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Rapid Thermal Annealing effects on the electrical and structural properties of the AZO thin film deposited at a room temperature

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ABSTRACT :Transparent and conductive Al-doped Zinc Oxide (AZO) thin films were prepared on a glass substrate at a room temperature (R.T) by the RF magnetron sputtering method, and then annealed at the temperature of 600℃ in the 98% N_{2+},2% H_2 ambient gas a with the rapid thermal annealing.
Rapid thermal annealing effects on the electrical and structural properties of AZO films have been investigated. The 600℃ temperature annealing with different annealing time efficiently affects the structure characteristics and the electrical properties of AZO thin films. The films were characterized by X-ray diffraction (XRD), scanning electron microscope (SEM), hall measurement, and UV/VIS spectrometer.

Keywords: Al-doped ZnO Thin Films, Rapid Thermal Annealing, RF Magnetron Sputtering, XRD

Ⅰ. INTRODUCTION

The transparent conducting oxide (TCO) thin film is important as a photoelectric cell that uses light.
It is applied to the transparent electrode that is used in the organic light-emitting material, solar cell, electromagnetic wave shielding film, although its applicability may be variable depending on the applicability of TCO. [1,2]
Currently, the most widely used material for TCO thin film is indium tin oxide (ITO), which has advantages in its excellent optical and electrical properties but many disadvantages such as the material expense increase due to the short supply of the raw material (In), difficulty of deposition at a low temperature, deterioration in the hydrogen plasma, reducibility of In and Zn. [3,4]
However, considering the transparency and electrical resistivity of ITO, it is true that there is almost no candidate to replace ITO among the TCO thin films known so far. But, as a material to overcome the disadvantages of ITO, SnO_2, AZO (Al-doped ZnO), FTO (Fluorine doped Tin Oxide) are mentioned.
This research studied on the manufacture of the Al-doped ZnO thin film as a material that can be used for the TCO material and can reduce the manufacture process.
In particular, the Al-doped ZnO(AZO) thin film that is deposited on a transparent glass substrate attracts much attention because it can be developed to the transparent electrode of the organic light-emitting cell, such as the liquid crystal display & plasma display, and solar ray emitting cell, due to its high chemical and physical stability, optical transmittance higher than 85%, low electrical resistance of $1X10^{-3}$ Ωcm, 3.2 eV energy band gap, and thermal stability in the manufacture process. At the same time, the low raw material price helps it be considered as one of the candidates to replace ITO. The AZO thin film can be manufactured as a crystalline thin film by various methods, such as MBE (molecular beam epitaxy), RF magnetron sputtering, chemical vapor deposition, and pulse laser deposition. [5-7] But,

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in most of these processes, the crystal phase is formed first and then the annealing treatment is applied, and there are many studies on this. [8]
In general, to apply for a flexible display or a solar cell as a substrate material, a low temperature process is necessary below 150℃. However, in the TCO thin film formed at a low temperature, local crystallization can occur depending on the change of the exterior environment.
Hence, we need a method that overcomes this disadvantage and crystallize a high quality TCO, to promote the crystal growth, and one of those methods is to apply the annealing. There are various annealing methods, but unlike other simple annealing methods, rapid thermal annealing (RTA) is known to have a larger effect of characteristic improvement on the thin film, by the flash annealing, than other general annealing methods. [9]
Therefore, in this research, after making a new AZO thin film on a glass substrate using the RF magnetron sputtering that is now the most frequently used in the semi-conductor process, RTA was applied to verify the material property of the AZO thin film in accordance with the annealing atmosphere and devices, using a mixture gas of 98% nitrogen and 2 % hydrogen, and high purity 99.999% oxygen, which were not used in papers previously, at the temperature of 600℃ with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min. Then, the transmittance and surface change of the treated Al-doped ZnO (AZO) thin film were observed, and at the same time changes of the electrical and optical characteristics were investigated, and also the possibility as the transparent electrode and ultra-violet emitting cell was considered.

