

Hillock Behavior on Aluminum Thin Films Deposited on Polyimide Film

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Polyimide 박막상에 증착된 알루미늄 박막의 Hillock 거동

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초 록 Polyimide를 입힌 SiO₂ wafer상에 증착된 알루미늄 박막의 두께 및 소둔 여부에 따른 hillock의 거동을 atomic force microscopy (AFM)을 이용하여 분석하였다. 증착된 상태의 박막에서 성장 hillock이 관찰되었으며 박막 두께가 증가할수록 hillock의 크기는 증가한 반면 밀도는 감소하였다. 소둔 후 hillock의 평균 크기는 증가하였으나 밀도는 감소하였다. 이러한 hillock 밀도의 감소는 견고한 wafer상에 직접 증착된 알루미늄 박막에서와 다르다. 이는 유연한 polyimide 박막에 의한 응력 완화로 응력유기 입계확산이 이루어지지 않아 hillock이 추가로 형성되지 않은 상태에서 큰 hillock이 성장하면서 작은 hillock을 흡수하기 때문으로 판단된다.

Abstract Behavior of hillocks on aluminum films deposited on polyimide-coated SiO₂ wafer has been investigated using an atomic force microscopy with variation of the film thickness and annealing treatment. Growth hillocks were observed on as-received films and hillock density was decreased while hillocks grew in size with the film thickness. After annealing, average hillock size was increased but density was decreased. The reduced hillock density in these films is in contrast with the results from the films deposited directly on a rigid substrate. This is attributed to the presence of soft polyimide layer which relaxes the stress and thereby lacks the stress-induced grain boundary diffusion in aluminum films. It is suggested that, in this situation, no additional hillocks emerge and small hillocks are consumed by growing large hillocks.

1. Introduction

Hillocks are numerous outgrowths on the surface of thin films, which cause a number of problems in performance and reliability of microdevices. Hillocks on the surface of aluminum mirrors in spatial light modulators degrade the optical performance of these devices.¹⁾ In aluminum interconnect metallization, hillocks form on the sides of the patterned lines as well as the top surface.²⁾ The side hillock can cause electrical shorts between patterned interconnect lines and the surface hillock can lead to interlayer shorts in multilayer metallization.

Hillocks on thin films are generally classified into two types: growth hillocks and annealing hillocks. Growth hillocks are characterized by crystalline appearance with well-defined surfaces and edges,^{3,4)} and they are considered to form during the growth of the film. Annealing hillocks form during thermal cycling, and are commonly recognized by their rounded shapes with no distinct crystalline features. A number of experimental observations and

mechanistic studies have been done on the formation of hillocks.^{3~10)} The results suggest that hillocks form as a result of the relaxation of compressive stresses during film deposition and thermal cycling, although the hillock formation mechanisms have not been understood completely yet. Hillock formation is irreversible and thus hillocks do not disappear when the compressive stress is removed from the film. The growth of a hillock involves atomic diffusion along grain boundaries and, presumably, along the film surface as well as the interface between film and substrate. It is known that the microstructures, properties, and stress states of deposited thin films are influenced by the substrate. Therefore, the characteristics of hillocks depend on the nature of substrate. Most of the previous works,^{3~10)} however, have been focused on rigid Si and SiO₂ substrates. As organic polymers draw increasing interest for the advanced intermetal dielectrics in microelectronics,¹¹⁾ it is necessary to understand the hillock behavior on the flexible substrates in both fundamental and practical view

points. In this study, it has been investigated the evolution of hillocks on aluminum films deposited on the soft polyimide with the film thickness and annealing treatment.

2. Experiments

PMDA-ODA (DuPont Pyraline PI2540) polyimide precursor solution was spun onto an oxidized 6" Si wafer and then cured at 300°C under the nitrogen flow. Thickness of polyimide was approximately 3.5 μm . Aluminum (Al>99.99%) thin films were deposited onto polyimide-coated Si wafers using an DC magnetron sputtering system at room temperature in the thickness range of 60–480nm. Aluminum films were annealed at 400°C for 30 minutes in vacuum under constant flow of a forming gas. Specimens were prepared by cutting wafers into small squares which measure 1cm \times 1cm.

