Effects of Vapour Flowing Rates and Concentration on Surface Growth at Nano–thickness Range: Kinetic Monte Carlo Simulations

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ABSTRACT

In this study, we use the kinetic Monte Carlo simulation to investigate the growth dynamic of the films on a substrate by imitating the chemical vapour deposition process. As a prototype, the system is confined into two–dimensional slap where the deposit chamber is divided into array grids. The vapour flows in and out with varying rates and concentrations and the growth process takes place on the substrate placed at the bottom of the array. Interaction among atoms is considered to take Ising–like, and Kawasaki algorithm (Kawasaki, 1965) is used in this study to observe the average thickness and its variance. The result shows that for the increasing temperature, the average thickness decreases at fixed rates and concentration. With turning on the rates, at fixed concentration, the averaged thickness decreases at low temperature. Furthermore, the averaged thickness greatly increases at low temperature when the concentration, at fixed rates.

Key words: Nanotechnology, Surface growth, Chemical vapour deposition, Monte Carlo

INTRODUCTION

Thin–film surface morphology, especially at nano–thickness range, controls many important physical and chemical properties of the films leading to many essential applications e.g. gas sensor (Choopun et al., 2007). It is therefore of great interest to understand the evolution of the surface morphology during thin film growth. Nevertheless, our scientific understanding of their growth mechanisms is still lacking. Currently, theory, experiment and computer simulations are used together in order to better understand the underlying mechanisms behind thin-film growth.

Computer simulations are often used to verify theoretical approaches or to help guide experimental procedures (Levi and Kortla, 1997). In turn, new experimental results or theoretical advances are then used to improve simulation models. Computer simulations can explain about the nature of thin-film growth which is unobtainable by theory of experiment (Gyure et al., 1998). Nevertheless, the formation of a growth front is a complex phenomenon and often occurs far from equilibrium, so modeling of the growth phenomena cannot be trivially taken. For instance, when atoms are deposited on a surface, atoms do not arrive at the surface at the same time causing surface growth front roughness especially at low temperatures or at high growth rates. This makes the problem very difficult to model, so the simulation technique is required.

One way to do such simulations is the kinetic Monte Carlo method. The kinetic Monte Carlo methods have proven very successful at simulating dynamic properties of non-equilibrium system (Binder and Heermann, 1992; Kalos and Whitlock, 1986). Therefore, we studied the effects of vapour flowing rates and concentration on surface growth via using the kinetic Monte Carlo simulation. The considered system located on square lattice grids in spatial space, being influenced by the direction-independent interaction with varying the temperature. The Kawasaki algorithm was used as the configuration update-algorithm. The result was taken in term of thickness of film.

METERIALS AND METHODS

We consider the Ising Hamiltonian

$$H = -\sum_{<ij>} J_{ij} S_i S_j$$

where $S_{i(j)} = \{1,0\}$ referring to an occupied and a vacant atom on array grids in spatial space. In the equation, the sum takes on only nearest-neighbor pair and J_{ij} was assumed direction-independent interaction. In real system, we found that interaction between vapour is weaker than solid. So in this study, we assume $J_{ij} = J$ for the vapour's interaction and $J_{ij} = 1.5J$ for the solid's interaction or the interaction between atoms at the bottom of the system. The unit J and J / k_B are used for energies and temperatures where k_B is the Boltzmann's constant.

In making simulation, the lattice with occupancy of $N=L_x \times L_y$ sites, where L_x and L_y represent the number of atomic sites along the x and y directions of the system and N represents the total atom of the system, the time unit was defined in terms of Monte Carlo step (mcs), i.e., 1 mcs is number of times that were used for random atom in N times . In this study we used $L_x=L_y=L=100$, the concentration (C) varies from 0.001 to 0.01 percent, temperature (T) varies from 0.01 to 10 J/k_B and the flowing rate (R), that demonstrate the number of time for flowing in and out atoms to the system, varies from 1 to 10 mcs^{-1}. Next, we input atoms that are relative to concentration by random the position of the atoms. During the thermal diffusing process (the kinetic Monte Carlo simulation), simulations used the atom-vacancy exchange dynamic that the exchanging process consists of random atom-vacancy exchanges throughout the lattice. To describe, we put at random atom. Then, we randomly exchanged direction. After that, we proposed the exchange between these two with the Kawasaki probability.

$$\text{probability}(p) = \exp\left(-\frac{\Delta H}{k_B T}\right)$$

where ΔH is the energy difference between the before-exchange states.