Ⅱ. EXPERIMENTAL METHOD

Ⅱ.1 Experiment and analysis of thin film

In this research, the AZO film was under deposition at a room temperature by the RF magnetron sputtering method, and as a result an amorphous AZO thin film was developed. In general, in the AZO thin film, the C-axis (002) plane orientation is known to occur first, but the final characteristic of a thin film is heavily affected by the substrate temperature, pressure of argon and oxygen, power, and content of Al. There could be several reasons, but the process of this paper developed an amorphous AZO thin film.
And, to crystallize the amorphous thin film, an amorphous AZO sample received the RTA treatment using a mixture gas of 98% nitrogen and 2% hydrogen, at the temperature of 600℃ with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min, and the crystallized samples were used for manufacture.
As a substrate, no-alkali Corning glass was used, and it was under the ultra-sonic cleaning in the order of acetone, methanol, distilled water, 5 minutes each. As a target, a 2 inch thick ZnO (purity 99.99%) doped with 0.25 inch aluminium oxide (2 Wt%) was used.

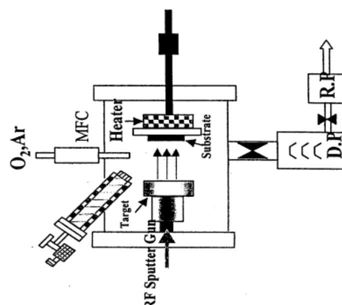

Fig. 1. A RF magnetron sputtering system

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Figure 1 shows a schematic drawing of the sputtering device used in this research. The initial vacuum level in the chamber was lowered to $1.5X10^{-6}$ Torr by a rotary pump and a turbo pump, to prevent contamination of the thin film caused by the residual gas. As the reacting gas at the thin film deposition, Ar 50 sccm was used, and the pressure in the chamber while the film formation (?) was on progress was kept at 20 mTorr by the butterfly control. The distance between the target and the substrate was 7 cm and the RF power of 100 W was supplied to conduct the experiment. The working pressure was $2.0X10^{-2}$ Torr in the experiment, and the sputtering of the AZO thin film was deposited for about 15 minutes to obtain about 600 nm thin film. This was measured by using the KAL-TNCO instrument, and repeating the measurement 5 times all gave similar thickness.
And, to crystallize the deposited amorphous thin film, the Ultech RTA device was used to inject the mixture gas of 98% nitrogen and 2% hydrogen at the C/G Pressure : 495mm Torr. Here, in the RTA process, the temperature elevation was not over 20 sec and each sample was annealed at the temperature of 600℃ with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min, and the crystallized samples were used for manufacture.
To analyze the grown thin film, an X-ray diffraction experiment was conducted using the Cu-Kα radiation (wavelength 1.54 Å), and the fine shape of the thin film was observed by using FE-SEM and AFM. The electrical property of the thin film was measured by using the hall measurement device and the area resistance measuring instrument, and the absorption rate of the thin film was measured at the wavelength range of 300-900 nm by using the PL instrument. Also, the optical transmittance at the visible light region was analyzed by using the UV visible NIR Spectrophotometer.

Ⅲ. RESULT AND DISCUSSION

The thin film developed by the optimized magnetron sputtering is known to exhibit the C-axis (002) plane orientation first even at a room temperature deposition, and metal targets exhibit high deposition rate under reactive sputtering, but the final characteristic of the thin film is affected by the substrate temperature, pressure of argon and oxygen, power, content of Al and, thus, can be different, and in this research an amorphous thin film was formed. To observe the crystal phase and phase change, samples at a room temperature and those samples crystallized by applying RTA under the atmosphere of 98% nitrogen + 2% oxygen at the temperature of 600℃ with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min, were used for manufacture. Figure 2 shows the analysis result of XRD that was annealed in the above time intervals. The X-Ray analysis shows that the amorphous AZO thin film exhibited different degree of crystallization as the annealing temperature increased, irrespective of the annealing atmosphere.
The component ratio of the AZO thin film used in this experiment was 8:2, which means that the Al-doped ZnO phase is formed as the crystallization progresses at the time of RTA treatment. In other words, the peak that occurred in AZO was the peak that occurred at 34.3-4, and it could be verified as the (002) peak from the JCPDS (36-1451) card.
As the annealing temperature increases, the amorphous Al-doped ZnO phase begins to show crystallized state, which becomes the reason to enhance the crystallizability along with the growth of the C-axis and the reason to grow the crystallizability of the Al-doped ZnO thin film.

