the thin film thickness and surface roughness. Closed-loop system simulation results demonstrate that the model is adequately accurate and that the controller is effective.**

I. INTRODUCTION

Operating thin film deposition under real-time feedback control is one of the most prominent solutions to meet the increasingly stringent requirements on the quality of thin films and reduce thin film variability. Significant research efforts have been made on the feedback control of thin film deposition processes with emphasis on deposition spatial uniformity control (see [25], [3] for results on rapid thermal processing (RTP) and [1] on plasma-enhanced chemical vapor deposition (PECVD)) and on thin film composition control (see [21] for experimental results on real-time carbon content control in a PECVD process). More recently, there has been great attention on feedback control of thin film microstructure arising from the vast need of thin films with well-defined microstructure. In a thin film growth process, the film is directly shaped by microscopic random processes (e.g., molecule adsorption, desorption, migration and surface reaction). Models that describe these microscopic processes and directly account for their stochastic nature are needed for precise control of film microstructure. Examples of such models include: 1) kinetic Monte-Carlo (kMC) methods [9], [6], [10], and 2) stochastic partial differential equations (PDEs) [4], [27].

Methodologies for estimation-based feedback control and model-predictive control using kinetic Monte-Carlo models have recently been developed in [13], [14], [16] and [19], respectively. However, the fact that kMC models are not available in closed-form makes very difficult to use them for system-level analysis and the design and implementation of real-time model-based feedback control systems. Based on the fact that kinetic Monte-Carlo simulations provide realizations of a stochastic process which are consistent with the master equation that describes the evolution of the microscopic probability distribution, a method to construct reduced-order approximations of the master equation was reported in [8]. Furthermore, an approach was also reported in [24], [2] to identify linear deterministic models from outputs of kinetic Monte-Carlo simulators and design controllers using linear control theory. This approach is quite effective to control the macroscopic variables which are low statistical moments of the microscopic distributions (e.g., surface coverage, which is the zeroth moment of adspecies distribution on a lattice). However, to control higher statistical moments of the microscopic distributions, such as the surface roughness (the second moment of height distribution on a lattice), or even the microscopic configuration (such as the surface morphology), linear deterministic models may not be sufficient. This is because the effect of the stochastic nature of the microscopic processes becomes very significant in these cases and must be addressed both in the model construction and controller design.

Stochastic PDE models, on the other hand, which are available in closed-form, have been developed to describe the evolution of the height profile for surfaces in certain physical and chemical processes such as epitaxial growth [27] and ion sputtering [11]. More recently, Lou and Christofides presented a method for feedback control of surface roughness in a thin film growth process using 1-dimensional (1D) stochastic PDE models [17], [15]. Pole-placement controller design was carried out directly based on the stochastic PDE models which describe the surface height fluctuation, and the feedback controller was successfully applied to the kMC model of the process regulating the surface roughness to desired values.

In this work, we construct a 2-dimensional (2D) stochastic PDE model for a thin film deposition process and design a multivariable predictive controller based on the

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constructed model to control thin film thickness and surface roughness. We focus on a thin film deposition process governed by three microscopic processes including molecule adsorption, migration and desorption. A 2D linear stochastic PDE model is initially constructed following the methodology proposed in our previous work [18]. Then, a stochastic PDE model-based multivariable controller is designed using the constructed stochastic PDE model. The control problem is formulated as a predictive control problem, in which the constructed stochastic PDE model is used to predict both the thin film thickness and the surface roughness. Moreover, the controller design is performed based on a finite stochastic ordinary differential equation (ODE) approximation of the stochastic PDE model to achieve high computational efficiency. The model-based predictive controller is applied to the kMC simulation of the deposition process to simultaneously control the thin film thickness and surface roughness. Closed-loop system simulation results demonstrate that the model is adequately accurate and that the controller is effective.

II. PRELIMINARIES

In this work, we consider a thin film growth process of deposition from vapor phase, in which the formation of the thin film is governed by three microscopic processes that occur on the surface, i.e., the adsorption of vapor phase molecules on the surface, the migration of surface molecules and desorption of surface molecules. A kinetic Monte-Carlo simulation code following the algorithm reported in [26] is used to simulate the deposition process on a 2D lattice.

