非晶質炭素膜による石英表面の平滑化

Evolution of Surface Roughness of Quartz Surface by Amorphous Carbon films

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1. Introduction

Amorphous carbon thin films attracted extensive attention recently owing to the sp^2 and sp^3 hybridized bonding which yields exceptional mechanical and tribological properties [1]. These thin films have been used as coating materials for several major applications such as semiconductors, hydrogen storage devices, and medical implants. The common factor between all these distinct industries is the ability to control the surface morphologies by either depositing smooth top coating or obtaining porous surface layers. Therefore, optimizing the surface roughness of end products is increasingly becoming meaningful as it can impact overall performance.

Experimental evaluations of the surface structure are extremely unreliable due to the lack of any experimental methodology that can accurately describe the surface morphologies from the atomic point of view. Thus, molecular dynamics (MD) provides a deep understanding of the surface roughness evolution during amorphous carbon thin film deposition. Additionally, MD simulation offers a clear explanation regarding the growth mechanisms along with the distribution of stress at each step of the deposition process [2].

In this paper, molecular dynamics is utilized to illustrate the different stages of surface smoothing as a result of carbon atoms deposition on a rough surface. A comparison between low deposition angles of (0°, 30°) and high deposition angles of (60°, 75°) is investigated in terms of surface roughness and the morphologies of the top carbon layer.

2. Simulation Methodology

Molecular dynamics simulation was employed to fully model the quartz SiO_2 substrate with the dimension of 76×44×56 $Å^3$ containing 13,500 silicon and oxygen atoms. The substrate was divided into three sets of layers; the bottom 11 layers (17.1 Å) were fixed, the intermediate 10 layers (15.5 Å) were subjected to a heat-bath at room temperature by Langevin equation, and the top 15 layers (23.4 Å) were free to interact with incident atoms.

To simulate a rough surface with a root mean square roughness (Rq) of 2 Å, 4,500 argon atoms accelerated with the kinetic energy of 50 eV were inserted to sputter the top free layers of quartz. The Si-O, Si-Si, and O-O interactions are described using Tersoff potential [3]. Next, carbon atoms were deposited on the rough surface using the reactive empirical bond order (REBO) potential for C-C interactions [4]. The boundary conditions were applied along the x-axis and y-axis with the time step of 1 fs. The time interval between two sequential deposition impacts of 200 carbon atoms was set at 1 ps with 10 ps relaxation period at 300 K before the first deposition to equilibrate the substrate energy. The carbon atoms were introduced from a position of 44 Å above the substrate surface with various deposition angles, θ = 0°, 30°, 60°, and 75° with the z-axis to test the effect of deposition angle on the smoothing process. The kinetic energy of each carbon atom was set to 75 eV as it was proven to be the optimum energy to form a dense diamond-like carbon (DLC) film [5].

3. Results and discussion

Figure 1 illustrates the changes in the surface roughness up to 2 Å after 270 nanoseconds of argon sputtering. After carbon deposition with 0° and 30° angles, surface roughness decreases to the minimum value of 0.85 Å and 1.1 Å, respectively at 321 nanoseconds. This is followed by an abrupt rise in roughness back to the original value of 2.2 Å at 395 nanoseconds. Then, surface roughness reduces again to 1.1 Å and remains relatively stable until the end of the simulation run. On the other hand, in the case of carbon deposition at high angles (60° and 75°), a limited reduction in roughness values between

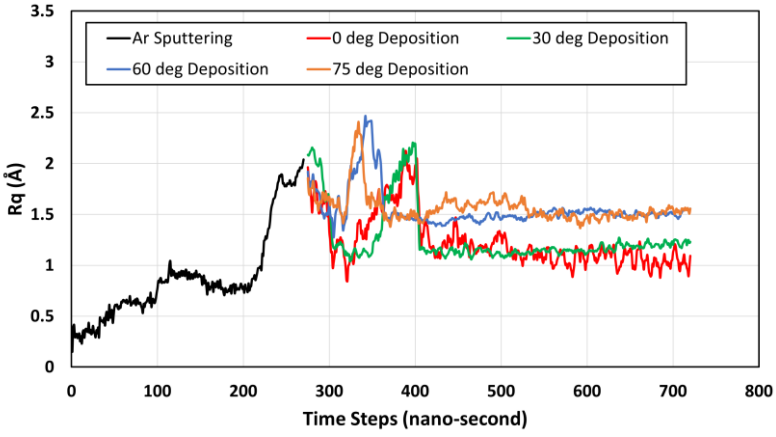

Fig. 1 MD surface roughness fluctuation after both argon sputtering and carbon deposition at different angles.