Observation of hillocks was made using a Digital Instruments Nanoscope III atomic force microscope (AFM) in contact deflection mode. The size, density, and height of hillocks were measured automatically by the microscope from the 10 μm \times 10 μm scanned images, although the 5 μm \times 5 μm images are

shown below for better morphological observation. The threshold height was chosen to be 10nm because the polyimide film cured on a SiO₂ substrate exhibited surface roughness of about 2.5nm which became 4.7nm after annealing at 400°C. In addition, it yielded a good agreement in the size and density of hillocks with the results measured manually from the scanning electron microscopy (SEM) pictures.

3. Results and Discussion

Fig. 1(a)–(d) show 3-D images of hillocks on as-deposited aluminum thin films of 60, 120, 240, and 480nm thickness. Hillocks on these films after annealing are shown in Fig. 2(a)–(d). Hillocks are observed on the surface of all aluminum films. Many hillocks are con-shaped, but the hillocks on a 480nm film look rather flat. The con-shaped hillocks are similar to the "spire-like" hillocks reported by Santoro,⁹⁾ which grow on the triple junction of grain boundaries. The size of hillock increases with the film thickness while the number of hillocks appears to be less influenced by the film thickness. It is noted that surface morphology

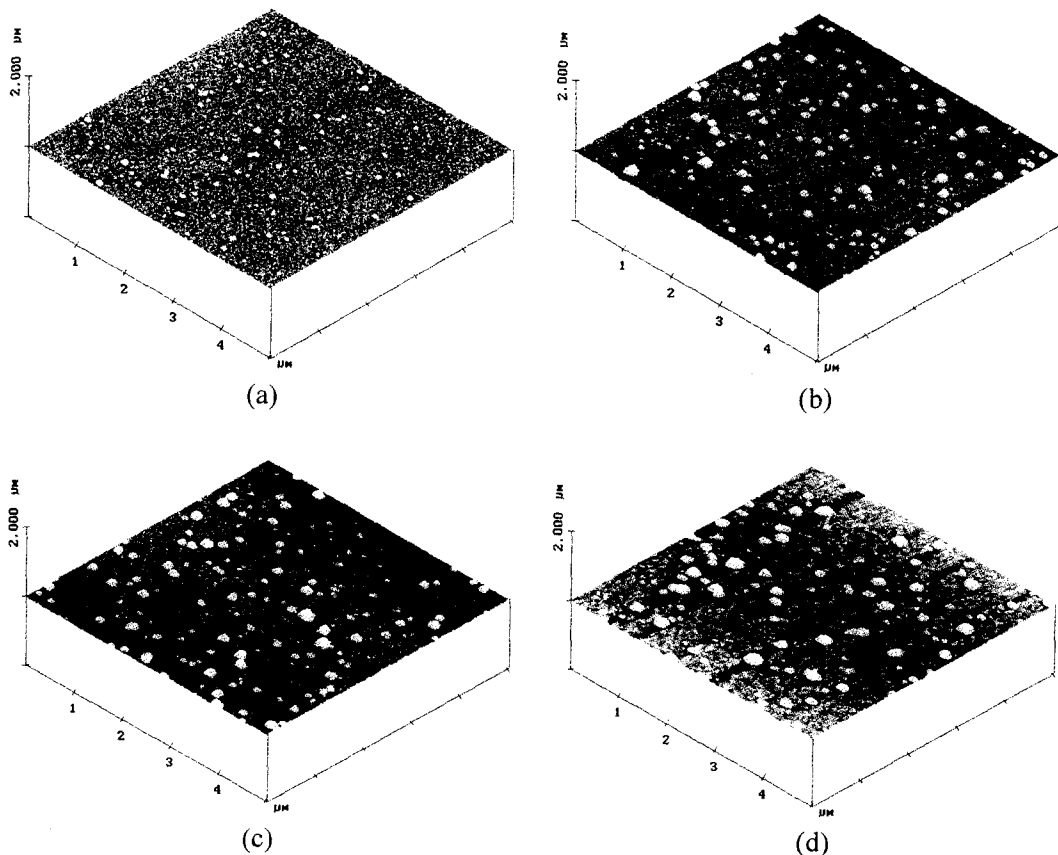


Fig. 1. 3-D AFM images of hillocks on as-deposited aluminum films with thicknesses of (a) 60nm, (b) 120nm, (c) 240nm, and (d) 480nm.