Without any *a priori* knowledge of the deposition process, we assume that there exists a 2D linear stochastic PDE of the following general form that can adequately describe the evolution of the surface of the thin film during the deposition:

$$\frac{\partial h}{\partial t} = c + c_1 \bigtriangledown h + c_2 \bigtriangledown^2 h + \cdots + c_w \bigtriangledown^w h + \xi(x,y,t) \quad (1)$$

where $x \in [0,\pi], y \in [0,\pi]$ is the spatial coordinate, t is the time, $h(x,y,t)$ is the height of the surface at position x, y and time t, and $\xi(x,y,t)$ is a Gaussian noise with zero mean and covariance:

$$\langle \xi(x,y,t)\xi(x',y',t')\rangle = \varsigma^2 \delta(x-x')\delta(y-y')\delta(t-t') \quad (2)$$

where $\delta(\cdot)$ is the Dirac function. Furthermore, the prederivative coefficients c and c_j in Eq.1 and the parameter ς^2 in Eq.2 depend on the process parameters: the substrate temperature T and adsorption rate W (directly determined by vapor phase concentration).

The stochastic PDE of Eq.1 is subjected to the following periodic boundary conditions:

$$\begin{aligned} &\bigtriangledown^j h(0,y,t) = \bigtriangledown^j h(\pi,y,t) \\ &\bigtriangledown^j h(x,0,t) = \bigtriangledown^j h(x,\pi,t) \quad j = 0,\cdots,w-1 \end{aligned} \quad (3)$$

and the initial condition:

$$h(x,y,0) = h_0(x,y) \quad (4)$$

To study the dynamics of Eq.1, we initially consider the eigenvalue problem of the linear operator of Eq.1, which takes the form:

$$\begin{aligned} A\phi_{m,n}(x,y) &= c_1 \bigtriangledown \phi_{m,n}(x,y) + c_2 \bigtriangledown^2 \phi_{m,n}(x,y) \\ &\quad + \cdots + c_w \bigtriangledown^w \phi_{m,n}(x,y) \\ &= \lambda_{m,n}\phi_{m,n}(x,y) \\ &\bigtriangledown^j \phi_{m,n}(0,y) = \bigtriangledown^j \phi_{m,n}(\pi,y) \\ &\bigtriangledown^j \phi_{m,n}(x,0) = \bigtriangledown^j \phi_{m,n}(x,\pi) \\ &j = 0,\cdots,w-1 \quad m,n = 0,\pm1,\cdots,\pm\infty \end{aligned} \quad (5)$$

where $\lambda_{m,n}$ denotes an eigenvalue and $\phi_{m,n}$ denotes an eigenfunction. A direct computation of the solution of the above eigenvalue problem yields:

$$\begin{aligned} \lambda_{m,n} &= (I2m + I2n)c_1 + [(I2m)^2 + (I2n)^2]c_2 + \cdots \\ &\quad + [(I2m)^w + (I2n)^w]c_w \\ \phi_{m,n}(x,y) &= \frac{1}{\pi}(e^{I2mx + I2ny}) \\ m,n &= 0,\pm1,\cdots,\pm\infty \end{aligned} \quad (6)$$

where $\lambda_{m,n}$ denotes the m,nth eigenvalue, $\phi_{m,n}(x,y)$ denotes the m,nth eigenfunction and $I = \sqrt{-1}$.

