Effects of total gas flow rate and sputtering power on the critical condition for target mode transition in Al-O_2 reactive sputtering

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Abstract

Reactive sputtering is one of the most commonly used techniques for the fabrication of compound thin films, and the critical condition for target mode transition from metal mode to oxide mode is very important. We investigated the effects of total gas flow rate and sputtering power on the critical condition in Al-O_2 reactive sputtering. It was found that the ratio of the number of sputtered Al atoms (N_{Al}) to the number of supplied O atoms (N_O) at the critical condition was almost constant, and the ratio of N_{Al} to N_O was close to the stoichiometric ratio of Al_2O_3 (2 to 3). It is thought that the introduced oxygen is gettered by Al atoms almost completely and the target remains in the metal mode below the critical condition. By increasing the amount of supplied O atoms above the stoichiometric ratio of Al_2O_3, the oxygen supply overcomes the gettering effect. Then, oxygen concentration in the plasma increases abruptly and the target mode changes from metal mode to oxide mode.

Keywords: reactive sputtering, plasma emission intensity, target voltage, gettering effect

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1. Introduction

Reactive sputtering is one of the most commonly used techniques for obtaining compound thin films by sputtering a metal target in a reactive gas atmosphere [1]. It is well known that target mode change (target poisoning) is very important in reactive sputtering because a change in the target surface state induces dramatic changes in the deposition rate and the chemical composition of the deposited films. Reactive sputter deposition can be a highly complex process, governed, for example, by the physics of plasma discharge, transport of sputtered and gas species, kinetics of film growth and chemical interactions at the target and film surfaces. A number of models of reactive sputtering have been proposed to explain the effects of processing parameters on the hysteresis, the change in deposition rate, the target poisoning effect, and the critical amount of reactive gas [2-11]. However, certain parameters, such as sputtering yields, sticking coefficients, pumping speed and target ion current, had to be assigned appropriate values to fit the theoretical data to the experimental results, and many researchers have yet to find suitable experimental conditions for their own sputtering systems.

We have studied the processes of reactive sputtering of Ti-O_2, Si-O_2, and Si-N_2 systems experimentally [12-14], and determined the critical conditions for compound film formation by changing the reactive gas flow ratios. However, several different oxides or nitrides, such as TiO, Ti_2O_3 and TiO_2, could be formed in these systems, which may obscure the analyses. In our previous paper [15], the formation process of Al_2O_3 thin films was studied because Al_2O_3 thin films are widely used in many mechanical, optical and microelectronical applications owing to their excellent properties, namely, chemical inertness, high mechanical strength and hardness, transparency, high abrasive and corrosion resistance, and insulating properties. Furthermore, Al_2O_3 is the only oxide of Al, and thus we expect that more reliable data on the target mode change could be obtained. It was found that the target mode change occurred at an O_2 flow ratio of 8% under a constant sputtering power of 50 W and a constant total gas flow rate of 5 ccm. Under these conditions, the ratio of the number of sputtered Al atoms to the number of supplied oxygen atoms was close to 2 to 3, which is the stoichiometric ratio of Al_2O_3 [15]. In this study, we examined the effects of total gas flow rate and sputtering power on the critical conditions for the target mode transition in Al-O_2 reactive sputtering. Because the amount of oxygen supply and Al supply depend on the total gas flow rate and sputtering power, and we believe that the relationship between the Al and oxygen supply at the critical condition has been clearly established.