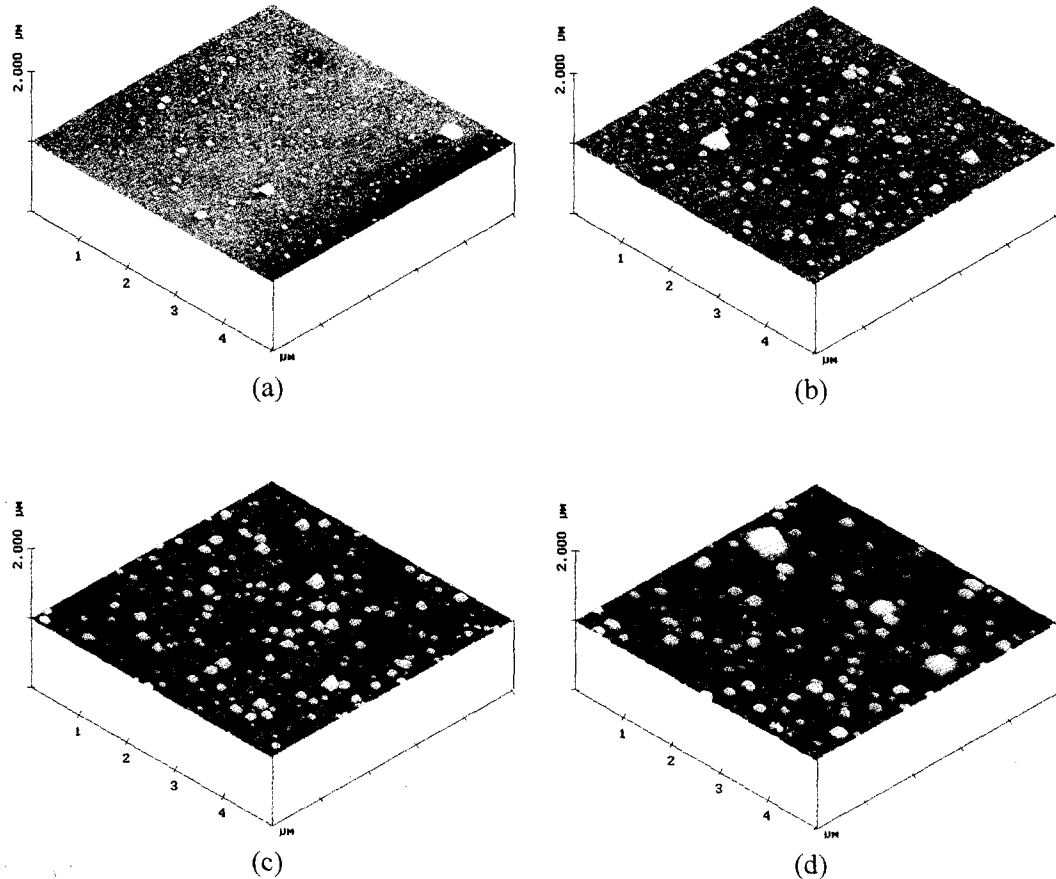


Fig. 2. 3-D AFM images of hillocks on annealed aluminum films with thicknesses of (a) 60nm, (b) 120nm, (c) 240nm, and (d) 480nm.

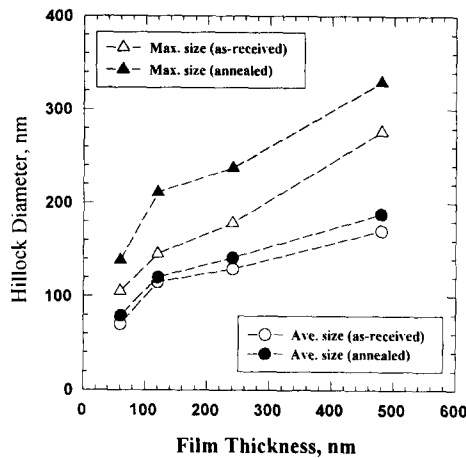


Fig. 3. Film thickness dependence of average and maximum hillock sizes for aluminum thin films before and after annealing.

of the films does not change much after annealing except the growth of a few large hillocks. Some hillocks were found to grow into a few μm size (not shown here) after annealing.