Next, we used random number (r) [0,1). If $\leq p$, the position of this atom will change but if $r > p$, this atom will be untouched. Then, we put the new atom at random and repeated this process relatively with the rate. After that, we removed the atoms that are in vapour state and input new atoms that are relative to concentration by random the position of the atoms and repeated this process until time pass by 1000 mcs. We chose 1000 mcs because it is suitable value to explain the result and make the simulation in short time.

In this study, we paid attention to the thickness of the system so we found the average thickness by using this equation.

$$\text{average thickness} = \frac{\sum_{i=1}^{L} h_i}{L}$$

where h_i is thickness of each array grids.

RESULTS AND DISCUSSION

From the simulations, the effect of the vapor concentration (*C*) on the atom deposition on the substrate surface at a fixed temperature has been revealed. As can be seen from Figure 1, which show the simulated growth films at temperature $T = 0.2\ J/k_B$ and time t =1000 mcs, the films gets thicker with increasing the concentration from 0.0025 to 0.01%. This is because at the higher concentration, the system has more atoms in the vapor phase and hence the chemical potential. Therefore, there is a high tendency that the atoms are driven from the vapor to solid phase and stick onto the substrate surface.

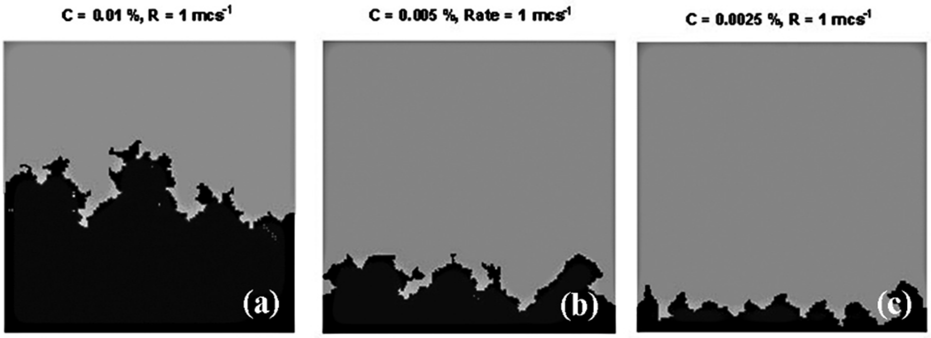

Figure 1. Simulated thin-film under the effect of concentration only, at temperature = 0.2 J/k_BT and time =1000 mcs. Light gray dots are the vacancy or the empty space whereas dark gray dots refer to the atoms that stick at the bottom of the system.

Nevertheless, on considering the effect of the flowing rate (R) only, Figure 2 shows the simulated growth-films at $T = 0.2\ J/k_B$ and $t = 1000$ mcs, the films becomes thinner with increasing flowing rate from 2 to 10 mcs^{-1}. This is expected since at high flowing rate, the atoms have less time for mobility and changing position before being taken away from system. Therefore, there are fewer tendencies if the atoms in the vapor phase have enough time to deposit themselves on the substrate.

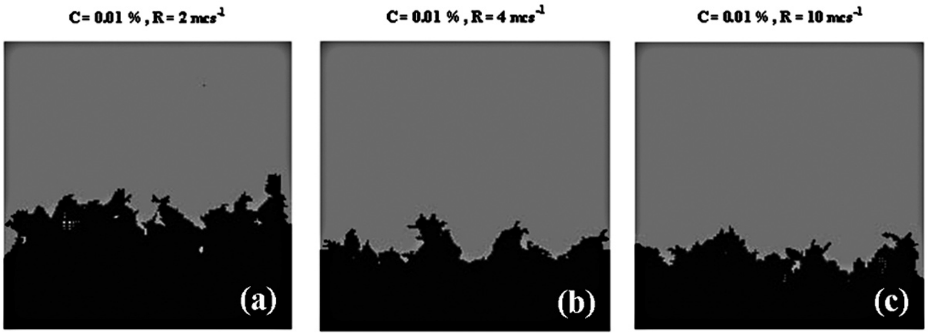

Figure 2. Simulated thin-film under the effect of rate only, at temperature = 0.2 J/k_BT and time = 1000 mcs. Light gray dots are the vacancy or the empty space whereas dark gray dots refer to the atoms that stick at the bottom of the system.

