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Stochastic Modeling for Deposition Process of Thin Film

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Abstract The deposition process which involves atom adsorption and migration(surface diffusion) is modeled via kinetic Monte Carlo(KMC) simulation in 3 dimensional space. In this paper, suitable physical quantities such as partial film sites occupancy ratio(PSOR), roughness, mass fractal dimension are introduced to describe the structure of film with porosity and the time evolution of film structure is mathematically modeled by stochastic differential equation and the coefficients of this equation are estimated on the basis of data obtained from the KMC simulation using least-square methods. Simulation results by the solution of stochastic differential equation show that the proposed mathematical model is enough to describe the deposition process of film and to demonstrate the possibility of the predictive control in the film deposition process.

Key words film deposition, stochastic differential equation, kinetic Monte Carlo simulation

Introduction

Microstructure of thin film including surface roughness and internal porosities, are important factors to estimate the quality of film. The previous researches[3, 4] on modeling and control of microstructure of thin film have focused on surface roughness of thin film. KMC methods[2, 5, 6] and stochastic differential equation(SDE)models[1, 4], have been developed to describe the evolution of film microstructure. However, KMC models are not suitable as model for control of deposition process thin film and SDE models including stochastic nature of the growth processes are suitable, but it is difficult to determine the coefficients of the equations, which are associated with practical physical processes. In the previous study [1], the method for modeling the deposition process of thin film by using SDEs and determining the coefficients of the equation on the basis of data of KMC simulation is introduced, but the deposition process of thin film was not considered in 3 dimension and the accuracy of solution of SDE with determined coefficients have not been considered.

In this paper, the deposition process of Fe film is modeled via kinetic Monte Carlo(KMC) simulation in 3 dimensional space. Physical quantities, such as partial film sites occupancy ratio(PSOR), roughness, mass fractal dimension are introduced to describe the structure of film with porosity and the time evolution of film structure is mathematically modeled by stochastic differential equation and the coefficients of this equation are estimated on the basis of data obtained from the KMC simulation using least-square methods. Simulation results by the solution of SDE shows that the proposed mathematical model is preferable to describe the film deposition process and to demonstrate the possibility of the predictive control in the film deposition process.

1. KMC Model for Deposition Process of Thin Film

In this paper, the growth of thin film of α-Fe (lattice constant $a_0 = 0.34\text{nm}$), which has body-centered cubic structure below 900℃, is considered.

Both of the adsorption and migration processes are contained in deposition process of thin film.

All particles are modeled as identical hard spheres and the center of particles deposited on the film are to place at the lattice sites. The new particles are always deposited from the top side of the lattice where the gas phase is assumed to be present. Particle deposition results in film growth along the direction normal to the lateral direction (z axis). The number of the nearest neighbors of deposited particles on the thin film ranges from 0 to 14, the coordination number of the body-centered cubic lattice. The particles on the film are allowed to diffuse to an unoccupied neighboring site with a appropriate probability that depends on its local environment. The substrate is initially set to be fully packed and fixed. There are no vacancies in this layer and the particles in this layer cannot migrate.

1.1. Adsorption process

In an adsorption process, an incident particle comes in contact with the film and is incorporated onto the film. In this paper, the microscopic adsorption rate W, which is in units of layers per unit time, is treated as a process parameter.

The incident particles are initially placed at random positions above the film lattice and move toward the lattice in random directions. The position of initial particle is uniformly distributed in the considered domain. The angle θ which is defined as the angle between the incident direction and the z-axis is chosen to be uniform in the interval $(0,\ \pi/2)$ and the angle φ which is defined as the angle between the incident direction and the x-axis in the interval $(0,\ 2\pi)$.

After the initial position and incident direction are determined, the incident particle A, travels along a straight line towards the film until contacting the first particle B, on the film. Upon contact, particle A stops and sticks to particle B at the contacting position. Then particle A moves (relaxes) to the nearest vacant site among the neighboring sites of particle B.

