Thermal Probe Measurements of Energy Flux onto a Substrate during ICP Assisted Sputter-Deposition

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Abstract: Energy flux onto a substrate in the inductively coupled plasma (ICP) assisted magnetron discharge was measured with a thermal probe. Changes in the energy flux against the ICP power and substrate bias were investigated. As a result, we found that i) the energy flux was proportional to the ICP power in a pure ICP and was predominantly determined by the ICP power in the ICP assisted sputtering, ii) the experimental substrate bias dependence of the energy flux was in good agreement with the calculated one. The influence of thin film deposition on the energy flux measurement is also reported.

Keywords: thermal probe, energy flux, ICP, transparent conductive films

1. Introduction

Tin-doped indium oxide (ITO) has been widely used as a transparent conductive film. The reason for the wide use of ITO is that a large area sputter-deposition of transparent conducting films with low-resistivity and high optical transmittance can be made with low-cost and good reproducibility. Alternative materials to the ITO have been anticipated, however, because the indium is a rare metal and ITO powder has toxicity. Recently, aluminum doped zinc oxide (AZO) received attention as an alternative material. The AZO is stable in the reduction atmosphere, low-cost, and harmless in the human body. However, fabrication process is unstable and its reproducibility is scarce. Therefore, it is necessary to establish the deposition process with high reliability. In addition, a further improvement of film quality is necessary. Therefore we have been studying deposition process of AZO films by using inductively coupled plasma (ICP) assisted magnetron sputtering. As a result, we can obtain good quality (high crystallinity, high conductivity, high optical transmittance) AZO thin films, in particular when the sputtering is assisted with increased ICP power.[1] However, not only the relation between the plasma parameters and thin film properties but also the role of ICP assist power on them are not clarified yet.

Energy flux onto a substrate has been measured with different thermal probes (TPs) by many researchers in the RF plasma [2-7], RF magnetron plasma [8,9], and DC grow discharge plasma [10] in the past. However, no measurement of the energy flux onto the substrate during the ICP assisted sputtering has been done as far as we know so far. Thus, the grasp of the energy flux onto a substrate during the ICP assisted sputtering is very important.

This paper reports the experimental results of the measurement of energy flux onto the substrate during the ICP assisted sputtering.

2. Experimental

Figure 1 shows the schematic of the TP made in this work. The TP is composed of a copper plate (10 mm in diameter, 0.1 mm in thickness) facing to the plasma, a ceramics rod (6 mm in diameter, 10 mm in length) producing temperature gradient, an aluminum rod (6 mm in diameter, 350 mm in length) connected to the outer heat sink, and outer tubes (Teflon and stainless steel tubes) for thermal and electric insulation, which prevents the energy flux from coming in from the side of the TP. The copper plate, ceramic rod and aluminum rod were tightly bonded. A tip of first thermocouple was attached on the ceramics rod at the distance of 6mm from the joint plane between the ceramics and the aluminum rods. The other end of the aluminum rod was water-cooled outside the vacuum vessel. The outer stainless steel tube for heat insulation was grounded to cut the electrical coupling between the TP and plasmas. The most thermal gradient appears on the ceramic rod that has a thermal conductivity of 1.6 W/(m·K). The length of the ceramics and the spacing of the two TP tips were determined by the tradeoff between the requirements of smaller thermal time constant of the ceramic rod and that of larger difference in temperature at both the ends of the ceramic rod. The thermal time constant is expressed as follows:

\[ \tau = CR = \rho cV \frac{L}{kS} = \frac{\rho c L^2}{k} \]

where \( C \) is thermal capacity, \( R \) is thermal resistance, \( \rho \) (kg/m³) is mass density, \( c \) (J/(kg·K)) is specific
heat, $k\, [\text{W/(m} \cdot \text{K)}]$ is thermal conductivity, $V\, [\text{m}^3]$ is volume, $S\, [\text{m}^2]$ is cross section area, $L\, [\text{m}]$ is length of the TP body. The shorter the length of ceramic rod is, the smaller the thermal time constant is. The quick response of the probe is favorable to improve the measurement efficiency. On the other hand, the longer the length of the ceramic rod is, the larger the temperature difference along the ceramic rod is as shown in eq. (1). The aluminum rod has large thermal conductivity and the temperature gradient in the aluminum rod is very small. It mostly works as a heat sink because the end of the aluminum rod was water-cooled.

