Improved Electrical and Optical Properties of IGZO Transparent Conductive Oxide Due to Microwave Treatment: Application to Silicon Solar Cells

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ABSTRACT The electrical and optical properties of IGZO-based transparent conductive oxide (TCO), fabricated by reactive-sputtering, are optimized using post microwave treatment (MWT), not rapid temperature annealing (RTA), for silicon solar cell. Compared to the sheet resistance and transmittance of IGZO-based TCO after RTA and MWT, we observed a transmittance of over 75 % in the visible and near ultraviolet range of 370-1,200 nm in common, while the lower sheet resistance of 15 Ω/□ was obtained, which was 3.5 times lower than that of the RTA sample, which resulted in higher current density in IGZO-based TCO after MWT. On the basis of trap density analysis, it is confirmed that the oxygen vacancy within IGZO-based TCO increased as process and power in MWT, which is the reason for the increase in conductivity. Furthermore, short circuit current density (J_sc) was compared to confirm the applicability in the solar cell, which is one of the fields that IGZO mainly uses as a TCO. J_sc of the IGZO based solar cell annealed in MWT at 400W for 60 s was 19.8 mA/cm².

INDEX TERMS Microwave treatment, TCO, IGZO, solar cell.

I. INTRODUCTION

Transparent conductive oxide (TCO) is mainly used in various industries such as solar cells, flat panel displays, TFTs, and optoelectronic devices due to their high carrier concentration, low sheet resistance, and high optical transmittance in the visible range. In general, indium tin oxide (ITO) is widely used and studied as a TCO material due to its excellent conductivity, and high transmittance. However, because of the high price and the toxicity of ITO [1], alternative TCO materials, such as zinc oxide (ZnO), aluminum doped ZnO (AZO), indium gallium zinc oxide (IGZO), and so on, have been intensively studied. ZnO has been intensively studied due to its excellent properties, such as a wide-bandgap, non-toxic characteristics, price competitiveness, and good stability in plasma [3]. AZO was also suggested as an alternative ZnO because of its lower electrical conductivity [4], [5]. However, annealing temperature ranges for AZO is constrained by the mismatch of thermal expansion coefficient between glass substrate and AZO. As an alternative TCO material having thermal-stress stability during the annealing, IGZO has attracted attention due to its high transmittance, high carrier concentration, low sheet resistance, and excellent thermal-stress stability [6]. Especially, IGZO is frequently used as a buffer layer in dye-sensitized solar cells because it perfectly compensates for the drawbacks of AZO [7]. In the case of IGZO-based TCO, the high annealing temperature of above 500 °C is commonly required to obtain the best conditions, resulting in an increase on the cost of the device due to the high cost of the glass substrate to use it at high temperature. In addition, it is difficult to apply IGZO to flexible devices because flexible substrates, such as devices on
PET and PEN, are vulnerable to high temperature. Therefore, it is important to have a low sheet resistance via an annealing process less than 500 °C. H.-W. Lee et. al. reported that IGZO annealed by rapid thermal annealing (RTA) at a low temperature and showed excellent conductivity compared to the IGZO annealed using conventional heat treatment [8]. However, high temperature annealing still causes thermal damage to devices during RTA. Microwave treatment (MWT) has been studied as an alternative way for heat treatments because MWT does not need to consume high energy and long-time in high temperature [9], [10]. In addition, in the case of MWT, it is known that has a low thermal budget [11], [12]. Unlike the conventional heat treatment, MWT uses microwaves to vibrate polar molecules for heating. Even though MWT has advantages in terms of heating, MWT is not widely used because the annealing conditions for MWT are not optimized, compared to the RTA method. In this paper, we directly compared the properties of IGZOs depending on the annealing condition of RTA and MWT, and we applied it to the solar cell to evaluate its applicability in applications requiring low-temperature processing.

II. EXPERIMENTAL DETAIL

A. IGZO DEPOSITION, RTA AND MWT

In order to evaluate the IGZO properties depending on conditions of RTA and MWT, the IGZO film was deposited on quartz substrates and silicon (Si) wafers by radio frequency sputtering (KVS-2000L). Quartz substrates and silicon wafers were cleaned using acetone, methanol and deionized water for 10 mins, respectively. The base pressure and working pressure of sputtering chamber during IGZO deposition was under 5 and 20 mTorr. The thickness of the IGZO was 36 nm. After deposition, RTA and MWT were carried out to compare the properties of IGZO. RTA was performed by MILA-3000 (ULVAC) in O₂ atmosphere varying temperature from 200 to 800 °C, while the treatment time was ranged from 30 to 90 s. MWT was performed using a microwave oven (KR-S341T) operating at a maximum power of 1,000 W with a frequency of 2.4 GHz. The power was varied from 200 to 1,000 W, while the time was ranged from 10 to 300 s.

