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Structural and Optical Properties of Al-doped ZnO Thin Films Prepared by Ion Assisted Deposition Method by Post-Step Post-Annealing

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ABSTRACT: AZO thin films were synthesized on glass substrate by ion-beam-assisted molecular deposition. In order to improve the crystallinity, thermal annealing process were performed in the temperature range of 400-700 ℃ for 30min in oxygen atmosphere. The effect of the annealed conditions on the electrical, the optical and the mechanical properties of AZO thin films were investigated. The optical properties of the AZO thin films were improved when the AZO thin films were grown by ion beam energy of 100 eV condition and the subsequent annealing condition at 600 ℃ for 30 min in 99 % oxygen gas atmosphere.

KEYWORDS: AZO, ion-beam-assisted MBE, annealing, optoelectronic devices, LED

I. INTRODUCTION

Recently in the 21st century, the direction of research on information communication and related materials has been toward ensuring new functionality beyond the existing paradigm, ensuring precision, combination and fusion, due to the development of short-wavelength optical devices and high-frequency electronic devices along with the continuous high-speed growth of the information industry. Since the luminescence phenomenon, one of current flow phenomena in solid materials, was discovered by the British radio engineer Henry Joseph Round in 1907 [1], industry and academia have been interested in developing new luminescent materials and devices. Among them, optical devices in the near-ultraviolet regions have recently been of increasing interest, and semiconductors with a wide band gap have been attracting attention. Although there are many materials, GaN is currently the mostly widely commercialized optical element material and is attracting attention. GaN-based optical devices are monopolized by two companies, Nichia (30%) in Japan and Cree (20%) in the USA, and their use requires payment of a lot of technical royalty fees. For this reason, it is necessary to solve the problem of obtaining original patents [2].

To overcome this problem, studies have recently been actively conducted on blue LEDs based on ZnO which is more stable and economically inexpensive than GaN while having characteristics similar to those of GaN, and interest in these LEDs is growing. ZnO thin films are direct transition-type Group II-IV compound semiconductors which have a hexagonal wurtzite crystal structure and also have a wide band gap of 3.37 eV at room temperature. They are known as materials showing n-type conductivity even in a non-impurity-dope state due to oxygen vacancies or internal lattice defects of Zn atoms in the lattice [3-4]. In material terms, ZnO thin films have large room-temperature exciton binding energy of 60 meV, which is greater than the room-temperature binding energy of GaN (25 meV) and ZnSe (19 meV). Thus, they are capable of emitting light by excitons at room temperature, may use homogeneous substrates, and are thermochemically stable [5-6]. Furthermore, they are more advantageous for actual device fabrication because they can be wet-etched, and are thermally or chemically significantly stable, thus providing devices having good durability [7-8]. Moreover, ZnO has a lattice mismatch of only 0.2% with GaN, and thus the same effect as homoepitaxy is expected even when ZnO and GaN are deposited and grown on each other. In addition, when fabrication of well-known ZnO wafer whose grown is easier than that of GaN crystals is performed, it is possible to pioneer a new industry of new optical devices based on ZnO thin films, and it is also possible to lead the next-generation compound photoelectric ceramics industry by securing unique technology. Accordingly, ZnO is expected to become an important research area. However, in general, non-impurity-doped ZnO thin films essentially have n-type polarity due to their oxygen

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vacancies and defects such as zinc interstitials. For this reason, in order to use them as semiconductor elements or LED light-emitting materials, it is necessary to control the concentration and transmittance of doping carriers. In addition, even though pure ZnO thin films show high conductivity due to the effects of oxygen vacancies and interstitial Zn, highly doped n-type ZnO thin films are required for device fabrication. This is because when non-impurity-doped ZnO thin films are exposed to the atmosphere for a long period of time, the electrical properties thereof change while the stoichiometric ratio of Zn and O changes due to the influence of oxygen, and when these thin films work at high temperatures, they become unstable. Thus, in order to fabricate ZnO thin films that have high transmittance and excellent electrical conductivity in the visible region, studies have been actively conducted to control light emission wavelengths by doping group III elements, such as Al, B, Ga and In, or substituting Zn sites with Mg or Cd to increase band gaps [9-10].

Among current physical vapor deposition methods, methods that are used to fabricate AZO thin films for UV light-emitting devices include RF magnetron sputtering [11], chemical vapor deposition [12], pulse laser deposition [13], molecular beam epitaxy [14], and the like. Among them, a method that is widely used in industry is a method based on an RF or DC sputtering process. The sputtering process has advantages in that large-area deposition is easy and deposition is possible even at low temperature [15]. However, when the sputtering process is used, there is a high possibility that defects occur on the thin film surface, and for this reason, many oxygen vacancies are formed and impurity light emission in the visible region occurs. This causes a problem in the light emitting device and deteriorates the optical characteristics. To overcome this problem, particularly, to improve light emitting characteristics, it is very important to minimize the light emission in the visible region for many defects that are frequently observed in ZnO thin films.

Since many defects are found in p-type ZnO thin films, conditions capable of minimizing the formation of various defects that cause n-type are required [16]. Thus, it is necessary to eliminate impurities in the visible region and form high-quality thin films that can exhibit only UV emission characteristics. In order to realize ZnO thin films as high-performance elements, thin films with good crystallinity should first be preceded. In order to obtain such high-quality thin films, it is an important goal to eliminate internal defects such as oxygen vacancies. This problem can be overcome through a deposition process or a subsequent heat treatment process. Looking at many existing studies, many studies have been conducted growth temperature and thin film thickness are controlled and AZO thin films are deposited at a high temperature of 200°C or higher, after which the crystallinity of the AZO thin films by heat treatment effects, thereby controlling light emission characteristics. However, it is difficult to find a study in which thin films are formed at low temperature and subjected to post-growth annealing.