The film-thickness dependence of size, density, and height of hillocks are plotted in Figs. 3, 4, and 5, respectively. It can be seen in Fig. 3 that avera-

ge size of hillocks increases with the film thickness both before and after annealing. Compared to the increment of the average hillock size, the maximum hillock size increases more pronouncedly after annealing, which is due to the appearance of a few large hillocks. On the contrary, as shown in Fig. 4, hillock density reduces as the film thickness increases after it reaches its maximum at 120nm. Furthermore, hillock density decreases rather than increases after annealing in the entire film thickness range. Fig. 5 shows that average hillock height does not vary with the film thickness and annealing treatment. However, maximum hillock height increases with the film thickness and annealing treatment in the entire film thickness range. This behavior is similar to maximum hillock size. In Fig. 6, the ratio of average hillock size to average hillock height is shown as a function of the film thickness. The average size of hillock is several times larger than average hillock height. Hillocks become larger in size but remain almost unchanged in height as they grow with the film thickness resulting in flat hillocks in a thicker film.

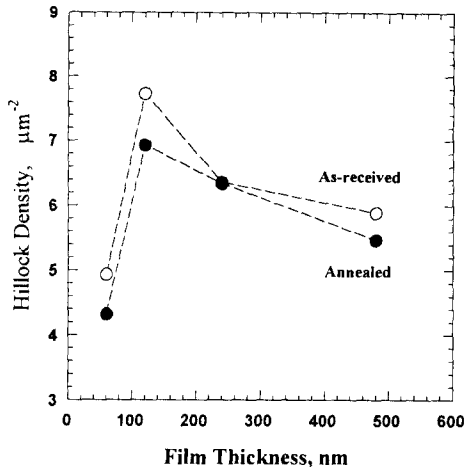


Fig. 4. Hillock density as a function of the film thickness for aluminum films before and after annealing.

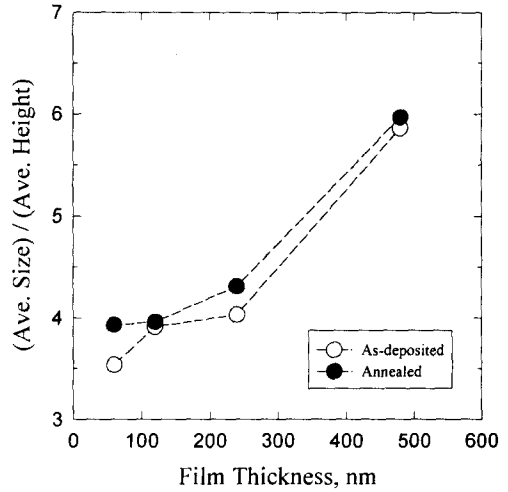


Fig. 6. The ratio of average hillock size to average hillock height as a function of the film thickness.

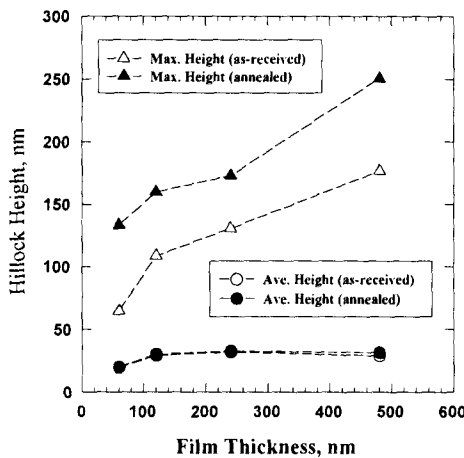


Fig. 5. Film thickness dependence of average and maximum hillock heights for aluminum thin films before and after annealing.