To present the method that we use for parameter identification of the stochastic PDE of Eq.1, we first derive an infinite stochastic ODE representation of Eq.1 using modal decomposition and parameterize the infinite stochastic ODE system using kMC simulation. We first expand the solution of Eq.1 in an infinite series in terms of the eigenfunctions of the operator of Eq.5 as follows (i.e., the Fourier expansion in the complex form):

$$h(x,y,t) = \sum_{m,n=-\infty}^{\infty} z_{m,n}(t)\phi_{m,n}(x,y) \quad (7)$$

where $z_{m,n}(t)$ are time-varying coefficients. Substituting the above expansion for the solution, $h(x,y,t)$, into Eq.1 and taking the inner product, the following system of infinite stochastic ODEs is obtained:

$$\begin{aligned} &\frac{dz_{m,n}}{dt} = \lambda_{m,n} z_{m,n} + c^z_{m,n} + \xi_{m,n}(t) \\ &m,n = 0,\pm1,\cdots,\pm\infty \end{aligned} \quad (8)$$

and the initial conditions:

$$z_{m,n}(0) = z_{m,n,0} \quad m,n = 0,\pm1,\ldots,\pm\infty \quad (9)$$

where $c^z_{m,n} = c\int_0^\pi \int_0^\pi \phi^*_{m,n}(x,y)dxdy$, $\xi_{m,n}(t) = \int_0^\pi \int_0^\pi \xi(x,y,t)\phi^*_{m,n}(x,y)dxdy$ and $z_{m,n,0} = \int_0^\pi \int_0^\pi h_0(x,y)\phi^*_{m,n}(x,y)dxdy$. Apparently, $c^z_{0,0} = \pi c$ and $c^z_{m,n} = 0$ when $m^2 + n^2 \neq 0$. Furthermore, $\langle \xi_{m,n}(t)\rangle = 0$ and $\langle \xi_{m,n}(t)\xi^*_{m,n}(t')\rangle = \varsigma^2\delta(t-t')$ ($\xi^*_{m,n}$ is the complex conjugate of $\xi_{m,n}$, the superscript star is used to denote complex conjugate in the remainder of this manuscript).

To parameterize this infinite stochastic ODE system, we first derive the analytic expressions for the statistical

moments of the stochastic ODE states, such as the expected values and covariances. By comparing the analytical expression to the statistical moments obtained by multiple kMC simulations, the parameters of the stochastic ODE system (i.e., $\lambda_{m,n}$ and ς) can be determined.

The analytic solution to Eq.8 is obtained as follows to derive the expressions for the statistical moments of the stochastic ODE states:

$$z_{m,n}(t) = e^{\lambda_{m,n}t} z_{m,n,0} + \frac{(e^{\lambda_{m,n}t} - 1)c^z_{m,n}}{\lambda_{m,n}} + \int_0^t e^{\lambda_{m,n}(t-\mu)} \xi_{m,n}(\mu) d\mu \quad (10)$$

III. FEEDBACK CONTROL

In this section, we design a model-based feedback controller to control the thin film thickness and surface roughness of the deposition process. The difficulty of obtaining in-situ surface measurements in real-time had been one of the obstacles for implementing feedback control system on thin film processes. Recently, researchers made possible to use some of the intrusive scanning probe based techniques such as the scanning tunneling microscopy (STM) [5], secondary electron microscopy (SEM) [12] and atomic force microscopy (AFM) [23] in-situ, to observe in real-time the growth of the thin film. More recently, it was reported in [22] that a non-intrusive grazing incidence small angle x-ray scattering (GISAXS) method was successfully used to monitor the thin film growth in-situ in real-time; the method was capable of sampling large surface areas with sampling frequency up to 10 Hz and a subnanometer resolution. Such advancements in surface metrology indeed opens up the possibility for implementing feedback control systems which rely on real-time surface state measurements.

Based on the results shown in the previous section, we extend the model construction method developed in our previous work [18] to the 2D case and construct a 2D stochastic PDE model for the deposition process (see [20] for detailed discussion). The linear stochastic PDE model identified for the deposition process is as follows (A detailed comparison of the kMC simulation and the stochastic PDE model with qualitative validation in terms of thickness and surface roughness can also be found in our full paper [20]):