B. ANALYSIS OF IGZO PERFORMED BY RTA AND MWT

In order to analyze the electrical properties of IGZO after RTA and MWT, a 4-point probe was used. Measurements were conducted 10 times in each condition to obtain an average of the sheet resistance. The current density of the IGZO film was also measured using a Keithley 4200 SCS. High power thin film-XRD (D8 ADVANCE) was used to examine the crystalline property of the IGZO films. The optical transmittance for the IGZO film deposited on quartz glass substrates were measured by a UV-Vis spectrophotometer (FC-PH10) in the spectral range from 300 nm to 1.2 μm.

C. APPLICATION: SOLAR CELL SIMULATION

So as to evaluate the feasibility of the IGZO films after MWT to the solar cell, a PV light house simulation was carried out because it is a useful simulation tool for analyzing the photo-generated short circuit current density (J<sub>sc</sub>) of the solar cell [13]. The simulation structure was Air/IGZO/Si wafer. The light intensity was set as 44 mA/cm². The transmittance of the IGZO was carried out the measured data by a UV-Vis spectrophotometer.

III. RESULT AND DISCUSSION

Fig. 1 shows the RTA and the MWT process for the IGZO based device. The left diagram of Fig. 1 describes the IGZO molecule was annealed from outside in RTA, so oxygen vacancy located outside can easily combine with surrounding gas. On the other hand, as shown in the right diagram of Fig. 1, when we use MWT, we can obtain low sheet resistance of the IGZO treated by MWT due to increase of temperature from inside of the IGZO molecule, thereby oxygen vacancies can be generated at inside of the IGZO layer without reaction with surrounding H<sub>2</sub> and O<sub>2</sub> gas or water vapor.

After the treatment process of both RTA and MWT, the sheet resistance of the IGZO showed a tendency to decrease as time and temperature increased. Especially, in Fig. 2(a),
at 600 °C, when increasing the annealing time, the sheet resistance of IGZO was decreased, and the minimum sheet resistance was 30.2 Ω/□ at 90 s. This decrease in sheet resistance can be explained by an increase of the carrier concentration caused by an increase of oxygen vacancies [14]. Generation of oxygen vacancy attributes to excite two electrons on the conduction band minimum, which indicates the formation of a shallow donor level. Note that oxygen vacancies generated at the shallow donor levels increase the carrier concentration and reduce the sheet resistance [14]. In contrary to the sheet resistance trend of the IGZO annealed above 800 °C, a reversal of the sheet resistance in which the annealing time increased was observed, which is because oxygen vacancies decrease when the metal cations of IGZO react with the surrounding H₂, O₂, and H₂O. Since temperature of the IGZO molecule was increased by RTA, the reaction of H + O₂ (g) → OH⁻ + e⁻ occurs in the IGZO film. The O-H group generated in this way is combined with metal ions, which leads to decrease in oxygen vacancies [15]. The effect of grain boundary scattering also causes a decrease in conductivity of the IGZO annealed by RTA [16]. Fig. 2(b) shows the sheet resistance was saturated in the range of 100 to 200 Ω/□, when the MWT time was 10 s, due to lack of time for the IGZO treatment. As the treatment time was increased up to 60 s, we observed the gradually decrease of the sheet resistance in raise power of MWT due to the temperature increases from the inside of the IGZO molecule [17], the increased oxygen vacancies without reaction with the surrounding gas. As a result, the minimum sheet resistance of 15.18 Ω/□ was manifested for the IGZO films treated by MWT at 1,000 W for 60 s. On the other hand, when the treatment power and time was above 600 W and over 120 s, the sheet resistance of IGZO was increased. It might be that sufficient heat energy is transferred at 800 W for 180 s to the molecular outer shell, which reacts with the surrounding gas or water molecules to reduce oxygen vacancy. The effect of grain boundary scattering also caused an invert of sheet resistance, similar to the results of RTA [16]. In order to further examine electrical characteristics of the IGZO, we analyzed the current density of the IGZO by measuring current—voltage (I−V) curve using Keithley 4200 SCS. Figs. 2(c) and (d) show the current density of the IGZO after RTA and MWT, at −0.65 V, respectively. As a result, we found that current density in both samples indicates a trend consistent with the sheet resistance. In the case of RTA samples, below 600 °C, increase of the current density was observed when raising annealing temperature. On the opposite, in the IGZO annealed at 800 °C, the current density was decreased over 120 s. Then, in the MWT experiment, we observed the increase of current density in the raised treatment power because of a generation of oxygen vacancies. As increasing the treatment time over 300 s, the current density is inverted with raising power of MWT, which might
be explained by effect of grain boundary scattering [16]. As a result, current densities for samples after RTA and MWT are inversely proportional to the sheet resistance results.