In recent years, a molecular beam epitaxy method having an excellent property of controlling thin-film crystal growth at low temperature has received attention as a method for improving the crystallinity of AZO thin films, and thus studies on light emission from AZO thin films have been published [17]. Therefore, in the present study, it was attempted to form thin films at low temperature by an ion-beam-assisted molecular beam epitaxy process capable of making the thin films precise, and conditions for light-emitting elements were optimized by changing heat treatment conditions in an oxygen atmosphere. The ion-beam-assisted molecular beam epitaxy process, a low-temperature process, has advantages in that it has excellent reliability and reproducibility, forms an excellent interface by reacting reactive ions with coating atoms, increases adhesion and density, and can produce almost all materials. In addition, a subsequent heat-treatment process for improving crystallinity serves to improve the crystallinity and light emission characteristics of thin films by increasing the island size and reducing lattice stress [18].

The present study attempted to identify the correlation between the light emission characteristics of AZO thin films and the heat-treatment temperatures in an oxygen atmosphere while precisely controlling AZO thin-film deposition at high temperature by ion-beam-assisted molecular beam epitaxy (MBE) deposition. In order to improve the thin-film crystallinity and control the thin-film surface, Al-doped ZnO (AZO) thin films were formed while irradiating low ion beam energy (100 eV), and the physical properties of the AZO thin films were studied by preforming post-growth annealing in an oxygen atmosphere. Then, the structural characteristics of the resulting thin films were analyzed, and the possibility of the thin films as blue light-emitting materials was investigated by optical analysis for the blue light emission mechanism.

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Table 1. Post-growth Annealing Process.

Post-annealing temperature	Temperature rise rate	Maintenance time	Cooling rate	Oxygen concentration
4000°C	200 °C/min	30 min	200 °C/min	99.99%
5000°C	200 °C/min	30 min	200 °C/min	99.99%
6000°C	200 °C/min	30 min	200 °C/min	99.99%
7000°C	200 °C/min	30 min	200 °C/min	99.99%

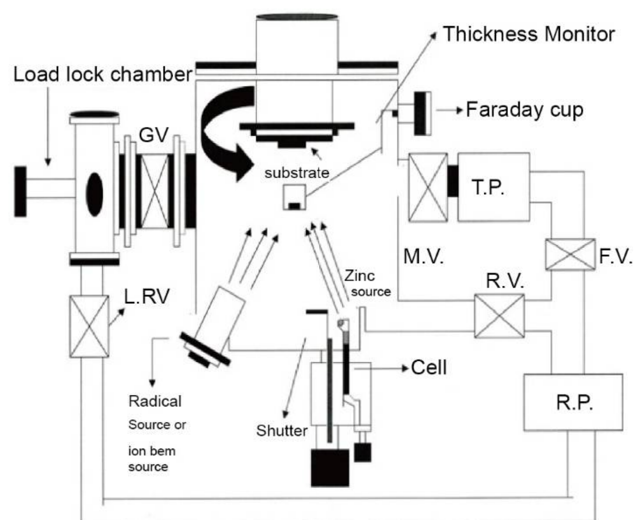

Fig. 1. A schematic diagram of the IBAD system.

II. EXPERIMENTAL METHOD

In the present study, ion-beam-assisted molecular beam epitaxy deposition was used to fabricate AZO thin films applicable to UV light-emitting devices, and irradiation with ion beam energy of 100 eV at room substrate temperature was performed. Thereafter, to fabricate n-type AZO light-emitting materials, annealing was performed in a high-purity oxygen atmosphere system at 400°C, 500°C, 600°C and 700°C at intervals of 100°C for 30 minutes. Experimental conditions therefor are shown in Table 1 above.

Before thin-film deposition, alkali-free glass was used as a substrate and ultrasonically washed sequentially with acetone, ethanol, methanol and distilled water for 5 minutes per washing, followed by drying with a high-purity (99.99%) nitrogen gun. The ion-beam-assisted MBE system used in this experiment was made in-house, and related parts were also made in-house. A Kundensen cell, a member for evaporating AZO thin films in the experiment, was also made in-house. As a crucible for an AZO thin film, a nozzle-free, open-shaped 3-cc BN crucible was used. To deposit an Al-doped ZnO thin film, AZO pellets with a ratio of 2 : 98 were made. As raw materials, ZnO (99.9%, CERAC. U.S.A) and Al_2O_3 (99.9% High Purity Chem, Japan) were used as pellet materials. Based on the literature, Al_2O_3 and ZnO were mixed with each other, and then crushed and mixed by a ball mill for 24 hours. Next, disk-type pellets were made using a cold press, and calcined in a box furnace at 1400°C, after which the pellets were made into an Al-doped ZnO source material.

For deposition of an Al-doped ZnO thin film, 2 sccm of high-purity (99%) oxygen was flushed into a chamber at room temperature for 5 minutes, and then the plasma discharge current was set at 300 mA, and the pellets were placed in the Kundensen cell. Next, for output stabilization of the ion gun, substrate washing and impurity removal from the substrate, the substrate surface was pre-sputtered with energy of 500 eV at 300 mA for 5 minutes. Then, the assisted

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ion beam current was set at 300 mA, an ion beam was irradiated at an ion beam voltage of 100 eV for 5 minutes, and at the same time, an Al-doped ZnO thin film was deposited while the Kundensen cell for evaporation of the AZO thin films was heated to a temperature of 750°C. The deposition rate of the Al-doped ZnO thin film on the glass substrate was measured to be 0.9□/sec by a thickness monitor with a quartz oscillator. While the Al-doped ZnO thin film was deposited until a thin film thickness of 200 nm was measured by the monitor, the Al-doped ZnO thin film was fabricated using ion beam energy. Next, annealing was performed at a temperature ranging from 400°C to 700°C in an oxygen atmosphere. The reason why such thin-film deposition conditions were selected was because, in a previous study experiment, a thin film irradiated with an ion beam (100 eV) showed possibility as a transparent electrode substrate, and when it was improved by a post-growth annealing process, it had a high possibility as a blue light-emitting element.