2. Experimental

Reactive sputtering of Al_2O_3 was performed using the RF (13.56 MHz) magnetron sputtering system shown in Figure 1. The sputtering chamber, 460 mm in diameter and 360 mm in height, was evacuated to a base pressure below 2.1×10^{-4} Pa using a turbo molecular pump with a pumping speed of 190 L/s. An Al (99.99% purity) target with a diameter of 50 mm was used for the sputtering. A gas mixture of Ar and O_2 was used for sputtering, and the O_2 flow ratio was varied from 0% to 100%. The total gas pressure during sputtering was maintained at a fixed value of 0.67 Pa. The total gas flow rate and RF input power were varied from 1 to 5 ccm (mL/min at 25˚C and 101.3kPa) and from 20 to 100 W, respectively. The flow rates of Ar and O_2 gases were controlled by mass flow controllers. The total pressure was measured with a capacitance manometer. The plasma condition during sputtering was determined by plasma emission spectroscopy using a multi-channel charge-coupled device (CCD) detector, and the target surface state was characterized by measuring the target voltage. The amount of Al sputtered from the Al target was determined by weighting the mass of an Al film deposited onto an aluminum foil with a thickness of 12 μm, which covered the surface area of a hemisphere (radius of approximately 120 mm) over the target [16], at an Ar flow ratio of 100%.

3. Results and discussion

3.1. Effects of total gas flow rate

The effects of O_2 flow ratio on the critical condition for target mode transition were studied. Figure 2 shows plasma emission intensity of oxygen atoms (777 nm) [17] as a function of O_2 flow ratio at a total gas flow rate of 1, 2, and 5 ccm. Sputtering power was adjusted to a constant value of 50 W. A rapid increase in the emission intensity of oxygen atoms takes place at critical O_2 flow ratios of 8, 17 and 33% for the total flow rates of 5, 2, and 1 ccm, respectively. As the total gas flow rate was decreased, the critical O_2 flow ratio for the target mode transition increased. Figure 3 shows the change of the target voltage as a function of O_2 flow ratio at total gas flow rates of 1, 2, and 5 ccm. The target voltage also decreases abruptly above the same critical O_2 flow ratios of 8, 17, and 33% as shown in Figure 2. These findings point to a change of the target mode from metal mode to oxide mode [1]. At low O_2 flow ratios below the critical points, oxygen molecules are gettered by Al atoms deposited on the sputtering chamber wall and the substrate, and the oxygen density in the plasma is very low. Owing to the very low density of oxygen in the plasma, the target surface remains in a metallic state and Al films or oxygen containing Al films are formed at the surface of the substrate. The transmittance and the resistivity of the oxygen containing Al films are slightly larger than those of pure Al films. In the high O_2 flow rate region above the critical conditions, the number of O_2 molecules supplied exceeds the gettering effect and the oxygen density in the plasma begins to increase. Because of the high density of oxygen in the plasma, the target surface is oxidized and transparent and insulating Al_2O_3 films are formed [15].

The effects of total gas flow rate on the critical conditions of the mode transition are studied in connection with the amount of supplied oxygen and sputtered Al atoms. The number of supplied oxygen atoms (N_O) at the critical O_2 flow ratio was calculated from the total gas flow rate and the critical O_2 flow ratio. N_O was found to be almost constant at about 3.1×10^{-5} mol/min at the total gas flow rates of 1, 2, and 5 ccm. The number of sputtered Al atoms (N_{Al}) in a 100% Ar atmosphere (metal mode) was estimated from the mass of the Al film deposited on the symmetrically arranged foil. N_{Al} was also found to be constant at about 2.2×10^{-5} mol/min for all total gas flow rates, as shown in Figure 4. These results indicate that the ratio of N_{Al} to N_O is constant and close to the stoichiometric ratio of Al_2O_3 (2 to 3), irrespective of the total gas flow rate, as shown in Figure 5. The slight increase in the ratio N_O/N_{Al} with increasing total gas flow rate observed in Figure 5 might be caused by the increase in effective pumping speed. At high total gas flow rates, we had to open the main valve (and thus increase effective pumping speed) to maintain a constant total gas pressure, and some of the oxygen atoms were evacuated by the vacuum pump without oxide formation. Thus, a larger amount of oxygen was necessary for the target mode change.