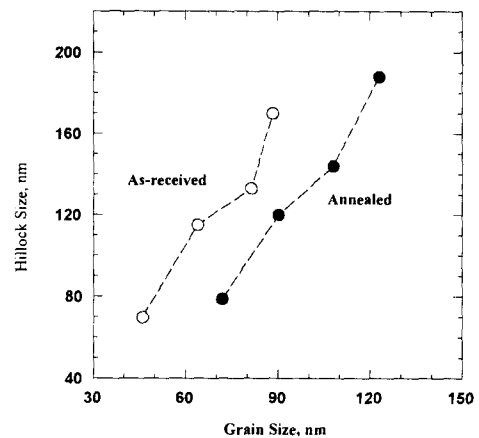


Fig. 7. Hillock size versus grain size for aluminum thin films before and after annealing.

Grain size dependence of average hillock size is shown in Fig. 7. Hillocks extend over more than one grain and hillock size linearly increases with the grain size both before and after annealing. However the curve shifts to the right side on grain size axis after annealing. This indicates that grain growth is faster than hillock growth upon annealing.

Observation of a large number of hillocks on as-deposited films is unusual because hillocks on unannealed aluminum films deposited on SiO₂ substrate at room temperature have seldom been reported.³⁻⁷ Nevertheless it is conceivable that the observed hillocks are growth hillocks formed during film growth at room temperature. The mechanism of growth hillock formation is not clearly understood yet, but it is believed to result from the relaxation

of compressive stresses during film deposition. However metal films deposited on Si or SiO₂ substrate usually exhibit tensile stresses, although the exact nature and amount of stress depends on a number of deposition parameters. When a polyimide layer is present between the metal and substrate, the amount of stress in the film is substantially reduced,¹² which is in favor of hillock formation. Since the surface morphology of as-deposited films depends on deposition conditions including chamber pressure¹⁰ as well as substrate properties, further investigation is necessary to elucidate the mechanism of hillock formation on as-deposited aluminum films on polyimide.

Studies^{3, 9} of hillock growth on rigid substrates found that the size and density of hillocks increase with the film thickness. This result was interpreted as the increase of grain size with film thickness

providing more grain boundary path for stress-driven diffusion. In this study, however, hillock density reaches maximum at a film thickness of 120nm and then reduces with the film thickness, while average hillock size slightly increases. The decrease of hillock density can be attributed to the presence of soft polyimide. The soft polyimide film relaxes the stress of metal film¹²⁾ thereby suppressing stress-driven grain boundary diffusion. In this situation no new hillocks emerge as the film becomes thicker and large hillocks preferentially grow out by consuming smaller ones in a manner similar to grain coarsening.

The role of polyimide film becomes prominent upon annealing. The observed decrease of hillock density after annealing is in contrast with other published results^{3,6)} for the aluminum films directly deposited on rigid substrates such as Si and SiO₂ wafers, where numerous hillocks emerged during heating. Annealing hillocks are considered to form during heating due to the compressive stress caused by the difference in thermal expansion coefficient (TEC) between aluminum and substrate. Therefore, the number and size of hillocks are expected to increase with the annealing temperature. However the stress state of aluminum film is altered when the polyimide interlayer exists between the film and SiO₂ substrate. Polyimide, which has TEC (~30ppm) close to that of aluminum (27ppm) and much lower elastic modulus, neither induce nor transfer the compressive stress to aluminum films. Thus the lack of compressive-stress-driven diffusion yields no annealing hillocks. Insteads large hillocks grow larger at the expense of small ones presumably via thermally-enhanced surface diffusion as observed upon isothermal annealing of hillocks.⁷⁾ Notice the hillock growth behavior with film thickness observed on as-deposited films is preserved after annealing treatment. This result indicates that growth of hillocks upon annealing dominantly depends on the presence of polyimide layer between aluminum thin films and a rigid substrate.

4. Conclusion

The characteristics of hillocks on aluminum films

deposited on polyimide-coated SiO₂ wafer have been investigated using an atomic force microscopy with the focus on the evolution of hillocks with the film thickness, in the range of 60–480nm, and annealing treatment. Growth hillocks were observed on as-received films and hillock density was decreased while hillocks grew in size with the film thickness. After annealing, average hillock size was increased but density was decreased. The reduced hillock density in these films is in contrast with the results from the films deposited directly on a rigid substrate. This is attributed to the presence of soft polyimide layer which relaxes the stress and thereby lacks the stress-induced grain boundary diffusion in aluminum films. It is suggested that, in this situation, no additional hillocks emerge and small hillocks are consumed by growing large hillocks.

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