The ion-assisted molecular beam deposition system used in the present study is shown in Figure 1. The system used in the experiment was configured such that the vacuum level of a high-vacuum chamber can be reduced to 10 Torr or less by a rotary pump and a turbo pump, and that the electron beam accelerating voltage was possible up to 7 kV. In addition, it was designed such that an assisted ion beam can be irradiated while thin films are deposited using the electron beam deposition system. Regarding deposition conditions, the chamber was evacuated to a pressure of 1× 10 Torr by a 1300 l/m rotary pump and a 2000 l/s (Seiko Seiki) TMP pump before deposition. As an oxygen ion beam source, an in-house made cold hollow cathode ion gun was used, and it was made by winding a 1/8-inch copper tube externally in the form of a solenoid such that it can apply RF power with cooling water. Oxygen introduction was performed using a variable leak valve (Varian Co) such that the oxygen partial pressure could be controlled to 10^{-8} Torr-10 Torr. In a process of producing ions with kinetic energy during introduction of high-purity oxygen, a process gas was introduced into an emitter in the ion gun, and when a discharge voltage was applied between an anode and the emitter, plasma was formed due to glow discharge. At this time, ionized gas atoms in the plasma were extracted out of the ion gun due to the potential of the discharge voltage, and the ionized gas atoms accelerated out of the ion gun had kinetic energy and were irradiated onto the thin film surface during deposition. Oxygen gas used as an atmospheric gas in the chamber and Ar gas for producing an assisted ion beam were introduced into the vacuum chamber in constant amounts and at a constant ratio through an MFC (mass flow controller), and the vacuum level of the chamber was measured using an ion gauge and a convection gauge. An annealing process was performed at a temperature of 400°C to 700°C for 30 minutes in order to anneal the grown AZO thin films in a high-purity (99.99%) oxygen atmosphere, and optimal light emission conditions as a function of post-grown annealing conditions were investigated. To analyze the grown thin films, an X-ray diffraction experiment was performed using Cu-Kα radiation (wavelength: 1.54 □), and the surface microstructures of the thin films were observed using FE-SEM and AFM. The electrical properties of the thin films were measured using a Hall system and a four-point sheet resistance meter. The light transmittance in the visible region was measured using an UV-vis-NIR spectrophotometer in a wavelength range of 200-800 nm. In addition, in order to examine the light emission characteristics of the AZO thin films, photoluminescence at room temperature was measured using a He-Cd laser at a wavelength of 325 nm.

III. RESULTS AND DISCUSSION

3-1. Phase Analysis and Characterization under Various Annealing Conditions

In order to identify the crystal structure of the AZO thin film deposited by ion-beam-assisted molecular beam epitaxy, analysis was performed by θ-2θ scanning of the XRD spectrum. In addition, after the deposited AZO thin film was annealed at each of 400°C and 700°C, the changes in the crystal structures in the XRD patterns before and after the annealing were comparatively analyzed.

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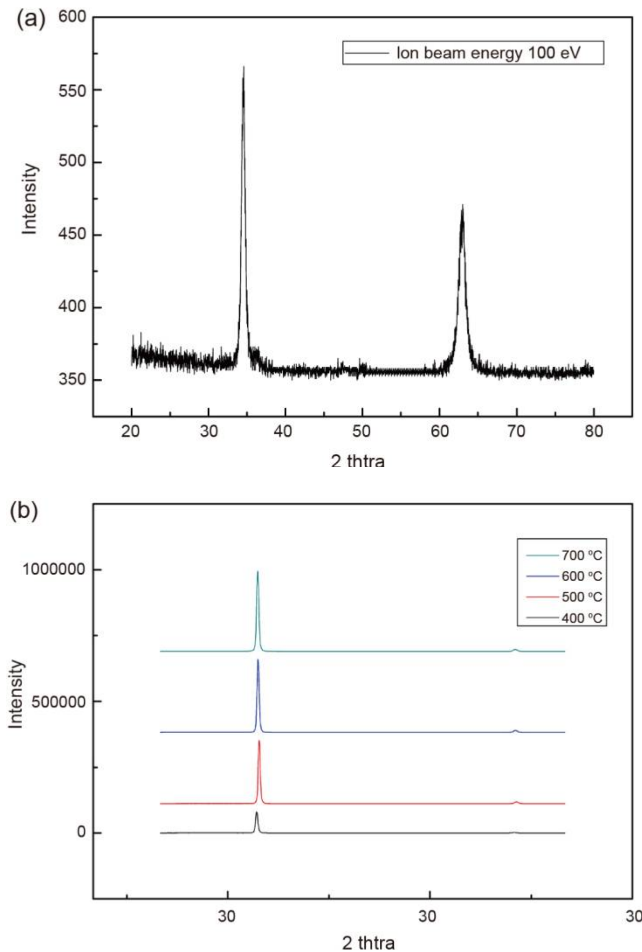

Fig. 2. (a) XRD Patterns of AZO thin films by Ion beam assisted deposition at 100Ev condition. (b) XRD Patterns of AZO thin films different annealing treatment range from 400°C to 700°C in 99% O_2 ambient gas.

First, looking at the state of the thin film irradiated with ion beam energy of 100 eV as shown in Figure 2(a), when the beam energy was 100 eV, XRD spectrum peaks (34.3º and 62.6º) corresponding to (002) and (103) planes were found, and formation of polycrystalline thin films was observed. This indicates that as the kinetic energy of the ion beam incident on the AZO thin film during deposition of the thin film increased, the XRD pattern peak corresponding to the (002) plane increased and a peak corresponding to the (103) plane having a new intensity peak appeared. The reason why the preferred orientation of the AZO thin films deposited by ion-beam-assisted molecular beam epitaxy appeared as the (002) plane and the (103) plane is mainly because of the ion channeling effect [19, 20] and the anisotropic radiation damage effect [21, 22]. In the case of the ion channeling effect, the incident direction of the assisted ion beam has the greatest effect on the preferred orientation, but in the case of the present study, the ion channeling effect by the ion beam irradiation direction was negligible, because the substrate was rotated during deposition of the ZnO thin films. Rather, in the case of the anisotropic radiation damage effect on each crystal plane, the most important factors are the incident assisted ion-beam energy and the momentum delivered per unit volume, and for this reason, as the ion beam energy increases, different radiation damage effects may appear in crystal planes having different atom densities and constituent atom distributions.

