Formation Process of Reactively Sputtered MoO_3 Thin Films and Their Optical Properties

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Electrochromic MoO_3 thin films are thought to be useful as a neutral light adjustment element for charge coupled device (CCD) and complementary metal-oxide semiconductor (CMOS) imaging device. Reactive sputtering has been commonly used for preparing oxide thin films, however, formation process of reactively sputtered MoO_3 films has not been studied in detail. In this paper the effect of oxygen flow ratio on the formation of MoO_3 films is studied

An RF magnetron sputtering system was used for sample preparation. A 2 inch disk of Mo target (99.9% purity) was sputtered in an Ar and O_2 mixed gas, and Mo and MoO_3 films were deposited on quartz glass substrates. O_2 flow ratio was varied from 0% to 100%. RF power, total gas flow rate and sputtering gas pressure were kept constant at 50 W, 5 cc/min and 50 mTorr, respectively. Plasma emission spectra were monitored by a CCD detector. Optical properties of the MoO_3 films were characterized by spectrophotometry and elipspometry.

Transmittance of the deposited films is plotted as a function of O_2 flow ratio in Fig. 1. The color of the films deposited at O_2 flow ratios of 0-10% is metallic silver, however, it becomes transparent and transmittance more than 70% is obtained above O_2 flow ratios of 15%. The transparent films are thought to be MoO_3 because refractive index of 1.8-2.1, which is close to that reported for MoO_3 films, was obtained. The deposition rate of the films decreases abruptly at an O_2 flow ratio of 15% as shown in Fig. 2. The decrease of the deposition rate is thought to be caused by the target mode change from metal mode to oxide mode. Figure 3 shows plasma emission intensity of oxygen atoms during the sputter-deposition as a function of O_2 flow ratio. The emission intensity of oxygen atoms is very low at O_2 flow ratios of 0-10% and it begins to increase above 15%. The amount of Mo atoms sputtered from the target was estimated to be 2.7×10^{-5} mol/min in the metal mode by the mass of the deposited Mo film, and the amount of supplied O_2 molecules is calculated to be 3.35×10^{-5} mol/min at an O_2 flow ratio of 15%. Therefore, the ratio of the number of supplied Mo atoms to that of oxygen atoms becomes 1:2.5, which is close to the stoichiometric value of 1:3 for MoO_3, at the critical O_2 flow ratio of 15%.

From these results the formation process of Mo and MoO_3 films is thought to be as follows. 1) The introduced O_2 molecules are gettered by the Mo atoms deposited on the sputtering chamber wall and Mo films are formed at O_2 flow ratios of 0-10%. 2) Above an O_2 flow ratio of 15%, oxygen density in the plasma begins to increase because the amount of introduced oxygen is more than that necessary to the formation of MoO_3. Due to the oxygen plasma the surface of the Mo target begins to be oxidized and the deposition rate decreases abruptly. As the results MoO_3 films are formed.

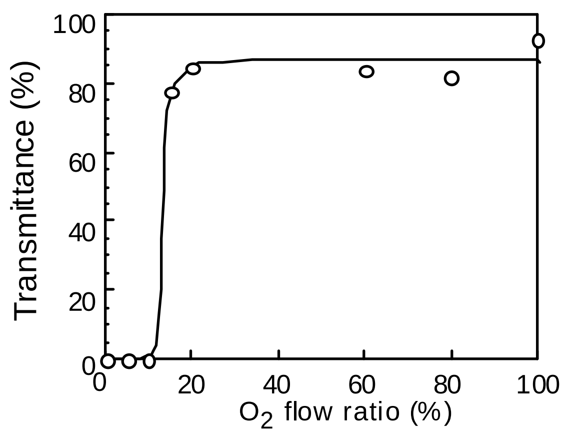

Fig. 1. Transmittance of the films at a wavelength of 600 nm as a function of O_2 flow ratio.

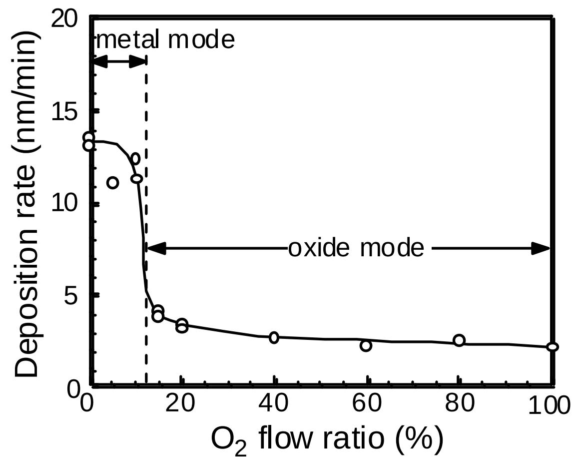

Fig. 2. Deposition rate of the films as a function of O_2 flow ratio.

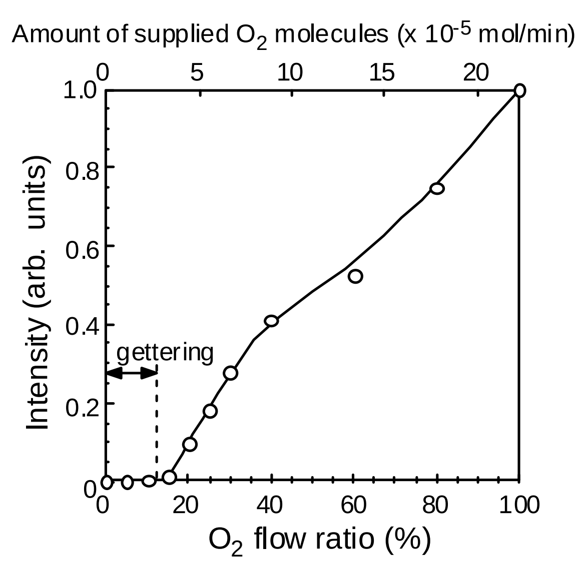

Fig. 3. Plasma emission intensity of oxygen atoms (777 nm) as a function of O_2 flow ratio.