As for the RTA treated sample, the XRD peak of the AZO thin film has the hexagonal structure (a= 3.25 Å, c=5.207 Å) that orients very highly to the (002) direction, their principal direction. [10,11,12].
As the RTA temperature increased, the diffraction peak of the AZO (002) increased and exhibited the maximum value with the 3 min treatment at the temperature of 600℃. This provides a variable for the improvement of the crystallizability along with the result that the peak of crystallizability increased in the direction of C-axis as the annealing temperature increased. In general, the sputtered ZnO thin film grows with the direction chosen in the crystallographical C-axis that is perpendicular to the substrate, which is because ZnO has tetrahedral coordinates due to the sp^3 hybridized orbits and the (001)-plane has the lowest surface free energy. This experiment also showed a strongly chosen direction of (0001) on the glass. [13]

However, after 6 min at the temperature of 600℃, when the annealing temperature increased, the diffraction peak showed a trend of decreasing strength. From this, we can see that the peak of crystallizability decreases in the direction

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of C-axis as the annealing temperature increases. As reasons of the decreasing crystallizability, penetration of oxygen into the ZnO:Al thin film due to the reversed spread of oxygen atoms, worsened crystallizability due to the adsorption on the surface, and existence of many flaws on the thin film can be considered. [14]
As can be seen from Fig. 2(a), it is difficult to judge clearly if the crystallizability of the AZO thin film is good only by the pattern of the AZO thin film, but just it is possible to verify approximately where the crystal direction is.
The result of the annealing in the reduction atmosphere shows that as the annealing time increased, the pak width at half height (PWHH) decreased from 0.68°to 0.38, and the height of the (002) peak increased abruptly.
Particularly, the AZO thin film annealed in the time of 180 sec has the highest peak and the least FWHH (0.38°), and this condition seems to affect largely on the crystallizability improvement of the ZnO thin film. However, when the annealing time was over 180 sec, FWHH increased,
Perhaps, this is because particles grew again and many flaws were created on the thin film.

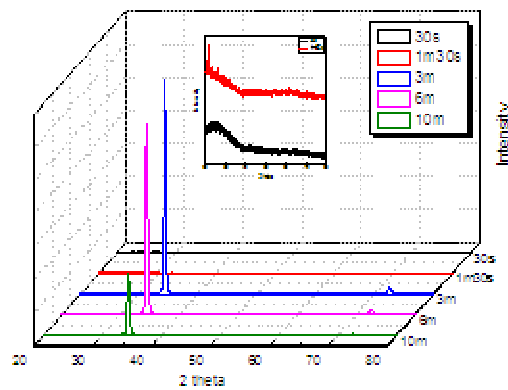

Fig. 2. XRD Patterns of ZnO:Al thin films at 600℃ with a different RTA Treatment and 98% N_2, 2% H_2 ambient gas.

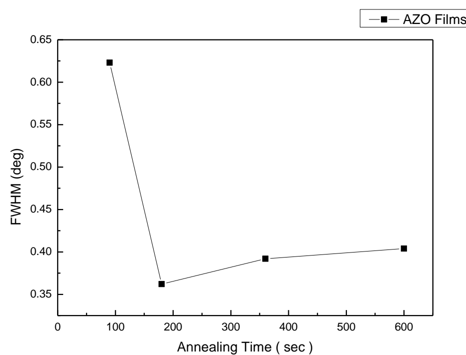

Fig.3 FWHM of ZnO films as a function of different annealing time

Figure 3 shows the SEM-measured shape of the AZO thin film that was obtained by applying RTA to the sample crystallized by annealing with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min, at the temperature of 600℃. Figure 3(a) shows a sample of non-RTA treated and Fig. 3(b),(c),(d),(e) show samples of RTA treated. The sample of non-RTA AZO thin film is the amorphous type in which the particle size is not observed.
The sample of the non-RTA AZO thin film show small ZnO particles, from a few to 20 nm size, formed on the surface. The particles are too small to allow the X-ray diffraction analysis.
We can see that the shape of the surface particles gradually crystal grow as the annealing time increases.
The surface cluster size of the ZnO:Al thin film is shown to grow, which is explained by that the heat energy in the crystallizing process was spent in the grain growth. Hence, if the annealing time increases in the reduction atmosphere, the crystal grain increases because of the decrease of defects, which improves the crystallizability, and this coincides with the XRD result.