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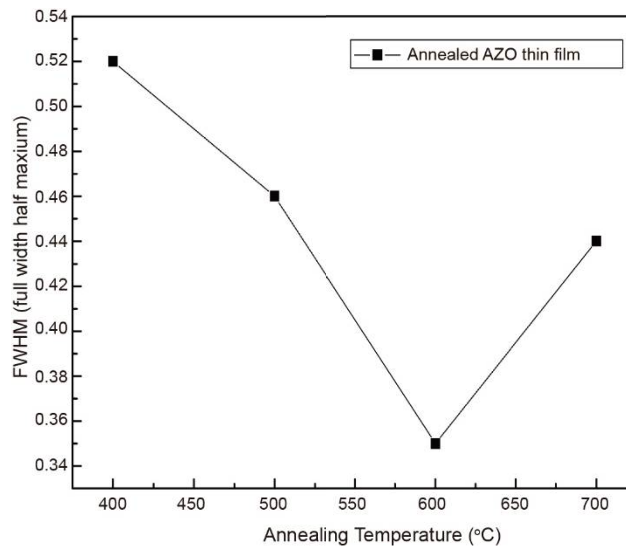

Fig. 3. FWHM of Al-doped ZnO films as a function of different annealing temperature ().

In addition, the ion gun used in the present experiment has a characteristic in that the ion beam current density increases when the ion beam energy is increased. Generally, when low energy or no energy is irradiated, the (002) plane where atomic density is the highest (more dense) tolerates the radiation damage relatively well at low current density, and when the ion beam energy increases and the current density also increases to 100 eV, the atomic mobility increases and also has an effect on the preferred orientation of the ZnO thin films. For this reason, it is believed that the (002) crystal plane and the (103) plane preferentially grow. However, when post-growth annealing in an oxygen atmosphere was performed at a temperature of 400°C to 700°C for 1 hour, only the (002) peak of the AZO thin films was observed regardless of the annealing temperature [23]. In addition, as the annealing temperature increased, the Al_2O_3 phase was not found. This is because Al ions (RAI = 0.054 nm) replaced Zn ions (RZn = 0.072 nm) in the hexagonal lattice, and because Al ions occurred in the interstitial sites of ZnO or Al segregates migrated to the grain boundaries, thus forming Al-O bonds [24].

Generally, prepared AZO thin films are deposited by various methods, including RF-magnetron sputtering, pulsed laser deposition (PLD), MOCVD, spray pyrolysis, etc. In the preferred orientation of AZO thin films deposited by such conventional methods, the (002) plane in the Wurtzite crystal structure grows mainly in the c-axis direction parallel to the substrate [25]. As a result of detailed analysis of the crystal structure of the AZO thin films, it can be seen that as the post-growth annealing temperature increased, the position of the peak shifted toward lower 2θ. This XRD shift is associated with the grain growth phenomenon and the stress present in the thin films [26].

Looking at the XRD results, in the case of samples annealed up to 400°C, compressive stress is dominant on the surface of the thin film and increases. Then, when annealing is performed at a temperature of 400°C or higher, the compressive stress shows a tendency to decrease. This is believed to be because when high-temperature annealing is performed in a high-purity oxygen atmosphere, oxygen vacancies present previously in AZO thin films are filled by the effect of the gas atmosphere, thus causing lattice rearrangement, and for this reason, the compressive stress is reduced [27].

This belief is associated with the grain boundary relaxation model. Generally, when post-growth annealing is performed, grains grow and, at the same time, tensile stress increases due to the interaction between the grains. Due to this tensile stress, compressive stress is offset, and as a result, the compressive stress weakly acts. Regarding this result, the results of a study conducted by J. A Thornton et al. assert that as the T/Tm (T: temperature of the substrate, Tm: meting point of the film) value increases, recovery, recrystallization and grain growth occur. This supports the experimental results of the present study [28].

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Among the samples subjected to post-growth annealing in an oxygen atmosphere, the sample annealed at 600°C showed the highest crystallinity. Looking at the XRD results, the (002) diffraction peak of AZO increased and showed the highest value at 600°C. This provides a parameter for improving crystallinity, with the result that the crystalline peak in the C-axis direction increases as the annealing temperature increases. This suggests that the lattice constant “a” is almost constant without linearly decreasing depending on the post-growth annealing temperature. This fact implies that as the temperature increases, Al does not replace Zn sites, but the crystallinity of the AZO film is improved by other factors [24]. This suggests that the crystallinity is increased due to a relative increase in the Zn or Al concentration depending on the post-growth annealing temperature.