1.4-1.5 Å occurs at around 310 nanoseconds. Thereafter, an instant increase to the maximum surface roughness number of 2.4 Å takes place between 330-350 nanoseconds. Finally, a decrease in the roughness between 1.5-1.6 Å appears at 380 nanoseconds and keeps stable to the end.

Figure 2 shows snapshots of the top view of surface morphologies at the time step of 430 nanoseconds. This time step is selected as it represents the end of the surface roughness fluctuations of all deposition angles and also it is considered to be the beginning of stabilized region of all curves according to Fig. 1. It is observed that the distribution of carbon atoms is well spread on asperities of the rough surface when atoms are deposited at low angles of 0° and 30° as shown in Fig. 1 (a, b). On the other hand, there is a poor atomic distribution of carbon exposing more areas of surface valleys indicated by yellow and green color in case of high deposition angles of 60° and 75° as seen in Fig. 2 (c, d). Furthermore, the sizes of the agglomerated carbon islands (red color) of high deposition angle substrates are bigger and denser compared to low deposition angle substrates. This implies that incident carbon atoms at low angles possess higher energy opposed to carbon atoms deposited at low angles which facilitates spreading of the incident atoms more evenly across the surface.

It is believed that the first stage of smoothing occurs due to the aggregation of multiple carbon atoms absorbed at the bottom of the valley. By further deposition, those clusters begin to congregate into islands, raising the roughness to the original values. This process is attributed to the shadowing effect [6, 7]. Finally, carbon atoms fill up all morphologies of the quartz surface, which helps to dissipate the kinetic energy of the incident carbon atoms due to the preferred C-C bonding energy leading to a decrease in surface roughness.

4. Conclusion

Molecular dynamics simulation reveals a unique smoothing behavior that occurs at the early stages of carbon atomic deposition. A low surface roughness of 1.1 Å is detected at low deposition angles of 0° and 30° compared to a high surface roughness of 1.5 Å at high deposition angles of 60° and 75°. Surface irregularities of carbon islands formed due to the shadowing effect are more emphasized in the case of high angles, which is the main reason for these two surface behaviors.

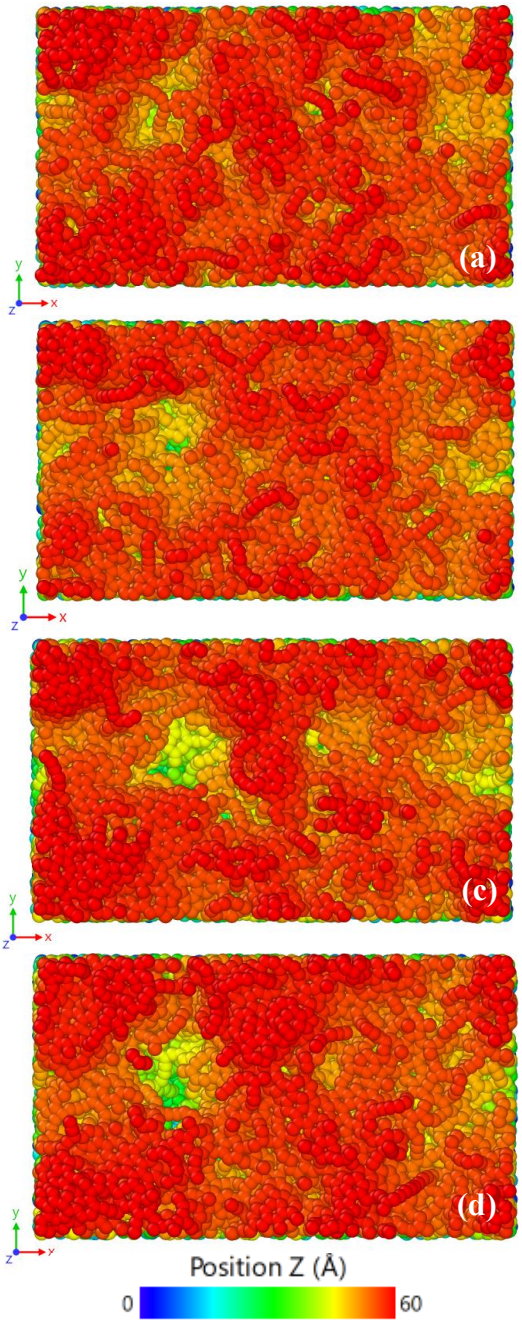

Fig. 2 Top view snapshots of the surface morphologies at 430 nanoseconds after carbon deposition under the deposition angle of (a) 0°, (b) 30°, (c) 60°, and (d) 75°. Color encodes the position of atoms in the z direction as indicated in the legend above.

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