But, judging from that the grain size decreases after 6 min at 600 ℃, recombination process may have occurred. The reason may be explained by the grain boundary relax model caused by the growth of the grain size, and with this model the growth of the grain size is known to cause the tensile stress on the thin film. [15, 16]

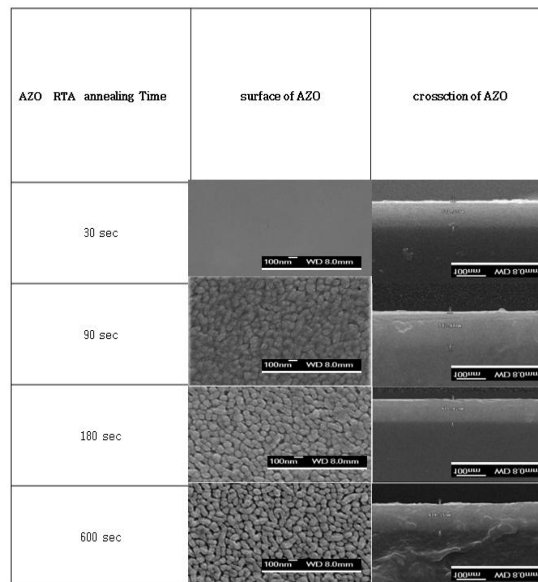

Fig. 4. (a) SEM images of AZO thin films annealed at 600℃ in with RTA 98%N_2+2%H_2

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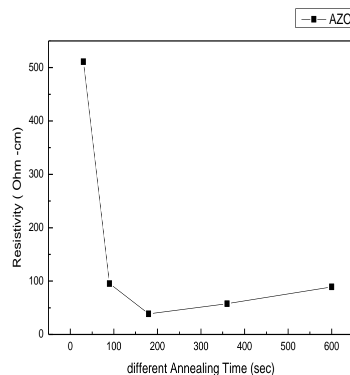

Fig. 5. Change of the resistance of the ZnO films with changing RTA heat treatment temperature In 98%N_{2} + 2% H_2 gas

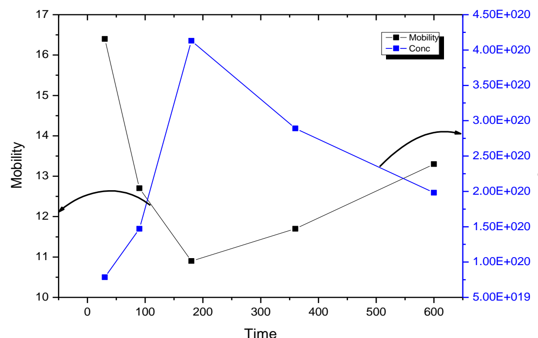

Fig. 6. Change of the Mobility and the Carrier concentration the ZnO:Al films with changing RTA heat treatment Temperature In 98%N_{2} + 2% H_2 gas

Figures 5, 6 and show the variation of the resistivity, carrier concentration, and atomic mobility, depending on time and the gas atmosphere, when the Al-doped ZnO thin film was RTA treated.
In general, the ZnO film that was deposited as an amorphous mostly shows high resistance outside the measuring boundary or untrustable results due to the unstable electrical property of samples. But, the thin film that was deposited at a room temperature was annealed at 600℃ with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min. In general, ZnO exhibits the n-type semi-conductor characteristics because of Zn penetration and the hydrogen impurities.
Annealing at the temperature of 600℃ with the time intervals of 30 sec, 1 min 30 sec, 3 min, the resistivity was reduced in the atmosphere of 98% nitrogen + 2% hydrogen, and at the annealing temperature of 600℃ with the time interval of 3 min, the electrical property is the best with the lowest resistivity and highest mobility. But, after 3 min of the annealing at 600℃ under the 98% nitrogen + 2% hydrogen gas, the resistivity all increased. This is because from a potential barrier that is formed for the crystal grain system in the annealing process, negatively charged oxygen species are desorpted and they play the role of trapping site for carriers. [17]` These negative charges develop a depletion region and reduces the carrier concentration and the resistivity. In the annealing process of the 98% nitrogen + 2% hydrogen gas, hydrogen atoms have the effect of removing the depletion region, around the crystal grain system, by the

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passivation of the crystal system face. The mechanism behind that the hydrogen removes the depletion region to increase the carrier concentration is known to be the boundary-passivation effect, which is that the hydrogen impurity atoms surround the Zn grain ions. [18.19] The reason that the resistivity of the AZO thin film decreased under the 98% nitrogen + 2% hydrogen gas is considered to be that the carrier concentration increased because the hydrogen atoms surrounded the grain border and the Zn ions. This result agrees with the XRD analysis result and allows the observation of the crystallizability change in accordance with the annealing process condition.

In general, the roughness of a transparent electrode is largely affected by the phase differences and fine structure of the transparent electrode, and affects the optical property.