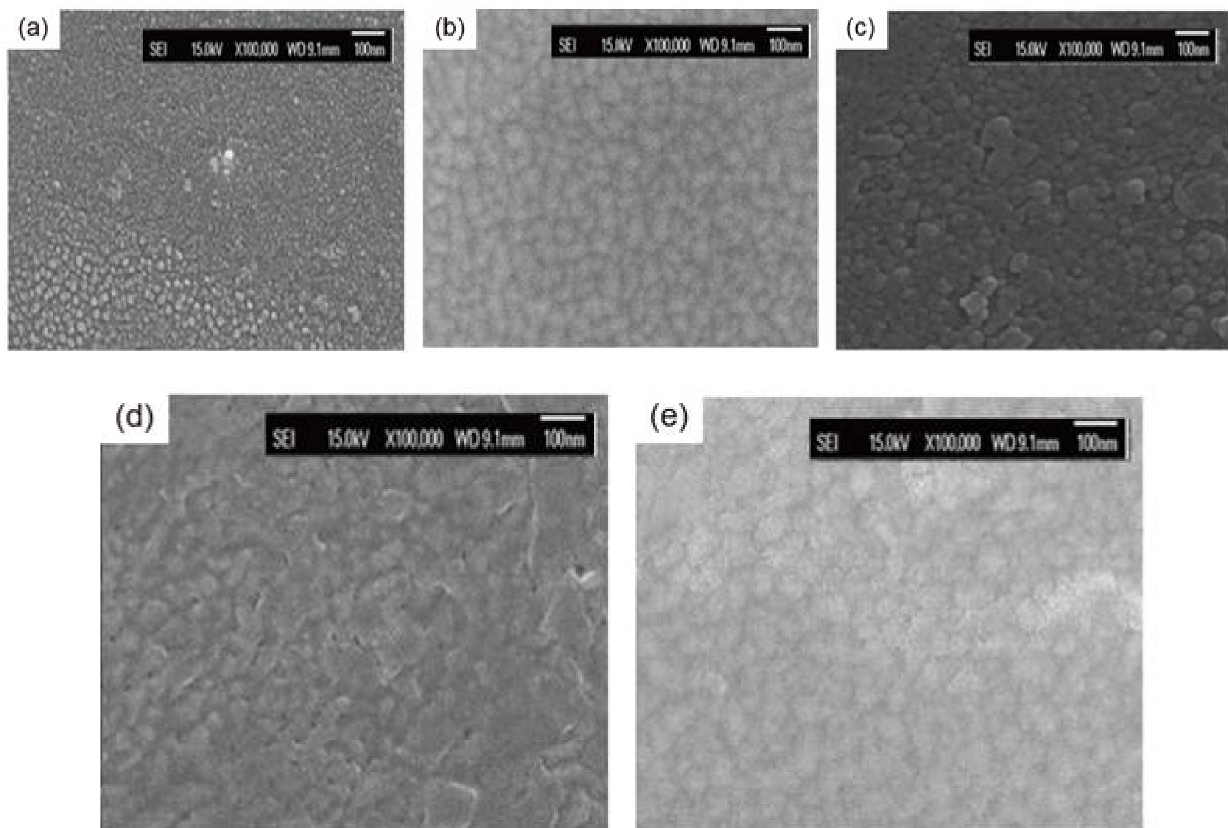

Fig. 4. FE-SEM image of AZO thin films grown at room temperature and different annealed with rapid treatment (a) AZO thin film by Ion beam deposition, (b) 400°C, (c) 500°C, (d) 600°C, (e) 700°C.

However, as the post-growth annealing temperature increased, the intensity of the peak relatively decreased. This indicates that due to oxygen adsorption or increased defects, the growth of crystalline grains decreases or recrystallization occurs. This study result is also observed in the measurement results of PL. To support the change in crystallinity of AZO thin films with this change in post-growth annealing treatment, the full width at half maximum (FWHM) was observed. In the case of a thin film irradiated with an ion beam of 100 eV, the FWHM was relatively high, suggesting that a plurality of defects was present in the AZO thin film. In order to overcome this problem, annealing was performed in a high-purity oxygen atmosphere at a temperature ranging from 400°C to 700°C. As the annealing temperature increased, the FWHM decreased. The FWHM was the lowest at 600°C, and decreased at higher temperature.The lowest FWHM value implies that the crystalline orientation of the AZO thin film is influenced by the lowest surface free energy and that crystal growth becomes better. Fujimura et al. asserted that particles with low surface energy become larger than the growth of the thin film and that the orientation of the thin film develops to a

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single crystallographic orientation with the lowest surface energy. This supports the results of the present study [29]. This suggests that the AZO thin film was preferentially oriented in the C-axis direction and well grew. Thus, as post-growth annealing is performed, many invasive defects of Zn and vacancy defects of O_2 disappear due to the effect of high-temperature oxygen adsorption and the annealing effect, and stoichiometric 1:1 combination of Zn and O_2 increases. For this reason, as recrystallization of defects on the ZnO thin film occurs, composition particles move and the ratio of micro crystal gets higher. This is attributable to the aggregation of adatoms caused by thermal energy that increased with increasing annealing temperature. These results are consistent with the results of FE-SEM, thus supporting reliability. However, as the annealing temperature increased to 700°C or higher, the FWHM intensity showed a tendency to increase. This suggests that as the annealing temperature increases, the increase of crystallinity in the C-axis direction decreases. Although the exact cause can be unknown, it is believed that as the annealing temperature in the oxygen atmosphere increased, the release of oxygen atoms from the surface of the AZO thin film increased, and as excess oxygen penetrated the AZO thin film, the crystallinity was deteriorated due to oxygen adsorption on the surface. At the same time, it is believed that as oxygen vacancies in the thin film behave like oxide s or metals, the crystallinity was deteriorated.

This indicates that due to a high temperature of 700°C or higher, Zn and O are released or Zn gets damaged, resulting in deterioration in the crystallinity. This supports the foregoing assertion, because the (002) diffraction peak decreased and then the FWHM value increased [30].

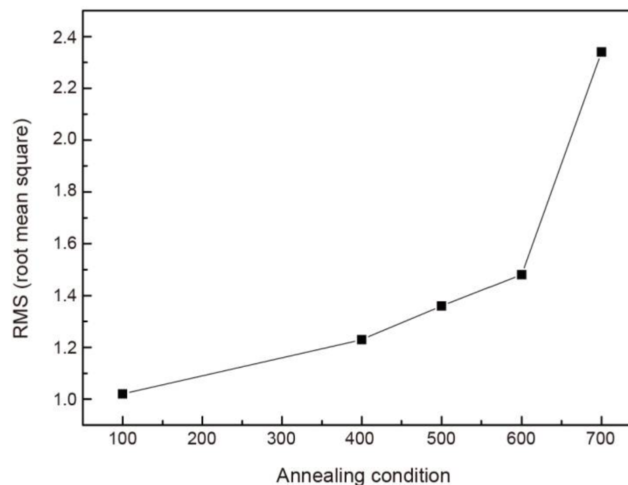

Fig. 5. The values of the surface roughness of the conventionally evaporated Al-doped ZnO film in oxygen ion environment and the Al-doped ZnO films deposited at 100 eV, and annealed at 400 °C, 500 °C, 600 °C, 700 °C.