The inflow energy flux (energy per unit area per second) $J$ was measured with the TP by two different ways: steady state operation or pulse operation. In the steady state operation, $J$ is evaluated by the equation

$$J = -k \frac{\Delta T}{\Delta x}.$$  \hspace{1cm} (2)

In case of the pulse operation, $J$ is evaluated by the equation

$$J = \frac{Q_{\text{in}}}{S} = mc \left[ \left( \frac{dT_s}{dt} \right)_{\text{on}} - \left( \frac{dT_s}{dt} \right)_{\text{off}} \right],$$ \hspace{1cm} (3)

where $mc$ is the thermal capacity ($m$: mass of probe head, $c$: specific heat), $S$ is probe surface area, and $[\left( \frac{dT_s}{dt} \right)_{\text{on}} - \left( \frac{dT_s}{dt} \right)_{\text{off}}]$ is the difference in temperature gradient at common substrate temperature difference $T_s$ during heating and cooling period.

The copper plate of TP was biased by connecting a lead wire. As a result of investigation of the influence of film deposition on probe surface, we confirmed that the measurement value decreased 4% by a film deposition of 100nm thickness. To clean the copper plate surface, the bias voltage of -100V was applied for fifteen minutes every time in the experiment.

Figure 2 shows the experimental setup for the energy flux measurement in ICP assisted sputter-deposition. A vacuum chamber (300 mm in diameter and 300 mm in height) was attached with a 3 inch DC planar magnetron, an argon gas supply system, and a pumping system (turbo molecular pump and rotary pump combination). A disk target of ZnO:Al$_2$O$_3$ (2wt%) of 60 mm diameter and 6mm thick was used as target. Between these diode electrodes, a single turn coil antenna of 100 mm diameter was installed and used for the production of 13.56MHz inductively coupled plasma. The antenna was covered with insulator and water-cooled. The distance from the target to the RF coil and the distance from the RF coil to the substrate were both 40 mm. The TP was set 8 cm from the target surface in the axial direction, and 3 cm from the center axis of magnetron. An argon gas was introduced at a flow rate of 50sccm by a mass flow controller. The operation gas pressure was set to 30mTorr by adjusting the main valve. After that, cooling water was flowed through the target, ICP antenna and the TP. The ICP electric power was turned on for some period until the thermal equilibrium was attained. Then, ICP power was turned off. Temperatures at two different positions were measured by two thermocouples installed in a TP, and were displayed on digital multimeters, then temperatures were recorded on a personal computer.

Figure 3 shows the temporal change of difference in temperature measured by the two thermocouples on the TP for the ICP RF power 100W and 200W when plasma on period was 60 s (pulse operation) and 700 s (steady state operation). The data shown in Fig. 3 indicates that the difference in temperature is mostly saturated with a time constant $\tau$ of 160
seconds and the thermal equilibrium condition is reached at the time after 3-4 \( \tau \) seconds. In the steady state operation, the energy flux was confirmed to be evaluated with eq. (2), but the time interval between each measurement took about 1-2 hours because the heating time of 3-4\( \tau \) and the cooling time of 30-40\( \tau \) were required. On the other hand in the pulse operation, the energy flux was estimated with eq. (3) by calibrating \( mc/\mathcal{S} \) with the energy flux evaluated in the steady state, and the time interval between each measurement was reduced by one order of magnitude.

Figure 3. Temporal change in temperature difference obtained by a thermal probe during plasma on and off period for ICP RF power of 100 and 200 W.

To evaluate the validity of the measurement in the pulse operation, we checked the difference in temperature gradient \( (dT_s/dt)_{on}-(dT_s/dt)_{off} \) during heating and cooling against common substrate temperature difference \( T_s \). Figure 4 shows the \( T_s \) dependence of \( (dT_s/dt)_{on}-(dT_s/dt)_{off} \) for ICP RF power of 100W (black circle) and 200W (white circle). For the common substrate temperature difference \( T_s \) which is 30-100\% of the maximum temperature difference, \( (dT_s/dt)_{on}-(dT_s/dt)_{off} \) was almost constant within the error range of \( \pm 5\% \). For the common substrate temperature difference \( T_s \) which is less than 30\% of maximum achieving temperature difference in ICP 200W, however, \( (dT_s/dt)_{on}-(dT_s/dt)_{off} \) deviated from the constant value. Thus, energy flux is able to be measured by pulse operation using \( (dT_s/dt)_{on}-(dT_s/dt)_{off} \) of the common substrate temperature difference \( T_s \) which is more than 30\% of the maximum temperature difference.