Then, in order to deeply understand a conduction property of the IGZO films on p-Si substrate, we investigated a change of oxygen vacancy in the IGZO layer by re-plotting the \( I - V \) curves with trap-limited space charge limited conduction (SCLC) and trap assisted tunneling (TAT) model, as shown in Fig. 3. First, in the case of the IGZO/p-Si structure after RTA, the conduction property is well-matched with trap-limited SCLC model, which is given by following equation, \( J = 9\mu \varepsilon \varepsilon_0 V^2 / 8L^3 (\theta / \theta + 1) \). Where \( \varepsilon_0 \) is the vacuum dielectric constant, \( \varepsilon \) is relative dielectric constant, \( \theta \) is the ratio of electrons in the conduction and the electrons captured by the traps. As shown in Fig. 3(a), where the \( I - V \) curves follow an ohmic behavior due to totally filled with injected electrons. Because the electron capturing inside the trap no longer exists, as it is totally combined with surrounding oxygen gas, the flow of electrons is free from the influence of traps and electrons can reach the counter electrode much easier and thus the IGZO resistance switches to a low resistance [18]. In addition, it appears that deep donor or electron-trap states are calibrated to shallow donor states by the annealing process [19]. Then, Fig. 3(b) shows the \( I - V \) curve of MWT, following an ohmic behavior with some MWT conditions, i.e., 1,000W/20s and 1,000W/60s. This is because, like RTA, the oxygen vacancy acting as a trap is filled due to the combination with surrounding gas and the states of deep donors (or electron traps) changed to shallow donor states after MWT. Therefore, using MWT processing makes IGZO less sensitivity to charge trapping [20], and the charge transfer between the IGZO and p-Si occurs by the SCLC model. On the other hand, when using a lower MWT power than 1,000 W, as shown in inset in Fig. 3(b), the TAT is dominant between the IGZO films and p-Si layers, resulting from the generated abundant traps via MWT process, and is given by following equation, \( J_{\text{TAT}} \propto \exp(-8\gamma \sqrt{2qm^*}(\theta_B)/3hE) \). Where \( q \) is the charge of an electron, \( h \) is Planck’s constant, \( m^* \) is the effective mass of an electron, \( \theta_B \) is the barrier height, and \( E \) is the electric field. This suggests that the TAT predominates at insufficient energies for the deep donors or electron-trap states to change to the shallow donor states. When compared to the power of MWT, oxygen vacancies are distributed from the inside of the IGZO film rather than that of the outside due to temperature-rise effect from the inside of the IGZO at higher power of MWT. Also, the amount of oxygen that can react with the IGZO is relatively low due to the IGZO based device was inflicted too low thermal energy to break bonding of surrounding gas. Therefore, gas in the exterior hardly diffuses into the IGZO layer compared to the RTA process. However, when a certain power is reached in the film, the thermal energy reaches the outside of the IGZO molecule, allowing it to react with the surrounding gas, thereby reducing the amount of trap. This is consistent with previous studies that oxygen vacancies decrease as microwave power increase [21].

Next, in order to confirm the structural characteristics of the IGZO after RTA and MWT, we analyzed the XRD peaks. Fig. 4 shows a XRD pattern of the IGZO after RTA, MWT, and as-deposited sample, which has the lowest sheet resistance, respectively. We commonly observed that peaks related to amorphous IGZO film appeared for MWT and as-deposited samples, while after RTA, it is found that IGZO film is poly-crystallized to (009), (104), (110), and (0018) planes, which are consistent with previous studies [22]. To more deeply study the XRD peaks, we calculated inter-planar spacings (d) using Bragg’s law and compared d with the Power Diffraction Pattern Database (PDF) #38-1104. The table in inset of Fig. 4 shows an average of grain size and FWHM of all samples. The grain of IGZO after RTA is bigger than these of the as-deposited IGZO film and the IGZO film after MWT. Also, each FWHM of the IGZO after RTA was the lowest due to crystallization while annealing in high temperature. These results show that sufficient energy to crystallize the IGZO film is applied to the device through the RTA process. The crystallinity of IGZO typically has an effect on the sheet resistance. Nomura, K. et al. reported that crystallized IGZO shows low sheet resistance owing to reduced trap states and formed the shallow donor level [23]. As a result of the measured sheet resistance, however, samples processed by MWT showed lower sheet resistance than RTA. These higher sheet resistances on RTA samples were because the concentration of oxygen vacancy which supplied the shallow donor level was decreased by the reaction with oxygen. In the
FIGURE 4. XRD patterns of and the as-deposited IGZO films and the IGZO films after RTA and MWT. (Inset) Average grain size and FWHM of the IGZO films.

case of MWT, since IGZO molecules were heated from the inside to the surface unlike RTA [24], oxygen could not react to the surface of IGZO molecule, which was not heated yet, resulting in low sheet resistance. Therefore, even though MWT samples showed less crystallinity than RTA, better conductivity on MWT was observed owing to the restricted reaction between oxygen and the surface of IGZO.