The AZO thin film deposited at a low temperature has a very uniform and flat surface as reflected by the surface roughness of 0.63 nm. However, since the AZO thin film exists in an amorphous form, its transmittance and electrical conductivity is so low that it is not appropriate to be used for the transparent electrode substrate. An experiment shows that the surface roughness is affected by the RTA treatment, and its rms value is considered to increase as the annealing temperature increases. The experimental result shows that in the RTA treatment at 600℃ at the atmosphere of nitrogen + hydrogen, the sample rms value of the surface roughness is relatively low as 0.98 nm for annealing from 30 sec to 6 min, but at a higher temperature the rms value keep increasing even to the value that is considered inapplicable to electronic elements. The AZO thin film that grew by the sputtering method has incomplete bonds on its surface, such as broken bond or dangling bond, but in the AZO thin film that was post-annealing treated at the oxygen atmosphere, it is considered that broken bonds or dangling bonds are reduced due to the oxygen absorption on its surface and crystal particles are re-crystallized very uniformly to achieve a relatively low roughness. [20]

For a high surface roughness, there is a high chance that poor pixels may appear, and for a high rms roughness value, etching can be problematic due to diffused reflection to light exposure and at the same time there can be dark spots on the surface due to the concentration of the applied electric power. [21] Hence, if the surface roughness increases, scattering of the incident light into the thin film also increases, which deteriorates the optical property of visible light. [22]. Therefore, the regulation of the dense particles and rms is considered to be important in the manufacture of touch panel or OLED. As a result of the experiment, the sample that was RTA treated from 30 sec to 6 min at the nitrogen + hydrogen atmosphere has a potential to be a good element, because it is not expected to exhibit these problems at manufacture.

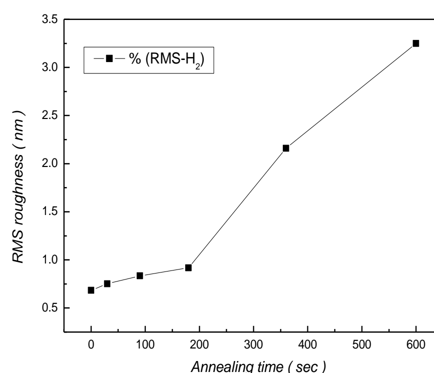

Fig. 8. Changing the root mean square Surface morphology according to the annealing Temperature

Figure 9 shows the transmittivity of the AZO thin film, in accordance with the post-annealing process, measured at the 200-800 nm range by using the UV-spectroscopy meter, and the optical transmittance measured with respect to 550 nm.

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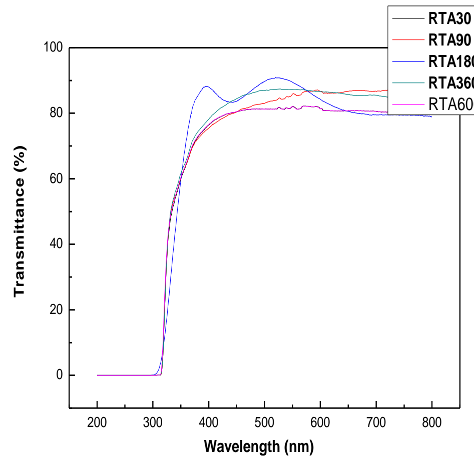

Fig. 9. Optical Transmittance at 550nm with respect of glass substrate with changing RTA heat treatment Time (min) at Various Temperature In H_2 ambient gas

The experiment shows that all annealed samples exhibited the optical transmittance higher than 80% at the visible light range (λ= 550nm). The transmittance of an amorphous Zinc-Al oxide thin film to the visible light of 550 nm wavelength was measured and displayed in Fig.9. Also, in the case that the thin film was annealed at the atmosphere of nitrogen + 2% hydrogen, all samples exhibited higher than 80% crystallizability and transmittance, but the transmittance showed a variation depending on the annealing time. The sample that was RTA treated for 3 min at 600℃ under the atmosphere of nitrogen + hydrogen exhibited the highest transmittance of 89%, but the transmittance showed a decreasing trend for annealing longer than that. Although the correct reason cannot be known, it is considered that the replacement of Zn by Al atoms occurred actively in the RTA process, and in the annealing the potential effect occurred on the thin film less frequently in the high temperature elevation case than in the low temperature elevation case, which eventually resulted in reduction in the electron effective concentration and thus increased the electrical resistivity.
The optical band gap was computed in accordance with this change of carrier concentration. The transmittance (T) is given by