$$\begin{aligned} \frac{\partial h}{\partial t} &= W\left(1 - \frac{k_w}{W^{a_w} e^{-\frac{k_B T}{E_w}}}\right) \\ &+ W\left(1 - \frac{k_w}{W^{a_w} e^{-\frac{k_B T}{E_w}}}\right) \bigtriangledown^2 h + \xi(x,y,t) \\ & \bigtriangledown h(0,y,t) = \bigtriangledown h(\pi,y,t),\ h(0,y,t) = h(\pi,y,t) \\ & \bigtriangledown h(x,0,t) = \bigtriangledown h(x,\pi,t),\ h(x,0,t) = h(x,\pi,t) \\ & h(x,y,0) = h_0(x,y) \end{aligned} \quad (11)$$

where $k_w = 3.3829 \times 10^{-12}$, $a_w = 0.6042$, $E_w = 2.7 \times 10^{-3}\ eV$, $k_c = 1.0274 \times 10^{-13}$, $a_c = 0.1669$, $E_c = 1.9 \times 10^{-3}\ eV$, $a_v = 15.55493$, $k_v = 20.64504$, $a_t = 0.02332$ and $k_t = 0.0261$. Furthermore, $\langle \xi(x,y,t)\xi(x',y',t') \rangle = \frac{\pi^2}{k^2_{max}} W[1 + \frac{e^{(a_t + k_t W)T}}{e^{a_v} + k_v W}]\delta(x-x')\delta(y-y')\delta(t-t')$.

A. Feedback control design

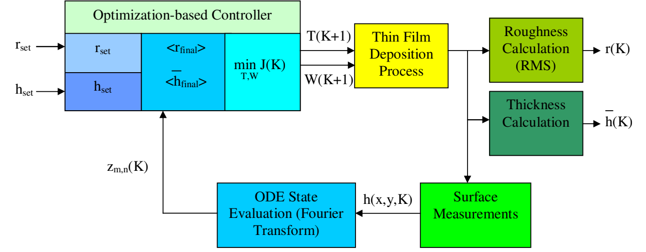

Fig. 1. Block diagram of the closed-loop system.

We now proceed with the design of the feedback controller. Since the thin film deposition is a batch process, the control objective is to control the final thin film thickness and surface roughness to the desired levels at the end of each deposition run. We use an optimization-based control problem formulation. The substrate temperature T, and the adsorption rate W (W can be adjusted by varying reactor inlet gas flow rate, chamber pumping speed, etc.) are chosen to be the manipulated variables. Furthermore, since the process is stochastic in nature, the controlled variable is the expected value of the final thin film thickness $\langle \bar{h}(t_{dep}) \rangle$ and surface roughness $\langle r(t_{dep}) \rangle$, where t_{dep} is the total deposition time.

Fig.1 shows the block diagram of the closed-loop system. The control system operates in a discrete time fashion, when a real-time surface profile measurement is obtained at time K (i.e., $t = Kt_s$, where t_s is the measurement interval as well as the control interval), the states of the infinite stochastic ODE system, $z_{m,n}(K)$, are computed. Then, a substrate temperature $T(K+1)$ and an adsorption rate $W(K+1)$ are computed based on states $z_{m,n}(K)$ and the stochastic PDE model, under the assumption that T and W are held at designated levels for the rest of the deposition. $T(K+1)$ and $W(K+1)$ are then applied to the deposition process at next measurement time $K+1$.

In order to design a model-based predictive controller, we first derive the analytical expression for the trajectory of $\langle \bar{h}(t) \rangle$ and $\langle r(t) \rangle$. The film thickness $\bar{h}(t)$ can be computed based on the ODE state as $z_{0,0}(t)\phi_{0,0}$ (see detailed discussion in [18]). The surface roughness $r(t)$, defined in the root-mean-square fashion, can be rewritten in terms of $z_{m,n}$ as follows (see detailed discussion in [18]):

$$r(t) = \sqrt{\frac{1}{\pi^2} \sum_{m,n=-\infty, m^2+n^2 \neq 0}^{\infty} z_{m,n}(t) z^*_{m,n}(t)} \quad (12)$$