1.2. Migration process(surface diffusion)

In a migration process, a particle overcomes the potential energy barrier and jumps to its vacant neighboring site. The migration rate of the i^{th} particle is calculated as follows [1].

$$r_{m,\,i} = v_0 \exp\left(-\frac{n_i E_0}{k_B T}\right) \tag{1}$$

where v_0 denotes a frequency of lattice thermal vibration, n_i is the number of the nearest neighbors of the i^{th} particle and can take the values from 1 to 13. E_0 is the contribution to the activation energy barrier from each nearest neighbor, k_B is the Boltzmann's constant and T is the substrate temperature of the thin film. Since the film is thin, the temperature is assumed to be uniform throughout the film. In this work, the values $v_0 = 10^{13}\text{s}^{-1}$, $E_0 = 0.47\text{eV}$

are taken, which are appropriate for a Fe film.

A particle can jump to either of its vacant neighboring sites with equal probability, but cannot jump off the film. It can only migrate on the surface.

2. KMC Simulation

2.1. Physical quantities to estimate the film structure

2.1.1. Partial film site occupancy ratio (PSOR)[1]

PSOR is defined as follows

$$\rho_{\mathrm{p}} = \frac{N_{\mathrm{p}}}{N_0 H_{\mathrm{p}}} \tag{2}$$

where ρ_{p} denotes PSOR, N_{p} denotes the number of particles from the top to H_{p} layers, H_{p} denotes the number from top layers of the film to considering layer and N_0 denotes the maximum value of the particles in a layer.

The choice of H_{p} affects the value of the PSOR, ρ_{p}. Specifically, PSOR cannot be correctly calculated when $H < H_{\mathrm{p}}$, but this problem is bypassed by assuming the existence of H_{p} fully-packed substrate layers in the film before the deposition process begins. Therefore, at the beginning of deposition, the PSOR starts from value 1 since all H_{p} layers are substrate layers and are fully occupied. PSOR is considered to be a physical quantity to describe the film structure in the vicinity of surface. In the simulation, the value of H_{p} is chosen to be 10.

2.1.2. Roughness

Roughness which is considered to be a physical quantity to describe the rough surface of film is defined as follows.

$$R = \sqrt{<(h-\overline{h})^2>} \tag{3}$$

where <…> denotes the average(mean) value.

2.1.3. Mass fractal dimension

The mass fractal dimension is defined as a physical quantity to describe a irregular and complex structure of film and the time evolution of film structure is estimated by it.

The mass of film with the porosities is estimated as follows.

$$M = m_0 \left(\frac{L}{a_0} \right)^{D_m} \tag{4}$$

where m_0 denotes the mass of a particle, a_0 the size of a particle, L the linear size of the growth film and D_m the mass fractal dimension.

Therefore, the mass fractal dimension is defines as follows.

$$D_m = \frac{\ln(M/m_0)}{\ln(L/a_0)} \tag{5}$$

2.2. Results of KMC simulation

The simulation input parameters are as follows.

The size of simulation cell $100a_0 \times 100a_0 \times 500a_0$, temperature of substrate $T = 600^\circ\mathrm{C}$, adsorption rate $W = 1\mathrm{layer/s}$, simulation time $t = 100\mathrm{s}$.

100 independent simulation runs are carried out to obtain the expected value and the variance of the physical quantities with time .

The grown film and its surface morphology when t=100s is shown in Fig. 1.

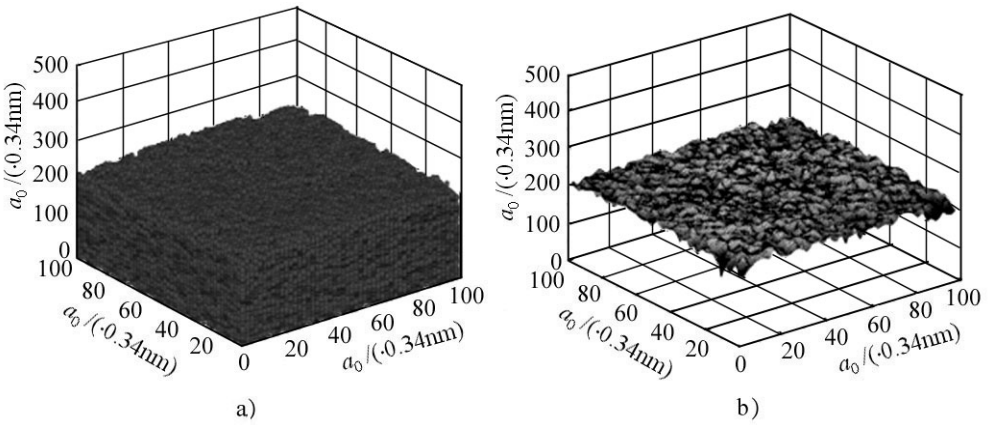

Fig. 1. The structure of grown film(a) and its surface morphology(b)

As shown in Fig. 1, it can be shown that lots of internal porosities are found in the film, and the surface structure of film is rough.