Figure 5 shows the ICP RF power dependence of energy flux for different target power of 0 and 40W. The energy flux onto the TP is evaluated to be 2700W/m\(^2\) for the ICP RF power 200W and target power 40W. The energy flux in the ICP assisted sputtering is not a simple addition of the energy flux in the pure planar magnetron and that in the pure ICP; i.e., the energy flux in the ICP assisted sputtering is predominantly determined by the ICP power.

![Figure 4](image-url)  
**Figure 4.** \( (dT_s/dt)_{on}-(dT_s/dt)_{off} \) against common substrate temperature \( T_s \) during heating and cooling period.

![Figure 5](image-url)  
**Figure 5.** ICP RF power dependence of energy flux (ICP assisted PM sputtering).

### 3.2 Substrate bias dependence

The change in energy flux against substrate bias was investigated experimentally and theoretically. The experimental values for the ICP RF power 200W are plotted with white circles in Fig. 6. According to Kersten et al. [4], the total energy flux onto a substrate \( J \) is expressed as the addition of the contribution \( J_i \) by the positive ions, the contribution \( J_e \) by the negative electrons, and the contribution \( J_{rec} \) by the recombination of positive ions and electrons at the substrate surface as shown in equation (4).

Each contribution is given by expressions (5), (6) and (7).

\[
J = J_{kin}^{i} + J_e + J_{rec}
\]  
(4)

\[
J_i = n_e \frac{kT_e}{m_i} \exp(-0.5)e_0(V_{ph} - V_s)
\]  
(5)

\[
J_e = n_e \frac{kT_e}{2\pi m_e} \exp \left(-\frac{e_0V_{bias}}{kT_e}\right) 2kT_e
\]  
(6)

\[
J_{rec} = J_i E_{rec}
\]  
(7)
Here, \( n_e \) is the electron density, \( kT_e \) the electron temperature, \( m_i \) the mass of ion, \( m_e \) the mass of electron, \( e_0 \) the base of natural logarithm, \( V_{pl} \) the plasma space potential, \( V_s \) the substrate potential, \( V_{bias} (= V_{pl} - V_s) \) the potential fall from plasma to the substrate, and \( E_{rec} \) the recombination energy of a positive ion.

Theoretical energy flux onto a substrate \( J \) was calculated for the pure ICP, because the experimental data on \( n_e \), \( kT_e \), and \( V_{pl} \) were available; i.e., the electron temperature, the electronic density and the plasma potential were measured respectively 2.5eV, \( 2.58 \times 10^{17} \)m\(^{-3} \) and 16.8V for the condition of ICP RF power 200W at 30mTorr. Thus, we obtained \( J \) by substituting the measured values and the physical constants such as \( m_e \), \( m_i \), \( E_{rec} (=15.7eV \) for argon ion) into equations (5), (6), and (7). Thus, the theoretical energy flux for 200W ICP was calculated against \( V_{bias} \), which is shown by a solid curve in Fig. 6.

From Fig.6, we notice that the experimental substrate bias dependence of the energy flux is in good agreement with the calculated one for ICP power 200W, and the experimental energy flux in an ICP is in good agreement with theoretically calculated value within 10% error. The result indicates that \( J_n \), \( J_e \), and \( J_{rec} \) dominantly contributes to the energy flux onto the substrate.

**Figure 6.** Substrate bias dependence of energy flux: Comparison of measured and calculated energy flux.

4 Conclusion

Energy flux onto a substrate in the inductively coupled plasma (ICP) assisted magnetron discharge was measured with a thermal probe. The energy flux was proportional to the ICP power in a pure ICP, and the energy flux in the ICP assisted sputtering is not a simple addition of the energy flux in the pure planar magnetron and that in the pure ICP; i.e., the energy flux in the ICP assisted sputtering is predominantly determined by the ICP power. As for the substrate bias dependence, the energy flux measured with the thermal probe was in good agreement with the calculated results, which were based on the model proposed by Kersten et al.

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