Due to the fact that the current deposition parameters ($T(K)$ and $W(K)$) would be used during the current control cycle before the new levels ($T(K+1)$ and $W(K+1)$)

are applied, the estimate of the film thickness (i.e., the estimate of $\langle z_{0,0}(t_{dep}\rangle)$ and the estimate of the final surface roughness cannot be computed directly ($\lambda_{m,n}$ and ς^2 are no longer constant due to the change of W and T). Therefore, we first need to derive the expressions of $z_{0,0}(t_{dep})$ and $z_{m,n}(t_{dep})$ ($m^2+n^2 \neq 0$) for this case. We consider such case that, at time $(K+1)$, the deposition parameters are changed from $W(K)$ and $T(K)$ to $W(K+1)$ and $T(K+1)$, respectively. Following Eq.10, by calculating the intermediate values $z_{0,0}(K+1)$ and $z_{m,n}(K+1)$ ($m^2+n^2 \neq 0$) using $z_{0,0}(K)$ and $z_{m,n}(K)$ respectively, and substituting $c^z_{0,0} = \pi c$ and $\lambda_{m,n} = -4(m^2+n^2)c_2$ ($m^2+n^2 \neq 0$, Eq.6), the expressions of $z_{0,0}(t_{dep})$ and $z_{m,n}(t_{dep})$ can be derived as follows:

$$\begin{aligned} z_{0,0}(t_{dep}) =& z_{0,0}(K) + \pi c(K)t_c + \bar{\theta}_{0,0}(K) + \hat{\theta}_{0,0}(K+1) \\ &+\pi c(K+1)[t_{dep}-(K+1)t_c] \\ z_{m,n}(t) =& z_{m,n}(K) \\ \times & e^{-4(m^2+n^2)\{c_2(K)t_c + c_2(K+1)[t_{dep}-(K+1)t_c]\}} \\ &+\bar{\theta}_{m,n}(K) + \hat{\theta}_{m,n}(K+1) \\ & m,n = 0,\pm 1,\cdots,\pm\infty \quad m^2+n^2 \neq 0 \end{aligned} \tag{13}$$

where $\bar{\theta}_{0,0}(K)$, $\hat{\theta}_{0,0}(K+1)$, $\bar{\theta}_{m,n}(K)$, and $\hat{\theta}_{m,n}(K+1)$ ($m^2+n^2 \neq 0$) are independent Gaussian random numbers with zero mean, and their covariances can be expressed as follows:

$$\begin{aligned} &\langle \bar{\theta}_{0,0}(K)\bar{\theta}^*_{0,0}(K)\rangle = \varsigma^2(K)t_c \\ &\langle \hat{\theta}_{0,0}(K+1)\hat{\theta}^*_{0,0}(K+1)\rangle = \varsigma^2(K+1)[t_{dep}-(K+1)t_c] \\ &\langle \bar{\theta}_{m,n}(K)\bar{\theta}^*_{m,n}(K)\rangle = e^{-8(m^2+n^2)c_2(K+1)t_c}\varsigma^2(K) \\ &\qquad \times \frac{e^{-8(m^2+n^2)c_2(K)t_c}-1}{-8(m^2+n^2)c_2(K)} \\ &\langle \hat{\theta}_{m,n}(K+1)\hat{\theta}^*_{m,n}(K+1)\rangle = \varsigma^2(K+1) \\ &\qquad \times \frac{e^{-8(m^2+n^2)c_2(K+1)[t_{dep}-(K+1)t_c]}-1}{-8(m^2+n^2)c_2(K+1)} \\ & m,n = 0,\pm 1,\cdots,\pm\infty \quad m^2+n^2 \neq 0 \end{aligned} \tag{14}$$