3.2. Microstructural Analysis under Varying Annealing Conditions

Fig. 4 shows SEM images of the surfaces of AZO thin films annealed at different temperatures under a high-purity oxygen atmosphere. Figure 4 (a) shows a sample irradiated with an ion beam energy (100 eV), and Figs. 4 (b), 4(c), 4(d) and 4(f) show samples annealed in an oxygen atmosphere. In the case of the thin film irradiated with an ion beam of 100 eV, the particle size showed a tendency to increase due to the increase in atom mobility caused by ion beam energy irradiation, but the shape of a polycrystalline thin film was observed in the XRD analysis results due to increase in the surface roughness and an increase in defects. However, in the case of the samples annealed at 400°C 500°C, 600°C and 700°C, it can be seen that as the annealing temperature increased, the surface shape had an island structure, and it gradually distinctly grew. As the post-growth annealing temperature increased, the growth of adjacent grains and recrystallized grains in the AZO thin films occurred. In the case of AZO thin films, thermal energy increases with increasing annealing temperature, and for this reason, the activation energy of Al, Zn and O atoms increases and actively moves in the crystals of the AZO thin films. Movement of the activated energy causes a gradual increase in the cluster size on the surface. In the experimental results, in the case of the AZO thin film annealed at 600°C, Al, Zn

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and oxygen atoms moved to a position with stable surface energy in the AZO thin film due to an increase in the activation energy. For this reason, grain growth occurred, vacancies were almost filled, and the atoms on the substrate were sufficiently diffused, and thus small grains showed a tendency to overlap and combine. It is believed that this thin film with a large grain size will show excellent electrical and optical characteristics, because the grain area, the grain boundaries and the defect structures present therein are smaller than those of thin films having fine structures [31]. However, in the sample annealed at 700°C or higher, it appears that the growth of recrystallized grains occurred due to annealing at the high temperature. In addition, the shape of microstructures did differ from that of other grains. As the density of crystalline grains increased, some spherical and hexagonal crystalline grains were formed, indicating that precipitation of some grains on the surface occurred. For this reason, it is expected that the surface roughness would be increased. For this reason, it is expected that the crystallinity of the thin film would be reduced and the optical and electrical properties thereof would be adversely affected [32].

3.3. Surface Morphology at Varying Annealing Temperatures

In order to examine the surface roughness of AZO thin films subjected to various pretreatment processes, the surface roughness of the thin films was measured using an AFM system. At this time, the RMS value, which is a measure of the surface roughness, was measured. In this experiment, for application to transparent electrodes and light-emitting materials, an AZO thin film irradiated with an oxygen ion beam (100 eV), and thin film samples obtained by annealing the AZO thin film sample at temperatures of 400°C, 500°C, 600°C and 700°C in an oxygen atmosphere were measured for their roughness (rms). Generally, for application to a transparent conductive substrate and a light-emitting substrate for a display device, it is important to determine the optimum process conditions for surface roughness. For a transparent electrode which serves as a pixel electrode for an organic EL device and is used for a hole injection electrode, an organic material is deposited directly on a transparent electrode layer, and thus the surface characteristics of the transparent electrode have a direct effect on the efficiency of hole transport to the organic layer. This is because when the height or step difference is great due to a rough surface, the possibility increases that the current is concentrated on the protruding portion, causing dark spots in the organic EL device [33]. In addition, when the surface roughness is poor, diffuse reflection during exposure can be severe, thus causing a problem in etching. In addition, light incident into the thin film can be scattered, thus adversely affecting the light transmittance. Such complex action can adversely affect the display device [34].

In addition, since an UV light-emitting device is fabricated through p-n junction, the surface roughness is an important factor for the light-emitting device. This is because the surface roughness of a thin film can cause carrier scattering which may affect the electrical conductivity and light emission characteristics of the thin film [35]. The results of the annealing experiment performed in a high-purity oxygen atmosphere indicated that the surface roughness value of samples annealed at a temperature of 400°C to 600°C was 2 nm or less, which was applicable to a transparent electrode for a display device.

In the experimental results, it could be observed that the AZO thin film irradiated with an oxygen ion beam (100 eV) showed a relatively smooth surface roughness of 1.02 nm, whereas the Rms values of the samples annealed at 400°C, 500°C, 600°C and 700°C in an oxygen atmosphere increased to 1.23 nm, 1.36 nm, 1.48 nm and 2.34 nm, respectively. From such study results, it is believed that suggest that that as the annealing temperature increases, the density of the AZO thin film increase, and the energy of particles adsorbed onto the substrate increases, thus further increasing the surface roughness of the thin film. Generally, microstructural features caused by annealing may be summarized as crystallization and grain growth, an increase or decrease in defects, a change in surface roughness, etc. Annealing conditions affect grain boundaries and surface roughness, which then affect the light transmittance and electrical conductivity of the thin film.

This reason is consistent with the Matthiessen's rule on electrical conductivity in that resistance varies depending on thermal vibration, impurities, and defects. A film crystallized on a smoother surface makes less carrier scattering and higher mobility, thus improving conductivity. Therefore, the regulation of the dense particles and Rms is considered important in the manufacture of touch panels, OLED devices, solar cell transparent electrodes, or light-emitting devices.

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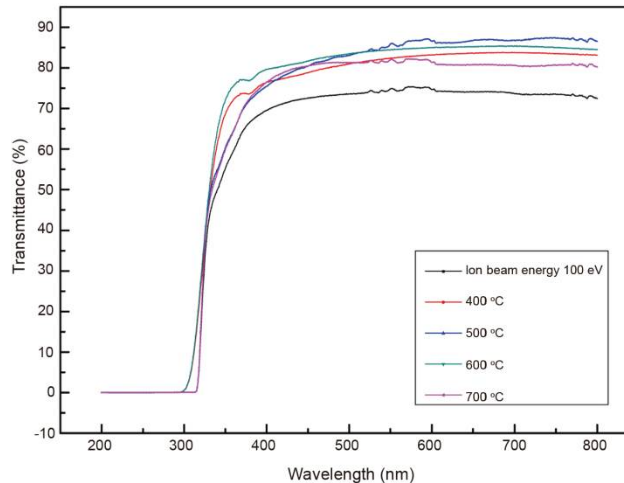

Fig. 6. Optical Transmittance at 550 nm with respect of glass substrate with changing of the Annealing treatment at Various Temperature in O_2 ambient gas.