3. Nonhomogeneous Linear Stochastic Differential Equation for the Deposition Process of Thin Film

3.1. Stochastic differential equation and its analytical solution for the deposition process

The kinetic model to describe the evolution of film structure mathematically is needed in order to control the deposition process of film. Since the time evolution of physical quantities is random, these are treated as stochastic processes and these are described by nonhomogeneous linear stochastic differential equation.

The nonhomogeneous linear stochastic differential equation to describe the evolution of the film structure is as follows.

$$dA(t) = -\frac{1}{\tau}A(t)dt + \frac{A^{ss}}{\tau}dt + \beta\, dW(t) \tag{6}$$

where A^{ss}, τ are two of model parameters, which denote the steady-state value of the physical quantity and the time constant. The parameter β, denotes the intensity of stochastic factor, and $W(t)$ is Wiener process with the following expressions for its mean and covariance:

$$< W(t) >= 0, \quad < W(t)W(t') >= \delta(t-t'), \quad \delta(t-t') = \begin{cases} 1(t=t') \\ 0(t \neq t') \end{cases} \tag{7}$$

The model parameters A^{ss}, τ, β are the coefficients which are related to the deposition conditions. The stochastic differential equation is subject to the following initial condition.

$$A(t_0) = A_0 \tag{8}$$

where A_0 is the initial value of the physical quantity.

The solution of the nonhomogeneous linear stochastic differential equation can be obtained as follows.

$$A(t) = A^{ss} + (A_0 - A^{ss})e^{-(t-t_0)/\tau} + \beta\int_{t_0}^{t} e^{-(t-s)/\tau}dW(s) \tag{9}$$

As a result, $A(t)$ is a stochastic process, the expected value and variance of which, $< A(t) >$ and $\mathrm{Var}(A(t))$ can be obtained as follows.

$$< A(t) >= A^{ss} + (A_0 - A^{ss})e^{-(t-t_0)/\tau} \tag{10}$$

$$\mathrm{Var}(A(t)) = \frac{\tau\beta^2}{2}(1 - e^{-2(t-t_0)/\tau}) \tag{11}$$

3.2. Parameter estimation

Parameters A^{ss} and τ can be estimated from the solutions of the equation and the KMC simulation data by solving the following least-square optimization problem.

$$\min_{A^{ss},\,\tau}\sum_{i=1}^{m}\{< A(t_i) > -[A^{ss} + (A_0 - A^{ss})e^{-(t-t_0)/\tau}]\}^2 \tag{12}$$

where m is the number of the data pairs, t_i, $< A(t_i) >$ are values from the KMC simulations.

β can similarly obtain by solving the following least-square optimization problem.

$$\min_{\beta}\sum_{i=1}^{m}\left[\mathrm{Var}\,(A(t_i)) - \frac{\tau\beta^2}{2}(1 - e^{-2(t-t_0)/\tau})\right]^2 \tag{13}$$

The coefficients which are estimated from the data obtained from the KMC simulation are as follows.

$$\begin{aligned} &R^{ss} = 7.118\,6\,\mathrm{lay}, \quad \tau_R = 10.594\,7\mathrm{s}, \quad \beta_R = 0.269\,2\,\mathrm{lay}\cdot\mathrm{s}^{-1} \\ &PSOR^{ss} = 0.029\,1, \quad \tau_{PSOR} = 0.990\,5\mathrm{s}, \quad \beta_{PSOR} = 0.029\,3\mathrm{s}^{-1} \\ &D_m{}^{ss} = 2.680\,7, \quad \tau_{D_m} = 13.896\,0\mathrm{s}, \quad \beta_{D_m} = 0.008\,5\mathrm{s}^{-1} \end{aligned} \tag{14}$$