3.2. Effects of RF input power

The effects of sputtering power on the critical condition for target mode transition were studied. Figure 6 shows plasma emission intensity of oxygen atoms (777 nm) as a function of O_2 flow ratio at a sputtering power of 20, 50, and 100 W. The total gas flow rate was adjusted to a constant value of 5 ccm. A rapid increase in the emission intensity of oxygen atoms occurs at critical O_2 flow ratios of 3, 8, and 12% for the sputtering powers of 20, 50, and 100 W, respectively. The critical O_2 flow ratio of target mode transition increased with increasing sputtering power. Figure 7 shows the change of the target voltage as a function of O_2 flow ratio at a sputtering power of 20, 50, and 100 W. The target voltage also decreased abruptly above the same critical O_2 flow ratios of 3, 8, and 12% as shown in Figure 6.

The effects of sputtering power on the amount of supplied oxygen at the critical conditions and that of supplied Al atoms are studied. N_{O} at the critical O_2 flow ratio was calculated to be 1.3×10^{-5} mol/min at 20 W and increased to 5.4×10^{-5} mol/min at 100 W. Corresponding to the increase of N_{O}, N_{Al} increased linearly with increasing sputtering power, as shown in Figure 8. However, the ratio of N_{Al} to N_{O} is almost constant, at close to the stoichiometric ratio of Al_2O_3 (2 to 3), irrespective of the sputtering power, as can be seen in Figure 9. The slight increase in $N_{\mathrm{O}}/N_{\mathrm{Al}}$ at low sputtering power might be also explained by the increase in effective pumping speed. At low sputtering powers, the amount of gettered oxygen was small and a relatively large amount of oxygen was evacuated by the vacuum pump.

3.3. Critical conditions for target mode change

The above two sets of experiments reveals that regardless of the total gas flow rate and sputtering power, the ratio of N_{Al} to N_{O} at the critical condition for target mode transition is close to the stoichiometric ratio of Al_2O_3 (2 to 3) in Al-O_2 reactive sputtering. It is believed that oxygen molecules introduced into the sputtering chamber are almost completely gettered by Al atoms deposited on the sputtering chamber wall and the substrate. Thus, the oxygen density in the plasma is very low and the target remains in the metal mode below the critical conditions. Above the critical conditions, oxygen supply overcomes the gettering effect and the oxygen density in the plasma increases abruptly, which induces the target mode change. The maximum number of gettered oxygen atoms is determined by the number of sputtered Al atoms and the stoichiometric ratio of Al_2O_3. Thus, the amount of supplied reactive gas necessary to form compound thin films by reactive sputtering is determined solely by the number of sputtered metal atoms under the metal target mode.

4. Conclusion

We investigated the effects of total gas flow rate and sputtering power on the critical condition for target mode change in Al-O_2 reactive sputtering. It was found that regardless of the total gas flow rate and sputtering power, the ratio of the number of sputtered Al atoms (N_{Al}) to the number of supplied O atoms (N_O) at the critical condition is almost constant and close to the stoichiometric ratio of Al_2O_3 (2 to 3). The critical amount of reactive gas (O_2) necessary to form compound films (Al_2O_3) is determined solely by the amount of sputtered metal (Al) under the metallic target mode.

Acknowledgement

The authors are grateful to Koji Shinya for the plasma emission intensity and target voltage measurements.

References

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Figure captions

Fig. 1 Schematic drawing of the RF (13.56 MHz) magnetron sputtering system used in the present experiments.

Fig. 2 Plasma emission intensity of oxygen atoms (λ = 777 nm) as a function of O_2 flow ratio at a total gas flow rate of 1, 2, and 5 ccm. The sputtering power was adjusted to a constant value of 50 W.

Fig. 3 Target voltage as a function of O_2 flow ratio at a total gas flow rate of 1, 2, and 5 ccm. The sputtering power was adjusted to a constant value of 50 W.