3.1. Optical Properties at Various Annealing Temperatures

Figure 6 shows the results of measuring the transmittances of AZO thin films, subjected to a post-growth annealing process in a high-purity oxygen atmosphere, by an UV-spectroscopy meter at a wavelength region of 200 to 800 nm. At this time, the transmittances were measured using 550 nm as a reference. In the experimental results, the thin film irradiated with an ion beam (100 eV) showed a transmittance of 80% or lower. This is believed to be because spot defects, such as oxygen vacancies, Zn interstitials and Al solute atoms, were increased by ion beam bombardment. In the case of the samples annealed in a high-purity (99.99%) oxygen atmosphere, all the samples showed a transmittance of 80% or higher in the visible region (λ = 550 nm), and the transmittance showed a tendency to increase with increasing annealing temperature. In the case of the samples annealed at 400°C and 500°C, the penetration of oxygen atoms due to oxygen adsorption or low diffusibility of Al and Zn was not enough to bond with Zn atoms or fill oxygen vacancies. However, in the case of the sample annealed at 600°C, activation energy enough to allow oxygen ions to penetrate into the ZnO thin film was delivered, and thus high-purity oxygen could be sufficiently adsorbed onto the thin film surface. This is because the grains themselves constituting the AZO thin film had many oxygen deficiency. As explained above, when annealing in the oxygen atmosphere is performed, the surface is activated by annealing, so that the oxygen vacancies, Zn interstitials and Zn vacancies of the thin film are reduced more quickly, thus reducing the oxygen vacancies, and at the same time, the structure of the stoichiometric composition changes. It can be understood that the increase in transmittance caused by annealing at 600°C or higher is attributable to an increase in the grain size and a decrease in the grain boundary density [36]. At the same time, since dense and large thin-film grains are grown, light scattering decreases, and thus the thin film can show excellent optical properties [36, 37]. At the same time, in the AZO thin film, blue shifting toward a shorter wavelength can be observed, and this phenomenon is considered caused by interference of the incident beam reflected from the AZO thin film and the substrate.

Such results are also consistent with the above-described SEM and XRD results. As the surface roughness of the AZO thin film decreases, factors that cause light scattering during the passage of light through the AZO thin film decrease, resulting in an increase in the transmittance. In addition, it is believed that as the C-axis crystalline orientation of the AZO film is improved, light scattering of the crystal structure decreases, resulting in an increase in the transmittance. In the experimental results, the sample annealed at 600°C showed the highest transmittance of about 88%. However, annealing at 700°C or higher showed reduced transmittance. Although there may be various causes, the light transmittance of the AZO thin film decreases due to increased surface roughness, quick and excessive oxygen adsorption of the thin film at high temperature, and precipitation of recrystallized particles on the AZO thin film.

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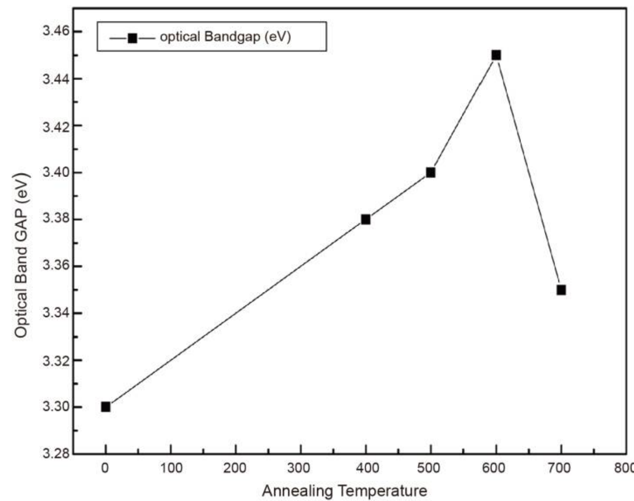

Fig.7. Variation of the optical band gap as carrier concentration change.

Figure 7 shows the correlation between the (αhν)1/2 and hν of the AZO thin film as a function of post-growth annealing temperature and the resulting change in the optical band gap. According to Tauc, the semiconductor optical band gap in a strong absorption region can be represented by the following equation (1):

α□(hν-Eg)1/2 (1)

wherein hν denotes photon energy, Eg denotes optical band gap (or Tauc gap), and n denotes a constant value. In addition, α is the absorption coefficient of thin film, and can be calculated from the following relation equation: α = (1 / t)in(1/T) (T is transmittance, and t is thin-film thickness). It can be seen that as the annealing temperature increases, the optical band gap of the AZO thin film changes. When the AZO thin film was irradiated with an ion beam (100 eV), the band structure of the AZO thin film had band tails due to many defect states present around band edges, unlike crystalline band structures. Depending on these defect states, the optical band gap greatly changes, and process conditions during thin film formation are greatly influenced [38].

The AZO thin film was annealed at 400°C to 600°C, the optical band gap increased, and when the thin film sample was annealed at 700°C, the optical band gap decreased. This change in the optical band gap may be explained by the Burstein-Moss shift. According to the Burstein-Moss shift theory, the optical energy gap increases as the free electron concentration increases. 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 [39]. At the same time, it is believed that depending on the oxygen atmosphere and the annealing time, an increase in the Al impurity concentration influences. Then, when the annealing process was performed at 700°C or higher, the release of oxygen from the AZO thin film increased, and the oxygen was filled in the oxygen vacancies in the thin film, resulting in an increase in the density of the Al-doped ZnO thin film, and optically behaved a metal, thus reducing the optical band gap.

When external energy is applied to the light-emitting material of the Al-doped ZnO thin film at varying annealing temperatures, electron-hole pairs (EHPs) are produced. The produced EHPs are unstable, and thus disappear through a recombination process, and the material returns to the original equilibrium state. Light emission refers to phenomena that emit light corresponding to the energy lost in the recombination process. Specifically, it refers to a series of physical phenomena that when external energy is applied to atom, molecule, polymer and crystal, the energy into the material by transition between the inherent electronic states is emitted as light and the material returns to its original equilibrium state. Namely, the light emission phenomena are classified by external energy that excites electrons. PL is a type of light emission by monochromatic proton energy such as a laser. Based on the PL results, the characteristics of AZO thin films at deposition parameters can be observed, and the stoichiometric characteristics thereof can be indirectly determined, and based on the light emission characteristics thereof, the applicability thereof to an optical device can be determined.