Therefore, the quantities that directly relate to the thickness and the roughness estimations, $\langle z_{0,0}(t_{dep})\rangle$, $\langle z_{m,n}(t_{dep})\rangle$, and $\langle z_{m,n}(t_{dep})z^*_{m,n}(t_{dep})\rangle$, can be derived as follows:

$$\begin{aligned} &\langle z_{0,0}(t_{dep})\rangle = z_{0,0}(K) + \pi c(K)t_c \\ &\qquad +\pi c(K+1)[t_{dep}-(K+1)t_c] \\ &\langle z_{m,n}(t_{dep})\rangle = z_{m,n}(K) \\ &\times e^{-4(m^2+n^2)\{c_2(K)t_c + c_2(K+1)[t_{dep}-(K+1)t_c]\}} \\ &\langle z_{m,n}(t_{dep})z^*_{m,n}(t_{dep})\rangle = \langle z_{m,n}(t_{dep})\rangle\langle z_{m,n}(t_{dep})\rangle^* \\ &\qquad +\langle \bar{\theta}_{m,n}(K)\bar{\theta}^*_{m,n}(K)\rangle \\ &\qquad +\langle \hat{\theta}_{m,n}(K+1)\hat{\theta}^*_{m,n}(K+1)\rangle \\ & m,n = 0,\pm 1,\cdots,\pm\infty \quad m^2+n^2 \neq 0 \end{aligned} \tag{15}$$

Accordingly, the expected final film thickness can be expressed as follows:

$$\begin{aligned} \langle \bar{h}_{final}(K)\rangle =& \frac{z_{0,0}(K)}{\pi} \\ &+c(K)t_c + c(K+1)[t_{dep}-(K+1)t_c] \end{aligned} \tag{16}$$

and a finite approximation, which only uses the first ($\pm N$th,$\pm N$th) states, of the expected final surface roughness can be derived as follows,

$$\begin{aligned} \langle r_{final}(K)\rangle^2 =& \frac{1}{\pi^2}\sum_{m,n=-N;\ m^2+n^2\neq 0}^{N} [\langle z_{m,n}(t_{dep})\rangle\langle z_{m,n}(t_{dep})\rangle^* \\ &+\langle \bar{\theta}_{m,n}(K)\bar{\theta}^*_{m,n}(K)\rangle \\ &+\langle \hat{\theta}_{m,n}(K+1)\hat{\theta}^*_{m,n}(K+1)\rangle] \end{aligned} \tag{17}$$

Therefore, the value of $T(K+1)$ and $W(K+1)$ are determined at each time by solving the following optimization problem:

$$\begin{aligned} \min_{W(K+1),T(K+1)} J(K) \ =& \ q_h(h_{set} - \langle \bar{h}_{final}(K)\rangle)^2 \\ &+q_r(r^2_{set} - \langle r_{final}(K)\rangle^2)^2 \end{aligned} \tag{18}$$

subject to,

$$\begin{aligned} \langle r_{final}(K)\rangle^2 =& \frac{1}{\pi^2}\sum_{m,n=-N;\ m^2+n^2\neq 0}^{N} \{z_{m,n}(K)z^*_{m,n}(K) \\ & e^{-8(m^2+n^2)\{c_2(K)t_c + c_2(K+1)[t_{dep}-(K+1)t_c]\}} \\ &+e^{-8(m^2+n^2)c_2(K+1)t_c}\varsigma^2(K) \\ &\qquad \frac{e^{-8(m^2+n^2)c_2(K)t_c}-1}{-8(m^2+n^2)c_2(K)} \\ &+\varsigma^2(K+1) \\ &\qquad \frac{e^{-8(m^2+n^2)c_2(K+1)[t_{dep}-(K+1)t_c]}-1}{-8(m^2+n^2)c_2(K+1)}\} \end{aligned} \tag{19}$$

$$\begin{aligned} \langle \bar{h}_{final}(K)\rangle \ =& \ \frac{z_{0,0}(K)}{\pi} + c(K)t_c \\ &+c(K+1)[t_{dep}-(K+1)t_c] \end{aligned} \tag{20}$$

$$\begin{aligned} c(K+1) =& W(K+1) \\ &-\frac{k_w}{W(K+1)^{a_w-1}e^{-\frac{k_BT(K+1)}{E_w}}} \end{aligned} \tag{21}$$

$$c_2(K+1) = \frac{k_c}{k^2_{max}W(K+1)^{a_c}e^{-\frac{k_BT(K+1)}{E_c}}} \tag{22}$$