Fig. 4 The number of sputtered Al atoms (N_{Al}) under metal mode (100% Ar) as a function of total gas flow rate. The sputtering power was adjusted to a constant value of 50 W.

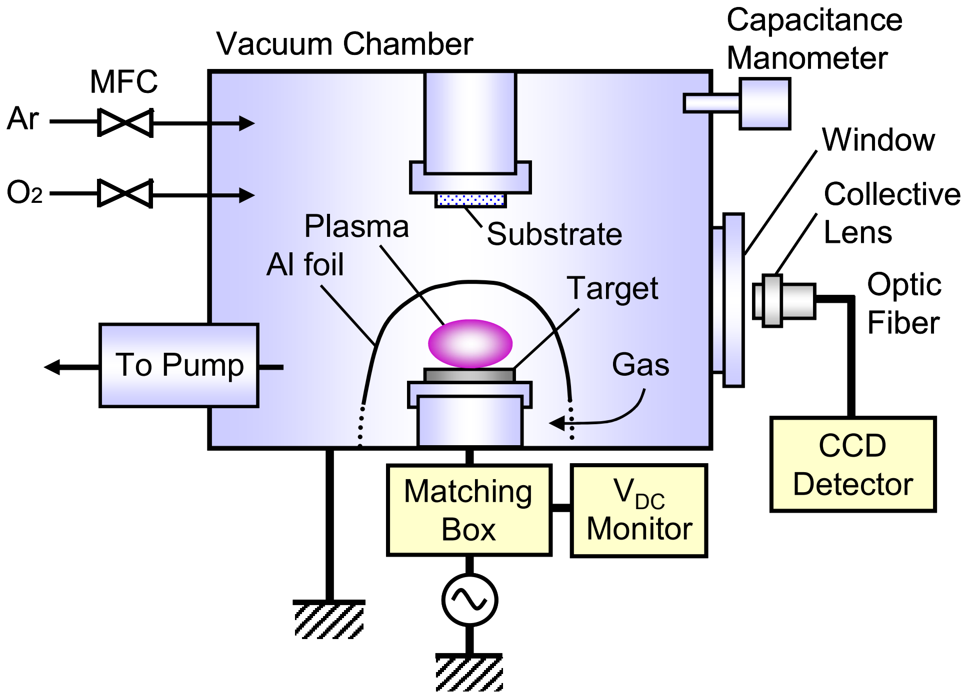
Fig. 5 The ratio of sputtered Al atoms (N_{Al}) to supplied O atoms (N_{O}) at the critical condition for target mode transition as a function of total gas flow rate.

Fig. 6 Plasma emission intensity of oxygen atoms (λ = 777 nm) as a function of O_2 flow ratio at a sputtering power of 20, 50, and 100 W. The total gas flow rate was adjusted to a constant value of 5 ccm.

Fig. 7 Target voltage as a function of O_2 flow ratio at a sputtering power of 20, 50, and 100 W. The total gas flow rate was adjusted to a constant value of 5 ccm.

Fig. 8 The number of sputtered Al atoms (N_{Al}) under metal mode (100% Ar) as a function of sputtering power. The total gas flow rate was adjusted to a constant value of 5 ccm.

Fig. 9 The ratio of sputtered Al atoms (N_{Al}) to supplied O atoms (N_{O}) at the critical condition for target mode transition as a function of sputtering power.



Vacuum Chamber
Capacitance Manometer
MFC
Ar
O2
Window
Collective Lens
Plasma
Substrate
Al foil
Target
Optic Fiber
To Pump
Gas
CCD Detector
Matching Box
V_DC Monitor