In order to evaluate the IGZO films as TCO, we measured transmittance and band gap, in the wavelength range of 300 to 1,200 nm using UV-Vis spectrophotometer. Figs. 5 (a) and (b) show a transmittance of the IGZO films after RTA and MWT in the visible range of 380 to 700 nm. Also, both enlarged inset figures in (a) and (b) show the trend of increasing transmittance over 75 % as the treatment temperature raises. These results represented that the fabricated IGZO had enough transmittance as TCO. So as to numerically analyze the IGZO, optical band gap energy ($E_g$) was examined by Tauc model (Figs. 5(c) and (d)) [25]. Fig. 5(c) shows the $E_g$ of the IGZO after RTA. As a result, an increase of $E_g$ in increasing the annealing temperature was observed due to the Burstein-Moss effect [26], [27]. When the carrier concentration exceeds the density of state at the conduction band edge the Fermi level moves toward the conduction band. Therefore, an electron at the valence band maximum does not elevate into the conduction band minimum due to Pauli’s exclusion, which results seem that the $E_g$ has widened [28]. Similar with the IGZO after RTA, the $E_g$ of the IGZO after MWT gradually increased as the power and treatment time increased. In addition, in order to intuitively confirm the transparency of the IGZO films, we took optical images for the 36 nm-thick IGZO films having lowest sheet resistance of the IGZO in RTA and MWT, on quartz, as shown in inset of Figs. 5(c)-(d). As a result, we observed the high optical transparency in both the IGZO films after RTA and MWT in range of visible region.

Finally, in order to evaluate the feasibility of IGZO as TCO for solar cell application, $J_{sc}$ absorbed in Si substrate was analyzed using a PV light house simulator depending on the
annealing conditions and methods for IGZO. Fig. 6(a) shows a schematic structure of IGZO based solar cell. Fig. 6(b) shows the $J_{sc}$ absorbed in Si substrate of Air/IGZO/Si substrate depending on the annealing conditions. Region 1 and 2 in Fig. 6(b) denoted $J_{sc}$ of IGZO treated by MWT at 200 W for 10 s and at 600 W for 60 s, respectively. In the case of IGZO after RTA at 800 °C for 30, 60, and 90 s, $J_{sc}$ was lower than other condition because destructive interference in the visible region was less than the others. When the temperature was increased, transmittance shifted toward long wavelength, which resulted in the change of the wavelength range that showing maximum transmission by the interference. The maximum $J_{sc}$ of 19.71 mA/cm$^2$ was observed on IGZO after RTA at 400 °C for 90 s. Like RTA, the $J_{sc}$ of IGZO after MWT at 800 and 1,000 W showed lower than other treatment condition. Region 1 shows the maximum $J_{sc}$ of 19.79 mA/cm$^2$ was manifested on IGZO treated by MWT, which was higher than that of IGZO after RTA. Through the transmittance of the IGZO annealed at 800 °C for 60 s is the highest, Fig. 6(b) shows its $J_{sc}$ is low due to reflectance from substrate and absorption from IGZO layer increase. Therefore, IGZO treated by MWT at 600 W for 60 s has an excellent $J_{sc}$ due to it has a balance between absorption and reflectance. Moreover, in the point of view of the solar cells, sheet resistance of TCO was important for collection of photogenerated current. Therefore, region 2 would show a better performance due to low sheet resistance (21.8 Ω/□), even though it represented slightly lower $J_{sc}$ of 19.75 mA/cm$^2$. Therefore, we demonstrated that IGZO after MWT results in a better performance due to less stress and fast speed than IGZO annealed by RTA.

IV. CONCLUSION
In this study, the electrical characteristic and optical characteristic of IGZO after RTA and MWT was analyzed. In both RTA and MWT, resistance and band gap change due to oxygen vacancy occurred, and mechanism was confirmed. However, RTA has the disadvantage of taking a long time and applying thermal stress to the device, whereas in the case of MWT, treatment was possible within a short time and efficiently. In addition, we evaluated the feasibility of the fabricated IGZO for solar cell application. As a result, the lowest sheet resistance of 21.8 Ω/□ was observed on IGZO treated by MWT at 600 W for 60 s, which was a lower sheet resistance of IGZO annealed by RTA. In addition, we obtained high transmittance of about 75 % in the visible range for all samples. According to the electrical and optical characteristics of IGZO, IGZO treated by MWT at 600 W for 60 s showed high $J_{sc}$ of 19.57 mA/cm$^2$ in solar cell. Therefore, it was confirmed that thermal treatment using MWT can achieve better electrical properties than RTA while applying less thermal damage to devices using IGZO as TCO.

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