In addition, the effect of simulated temperature on the average thickness of the films with varying the concentration C and rate R was also considered where the results are presented in Figures 3 and 4 in order. As can be seen in Figure 3, at fixed C, R and t, the films is thick at very low temperatures (e.g. at $T < 0.1\ J/k_B$) but with increasing the temperature, the thickness suddenly ceases and becomes very small at high temperatures (e.g. at $T < 1\ J/k_B$). This is due to, at high temperature, there are high thermal energy fed into the systems and hence the atoms have high kinetic energy. This causes the atoms to travel in a random fashion and less atom should be found at a particular position e.g. on the substrate. Therefore, this is why the agglomeration of the atoms is less happened and the films become very thin. In a same way, at a very low temperature, the atoms become less mobile since the thermal and kinetic energy is low. As a result, those atoms tend to stick to themselves making a large cluster of atoms. In addition, since the substrate surface is assumed to have a lower temperature than the vapor temperature and J for the atoms connecting to the substrate is set larger than normal to suit the situation, the atoms at the bottommost layer act as seed atoms for the thickness to grow. Moreover, at a fixed temperature, the increase of C also brings about the thicker films as a result of more atoms in the vapor phase is available.

Furthermore, in Figure 4, which instead fixes C but varies R, at low temperatures, the average thickness gets thicker with either increasing the concentration C or decreasing rate R. The reason is the same as previously given for Figures 1 and 2 that the increase of C allows more atoms to deposit on the substrate surface and so does the decrease of *R*. Similarly, at high temperatures, in an agreement with Figure 4, the thickness is very thin at high temperature due to higher level of thermal and kinetic energy.

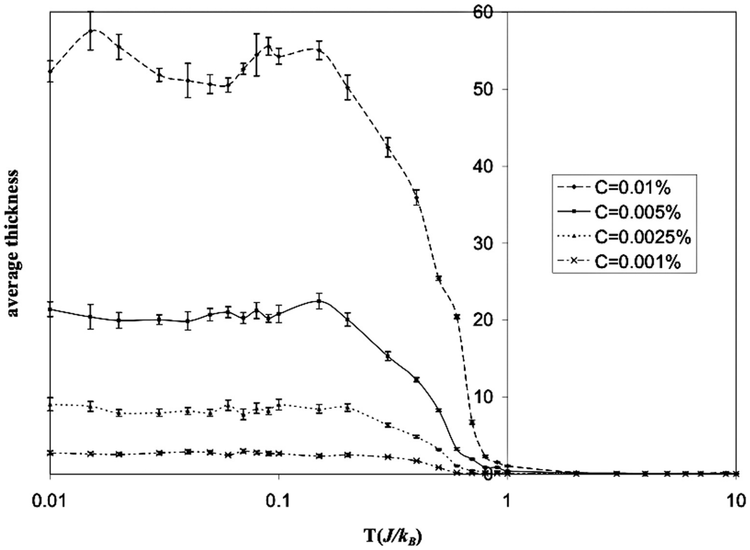

Figure 3. The average thickness of thin-film with varying concentration (C) as a function of temperature at rate (R) = 1.0 mcs^{-1} and time = 1000 mcs.

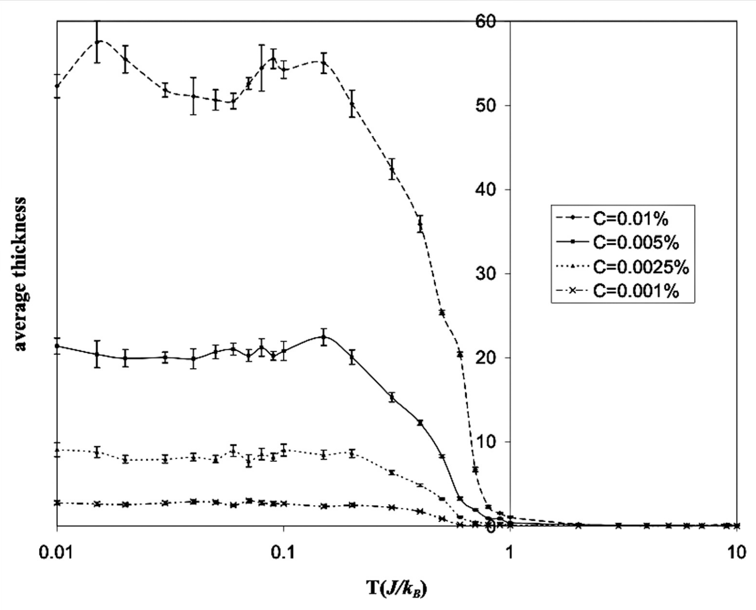

Figure 4. The average thickness of thin-film with varying rate (R) as a function of temperature at concentration (C) =0.01 % and time = 1000 mcs.

CONCLUSION

In this study, the effects of vapor flowing rates, vapor concentration and temperatures on surface growth at nano-thickness range via the kinetic Monte Carlo simulations were investigated. It was found that the films are well growth at low temperatures due to the suitable level of thermal energy on allowing the deposition to take place. In addition, with increasing vapor concentration the films get thicker as more atoms are available, and with decreasing the flowing rate, the thickness also gets thicker since there provides more time for the agglomeration process.

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