Fig. 1

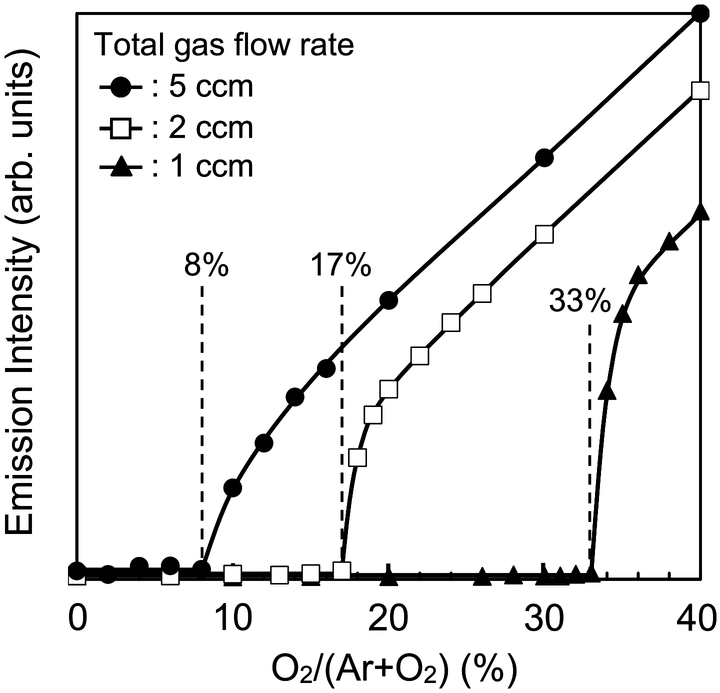

Total gas flow rate
: 5 ccm
: 2 ccm
: 1 ccm
8%
17%
33%
Emission Intensity (arb. units)
0
10
20
30
40
O2/(Ar+O2) (%)

Fig. 2

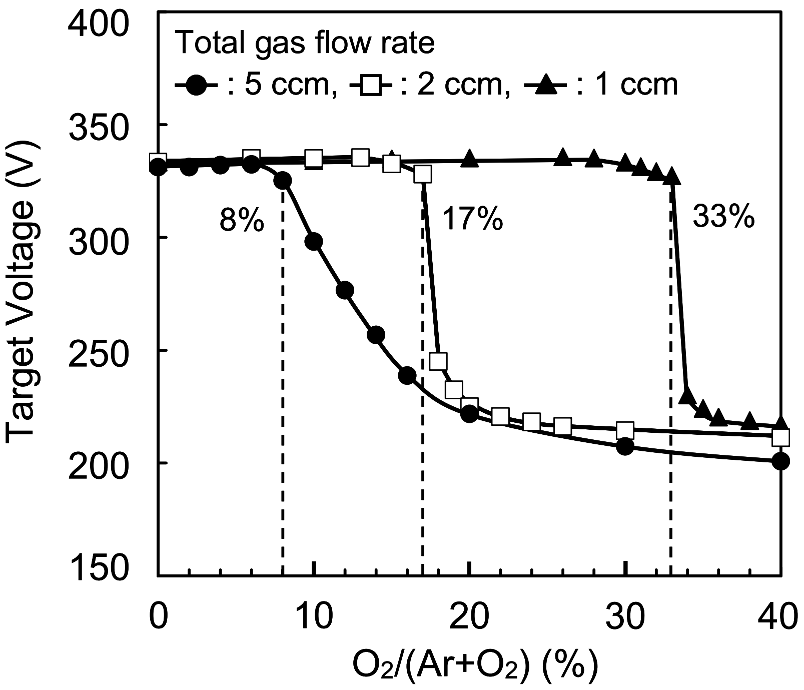

Total gas flow rate
●: 5 ccm, □: 2 ccm, ▲: 1 ccm
8%
17%
33%
Target Voltage (V)
400
350
300
250
200
150
0
10
20
30
40
O2/(Ar+O2) (%)