$$\begin{aligned} \varsigma^2(K+1) =& \frac{\pi^2}{k^2_{max}}W(K+1)\{1 \\ &+\frac{e^{[a_t+k_tW(K+1)]T(K+1)}}{e^{a_v+k_vW(K+1)}}\} \end{aligned} \tag{23}$$

$$c(K+1) \geq c_{min} \tag{24}$$

$$T_{min} \leq T(K+1) \leq T_{max} \tag{25}$$

$$W_{min} \leq W(K+1) \leq W_{max} \tag{26}$$

where q_h and q_r are the weights of the penalties on thickness and roughness respectively (we pick $q_h = 1/h_{set}^2$ and $q_r = 1/r_{set}^2$ in this study), c_{min} is the minimum growth rate (we pick $c_{min} = 0.1h_{set}/t_{dep}$ in this study), T_{min} and T_{max} are the lowest and highest substrate temperature, respectively (we pick $T_{min} = 400\ K$ and $T_{max} = 900\ K$ in this study), and W_{min} and W_{max} are the lowest and highest adsorption rate, respectively (we pick $W_{min} = 0.1\ s^{-1}$ and $W_{max} = 2.0\ s^{-1}$ in this study). We note that J corresponds to the difference between the square of the desired final surface roughness r_{set} and the square of the estimated final surface roughness $\langle r_{final} \rangle$ computed based on the current states $z_{m,n}(t_K)$. We choose to minimize the difference of the squares of the surface roughness, i.e., the mean square of the surface height, to simplify the calculation. The optimization problem is solved using a standard sequential quadratic programming (SQP) method described in [7].

Remark 1: Since Eq.19, is a finite approximation of the predicted final surface roughness, to achieve a control precision ϵ, m should be chosen large enough for each optimization computation so that the approximation error is less than ϵ (see detailed discussion in [18]).

Remark 2: Since the control action is computed using closed-form equations, the computation cost is proportional to the number of states used, $4N^2$, but independent of the optimization horizon $t_{dep} - t$; however, to evaluate the values of the $4N^2$ states, an additional computation time of the order of $4k_{max}^2 N^2$ is needed for each surface measurement. Nevertheless, even for a lattice size that corresponds to the largest physical dimension of the sampling area that can be achieved by common surface measurement techniques (i.e., a few microns), such computation can still be completed within the control interval using currently available computing power. On the other hand, such task is almost impossible to achieve using a kMC code, whose computation cost is on the order of $k_{max}^4(t_{dep} - t)$ for merely a single run. Furthermore, we note that the evaluation of each state is independent of other states, and therefore, can be executed in parallel, while the kMC code, being a serial calculation, is unsuitable for parallel processing.

B. Closed-loop simulations

A kMC code with a lattice size 100×100 is used to simulate the thin film deposition process. The measurement interval, as well as the control interval, is set to be $1\ s$. The highest order of the states of the stochastic ODEs used for optimization is $N = 10$.

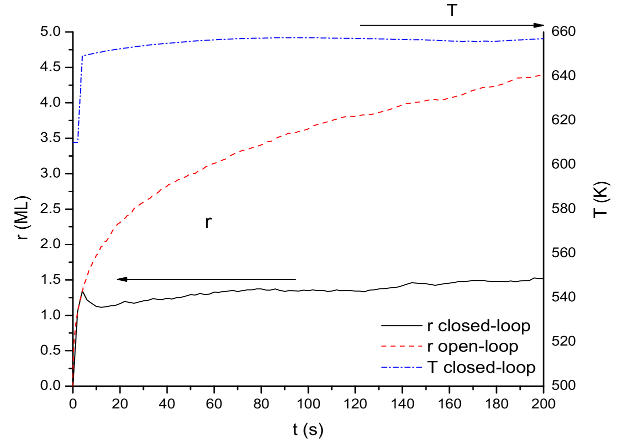

Fig. 2. Surface roughness and substrate temperature profiles of a 200 s closed-loop deposition process with a thickness setpoint of $100ML$ and a final roughness setpoint $r_{set} = 1.5ML$; the initial deposition conditions are $T = 610K$ and $W = 1.0s^{-1}$.