$$T=(1-R)^2 \, EXP(-\alpha d),$$

where R is reflectivity, α is the absorption coefficient, d is the thin film thickness. In the ZnO thin film, because reflection can be neglected in the absorption limit, the absorption coefficient α is given by

$$\alpha= In(1/T)/d.$$

Also, because the ZnO thin film has the transition band, on the assumption that the absorption limit transition probability is constant, αbecomes

$$\alpha \propto (h\nu - E_g)^{1/2}$$

where E_g is the transition energy gap, and hν is the photon energy. Therefore, the optical band can be obtained, if we measure the transmittance, and draw a graph of α^2 and hν and extrapolate the linear region to get the x-intercept. Fig 10 shows the change of the optical band gap in accordance with the annealing temperature.

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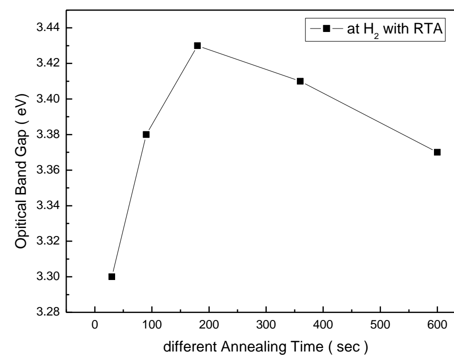

Fig 10. Variation of the optical band gap as carrier Concentration changed

The annealed thin film under the atmosphere of 98% nitrogen + 2% hydrogen, exhibited the increased optical band gap at the annealing time of 3 min, but samples that were annealed longer than that exhibited decreased optical band gap. The variation of the optical band gap as the annealing temperature varies is displayed in Fig, which can be explained by the Bernstein -Moss shift. According to the Burstein -Moss shift theory, the optical energy gap increases as the free electron concentration increases. [23] It is speculated that as the annealing temperature increased, more Al atoms replaced Zn atoms and the thin film was composed of the substitutional site, and the donor electrons emitted by Al atoms affected the Burstein - Moss shift.
Also, it is judged that this transmittance change is affected by complex elements such as change of the crystal growth and surface roughness as well as the electrical property. Because the crystal grain that constitute the ZnO:Al thin film is itself a structure of oxygen deficiency, as explained before, the atmospheric annealing ignites the surface activation, and the RTA treatment reduces much quickly the oxygen holes on the thin film, such as oxygen vacancy, Zn interstitial, and Zn vacancy, to change the stoichiometric structure and as a result to reduce the photo absorption due to defects. Depending on the annealing condition, it is possible to manufacture a transparent electrode that allows control of the fine structure and exhibits excellent optical characteristic. [21, 22]

Ⅳ. CONCLUSION

This experiment grew the Al-doped (2 wt%) ZnO thin film on a glass substrate by the RF- Magnetron sputtering method, to investigate its applicability to the transparent electrode substrate. In the experiment, an amorphous thin film was deposited on the glass at a room temperature, and after fixing other elements, the thin film deposited at the RTA device was annealed at 600℃, under the atmosphere of nitrogen and hydrogen, with the time intervals of 30 sec, 1 min 30 sec, 3 min, 6 min, 10 min. After the annealing, the electrical property, optical property, and change of the structural characteristics of the ZnO: Al thin film were observed, as a transparent electrode.

1. It was possible to verify that the resistivity of the sample decreased until 3 min, at 600℃ under the atmosphere of hydrogen, but after 3 min the resistivity increased again, which may be explained by that as the annealing time increased Al atoms replaced Zn atoms more actively to increase the carrier concentration, however at a high temperature the existing oxygen vacancies were filled to decrease the carrier concentration and hence the resistivity increased again.

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As a result, at the annealing condition of 3 min under the atmosphere of 98% nitrogen + 2% hydrogen, the sample exhibited higher than 85% optical transmittance to visible light and an excellent resistivity of $1.9X10^{-4}$ Ω.cm. Also, by exhibiting a low roughness, the sample demonstrated its applicability to the solar cell and opto-electronic device.

2. In the event that the thin film is deposited by sputtering, the compressive stress of the thin film is developed by the atomic peening effect, and until 3 min of RTA treatment at 600℃ the thermal expansion coefficient of the substrate seems to affect the AZO thin film. However, after 3 min of RTA treatment at 600℃, defects seems to be developed because of re-crystallization and growth of the grain size.

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