Fig. 3

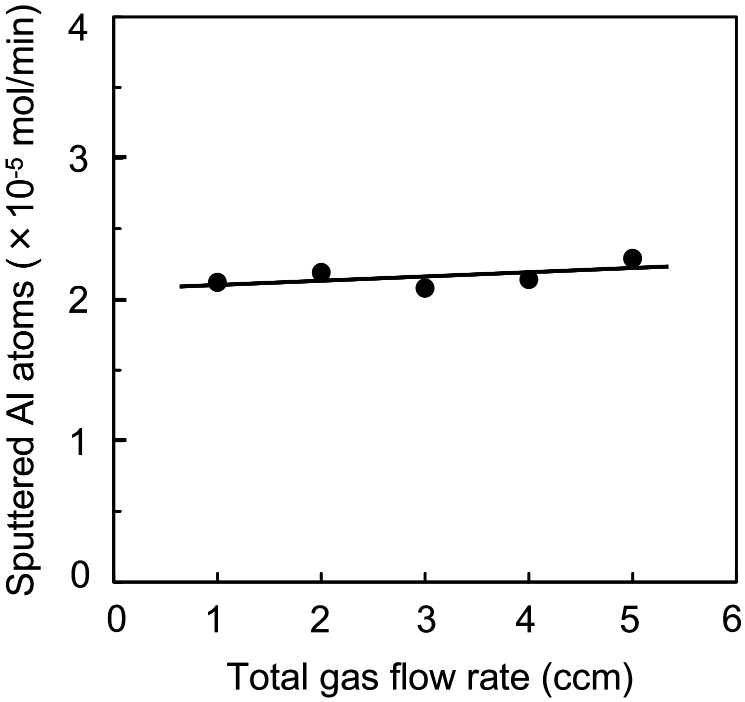

Sputtered Al atoms (× 10-5 mol/min)
4
3
2
1
0
0
1
2
3
4
5
6
Total gas flow rate (ccm)

Fig. 4

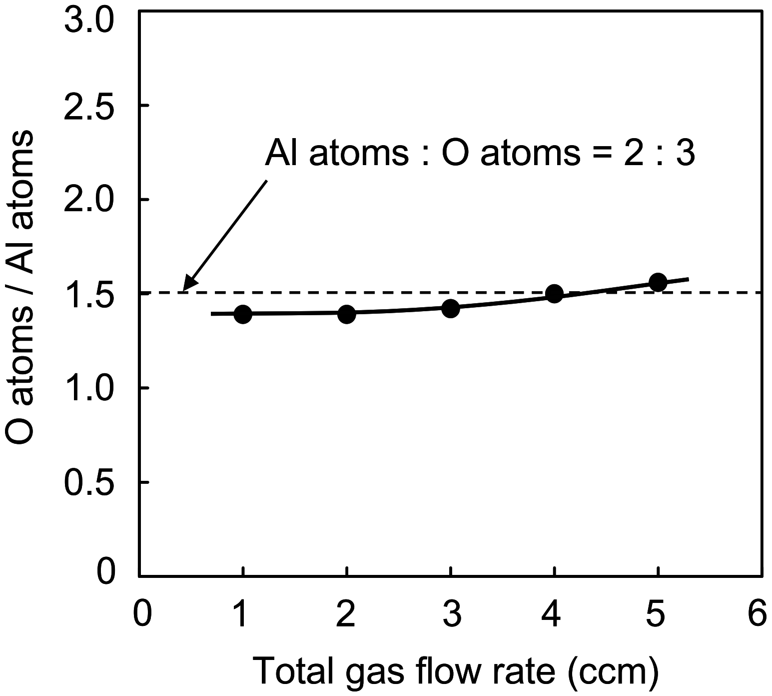

3.0
2.5
2.0
1.5
1.0
0.5
0
0
1
2
3
4
5
6
O atoms / Al atoms
Total gas flow rate (ccm)
Al atoms : O atoms = 2 : 3