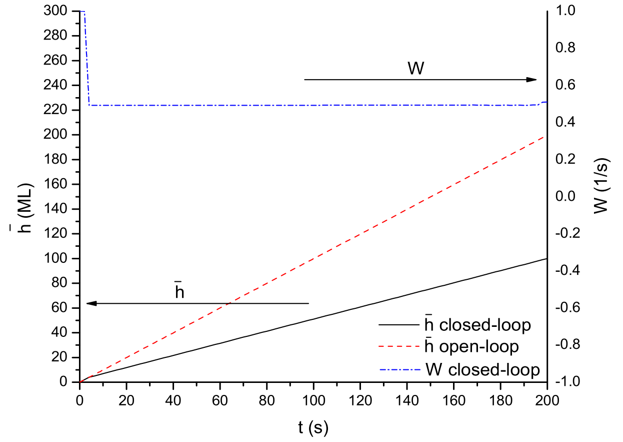

Fig. 3. Thickness and surface adsorption rate profiles of a 200 s closed-loop deposition process with a thickness setpoint of $100ML$ and a final roughness setpoint $r_{set} = 1.5ML$, the initial deposition conditions are $T = 610K$ and $W = 1.0s^{-1}$.

Fig.2 shows the surface roughness and substrate temperature profiles of a closed-loop deposition with initial substrate temperature $T = 610\ K$ and adsorption rate $W = 1.0\ s^{-1}$ and of an open-loop deposition with $T = 610\ K$ and $W = 1.0\ s^{-1}$. Fig.3 shows the thin film thickness and surface adsorption rate profiles of this closed-loop deposition and of open-loop deposition. The control objective is to control the thin film thickness to 100 $monolayers$ and to drive the final surface roughness to 1.5 $monolayers$ at the end of the 200 s deposition run. It can be seen that both the film thickness and the final surface roughness are controlled at the desired levels simultaneously while an open-loop deposition with the same initial deposition condition would lead to a 100% higher film thickness and a 100% higher final surface roughness as shown in Fig.2

and Fig.3.

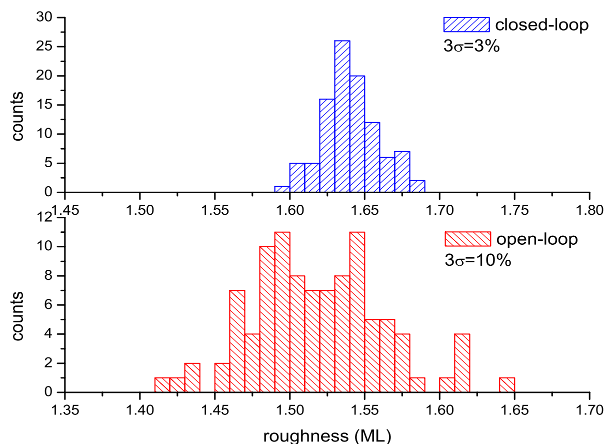

Fig. 4. Histogram of final surface roughness of 100 closed-loop and 100 open-loop thin film depositions targeted at the same surface roughness level.

Fig.4 shows the final surface roughness histogram of the thin films deposited using 100 different closed-loop depositions targeting thin film thickness of 100 $monolayers$ and final surface roughness of 1.65 $monolayers$ and 100 different open-loop depositions with fixed substrate temperature and surface adsorption rate. The average roughness of the thin films deposited by open-loop depositions is 1.52 $monolayers$, which is quite close to the average roughness of the thin films deposited by the closed-loop depositions (1.64 $monolayers$). However, the variance among the thin films from different open-loop deposition runs is over 300% higher than that of closed-loop depositions even though no process disturbance is considered in the simulations. This result suggests that, besides controlling the film thickness and surface roughness for the deposition, the proposed controller is also able to compensate for the stochasticity of the thin film growth process and reduced the variance among the thin films prepared by different deposition runs.

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