4. Results of the Stochastic Model for Deposition Process of Thin Film

4.1. Computational scheme on the solution of stochastic differential equation

The following computational scheme is used to find the solution of the stochastic differential equation.

$$A(t + \Delta t) = A(t)e^{-\Delta t/\tau} + A^{ss}(1 - e^{-\Delta t/\tau}) + \sqrt{\frac{\tau\beta^2}{2}(1 - e^{-\Delta t/\tau})}\;\xi_0 \tag{15}$$

where ξ_0 is normal random variable with the following expressions for its mean and variance: $<\xi_0>=0$, $\mathrm{Var}(\xi_0)=1$.

4.2. Comparison the results obtained from the stochastic model with the data obtained from KMC simulation

The time-evolutions of PSOR, roughness and mass fractal dimension are shown in Fig. 2－4.

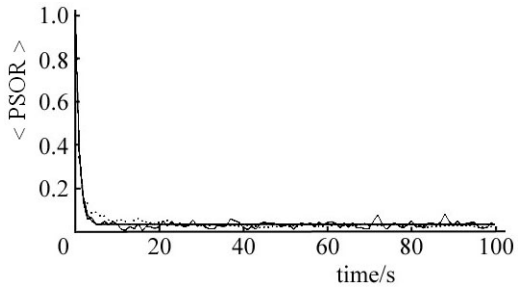

Fig. 2. Time-evolution of the PSOR
Point line–result of KMC simulation,
real line–result of SDE simulation

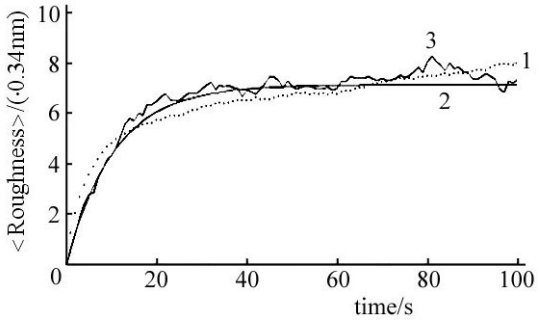

Fig. 3. Time-evolution of the Roughness
1－simulation result of KMC, 2－average value,
3－solution result of stochastic differential equation

In Fig. 2－4, the smooth curve(results of SDE simulation) denotes the evolution of the average value of physical quantity obtained from the solution of the stochastic differential equation and the broken curve denotes the calculation results by the computational scheme of the solution of stochastic differential equation.

As shown in Fig. 2, the simulation result is very similar to one of the previous research [1].

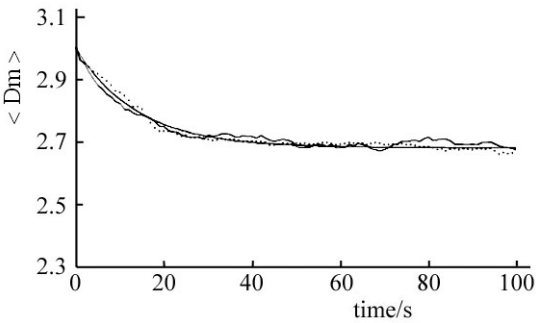

Fig. 4. Time-evolution of the mass fractal dimension
Point and real lines are equal to Fig. 2.

As shown in Fig. 3, the average value of roughness increases gradually with time at the beginning of deposition and remains unchanged, especially at increased time. This behavior describes the stages of thin film growth well. And, the results obtained by the stochastic model don't agree with one from KMC simulation completely, but it demonstrates the chaotic characteristics of roughness.

As shown in Fig. 4, the mean value of mass fractal dimension starts from 3.0 at the fully-packed substrate layers before the deposition process begins and decreases with time at the initial stage and then approaches constant value at increased time. This shows existence of the internal porosities in the film and the structural property remains unchanged with increased time.

Conclusion

It is shown that the stochastic model for the deposition process of thin film is preferable to describe the physical process as a random process by comparing the results of the stochastic model with the previous research and the data from KMC simulation.

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