Fig. 5

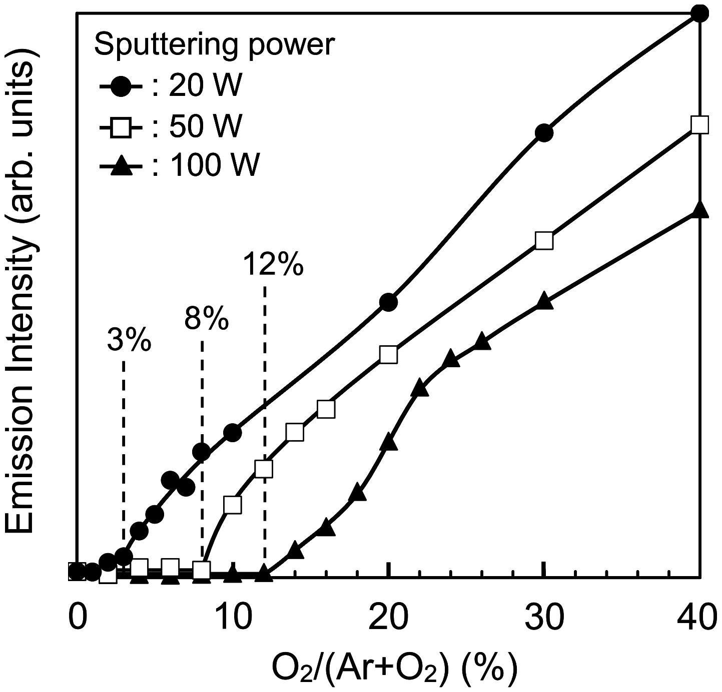

Sputtering power
●: 20 W
□: 50 W
▲: 100 W
3%
8%
12%
Emission Intensity (arb. units)
0
10
20
30
40
O2/(Ar+O2) (%)

Fig. 6

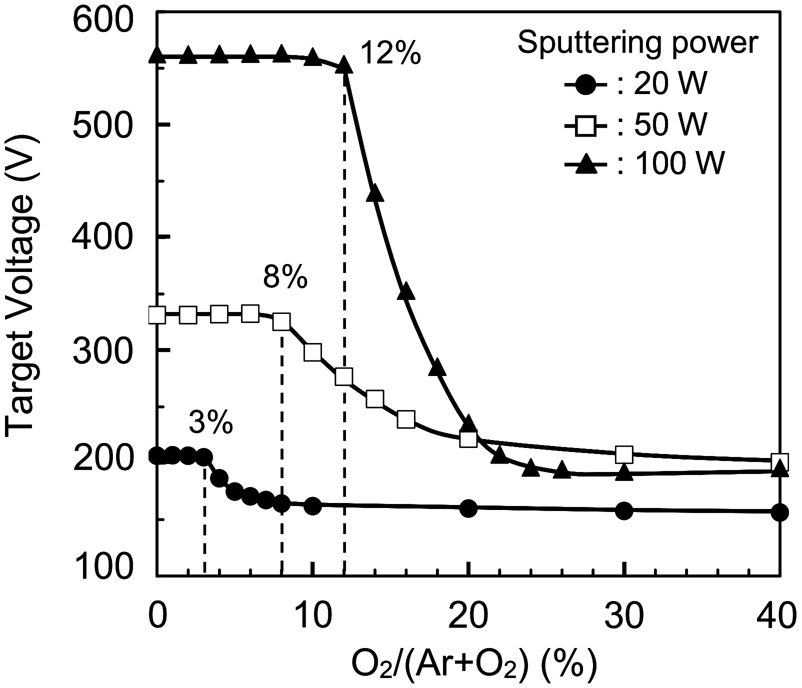

600
500
400
300
200
100
Target Voltage (V)
0
10
20
30
40
O2/(Ar+O2) (%)
Sputtering power
: 20 W
: 50 W
: 100 W
12%
8%
3%

Fig. 7

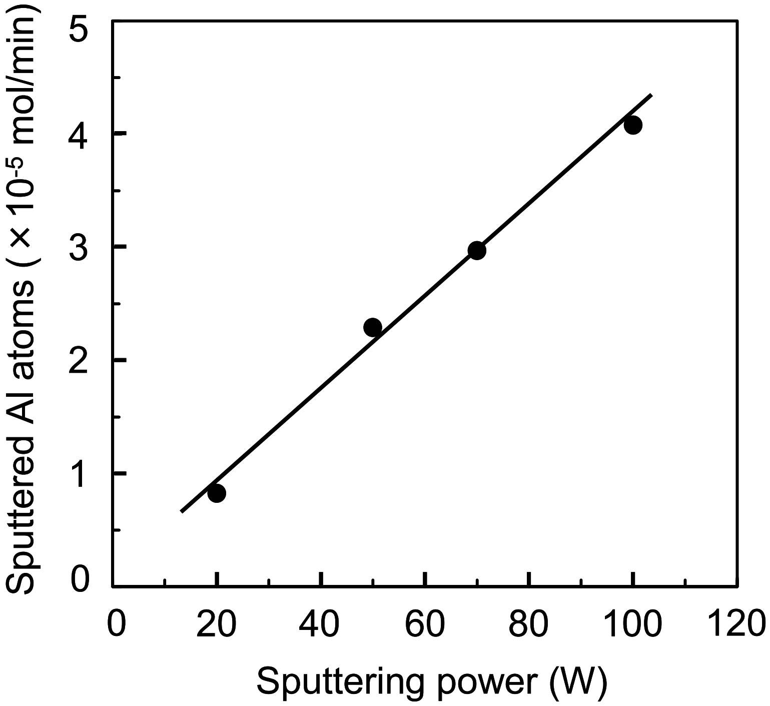

Sputtered Al atoms (× 10^{-5} mol/min)
0
1
2
3
4
5
0
20
40
60
80
100
120
Sputtering power (W)

Fig. 8

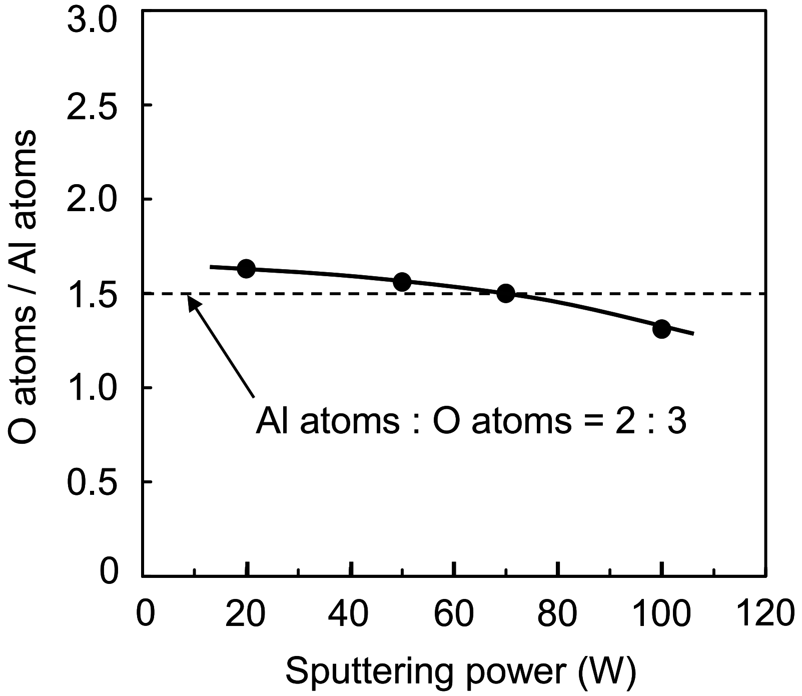

3.0
2.5
2.0
1.5
1.0
0.5
0
O atoms / Al atoms
0
20
40
60
80
100
120
Sputtering power (W)
Al atoms : O atoms = 2 : 3

Fig. 9