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AZO thin films are transparent thin films having a direct band gap of 3.37 eV, but in most cases, these films are non-stoichiometrically deposited due to an excessive content of zinc or lack of oxygen in a deposition process [40]. At this time, two electrons remaining after removing 6 electrons from 8 valance electrons around oxygen vacancies in defect structures of ZnO form a double donor [41].

Accordingly, an AZO thin film is an oxygen-deficient oxide in which defects occur, and thus it shows n-type characteristics. Ultimately, donor level-to-acceptor level transition or deep level transition from the longitudinal optical (LO) phonon energy level, as well as the band-to-band recombination that causes UV light emission, causes light emission in various visible regions having wavelengths longer than UV wavelengths [42]. Generally, ZnO thin films have an UV emission peak at around 380 nm, a green emission peak at around 510 nm, a yellow emission peak at around 590 nm, and a red emission peak at around 650 nm [43-45]. Generally, for UV emission from AZO thin films, it is important to reduce defects in deposited thin films, and to this end, an epitaxial thin film process is necessary. Thus, in this experiment, in order to form high-quality thin films, thin films were deposited by the MBE method, and post-growth annealing in a high-purity oxygen atmosphere was performed to reduce defects.

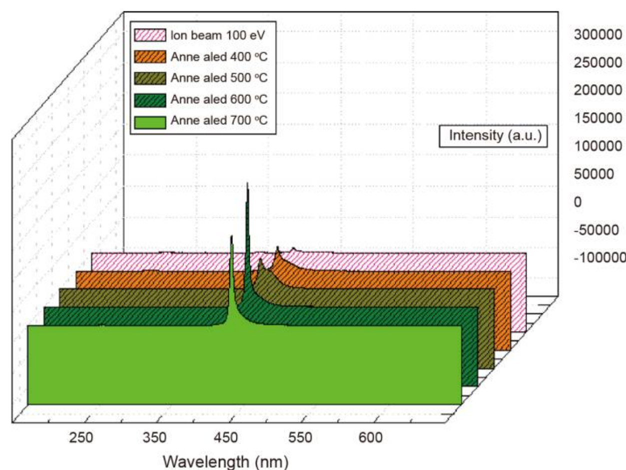

Fig. 8. The PL spectra of Al-doped ZnO thin films as annealing temperature.

Figure 8 shows the emission spectra of an AZO thin film deposited by ion-beam-assisted molecular beam epitaxy and AZO thin films annealed at different temperatures (from 400°C to 700°C). Generally, the quality of optical characteristics of AZO thin films can be determined by measuring the NBE emission ratio on PL data. In the present study, an UV emission peak at around 380 nm mainly appeared. According to the literature, in the case of UV emission that occurs in AZO thin films, free exciton transition (transition of free excitons in the conduction band to the valence band) and the transition of bound excitons with a shallow donor level, which is lower than that of the conduction band by several tens meV, to the valence band, result from intrinsic defect levels of ZnO including zinc interstitials (Zni), zinc vacancies (VZn), oxygen vacancies (VO), oxygen interstitials (Oi), antisite oxygen (OZn) and the like [46]. In this case, it has been reported that neutral or ionized impurity atoms or host lattice defects act as shallow donors or undergo transition to Zn vacancies as shallow acceptors, thus causing blue emission with 2.9 eV and 2.6 eV, respectively [47-48].

Some literatures reported that emission in the blue region results from electron migration between zinc interstitials and zinc vacancies, thus supporting NBE emission [49]. In addition, phonon replicas of free exciton transition and bound exciton transition also appear in UV emission. It is known that emission in the green region results from the transition of excitons with a deep donor level to the valence band. Many scientists have conducted many studies on deep donor levels, but these deep donor levels have not been clearly identified [50].

Studenikin et al. asserted that an oxygen vacancy (V O) as a donor and a Zn vacancy (V Zn) having one hole as an acceptor result in green emission by electron-hole combination [51]. In addition, Lin et al. asserted that green emission occurs depending on annealing conditions [52]. The experiments conducted by them indicated that when ZnO was grown on a p-Si substrate by a DC reactive sputtering method and annealed at different temperatures in different gas

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atmospheres, the intensity of green emission was stronger as the temperature and oxygen partial pressure were higher, and this is because of antisite oxygen which is a deep donor [52]. Many literatures assert that the cause of green emission is usually associated with the binding of oxygen. However, looking at the results of studies on the non-stoichiometric composition of ZnO and studies on green emission, it is considered that a deep donor that causes green emission in ZnO may also be a non-vacancy.

It is believed that if this non-stoichiometric composition of ZnO can be controlled, the green emission of ZnO can also be controlled. However, it is thought that the causes of green emission need to be considered in depth. However, it is certainly believed that it is necessary to improve the function of a UV light emitting device by decreasing the green emission in the visible region in order to obtain a high-purity AZO thin film having the luminescence characteristics. In this experiment, the results of PL analysis indicated that the thin film irradiated with an oxygen ion beam (100 eV) showed low AZO thin-film peak intensity. This suggests that a number of defect peaks are present in the thin film. Generally, zinc oxide thin films grown by a PVD process mostly do not have a 1:1 zinc-oxygen (Zn-O-Zn-O) structure, and these thin films contain a number of oxygen vacancies. These oxygen vacancies act as n-type dopants, but deteriorate optical characteristics by forming impurity levels in the band gap. In this experiment, annealing was performed at each of 400°C, 500°C, 600°C and 700°C under a high-purity oxygen atmosphere in order to reduce these oxygen vacancies and increase the applicability of thin films to UV light-emitting devices. It is generally known that highly crystalline thin films having less defects and impurities show stronger NBE peaks. It can be seen that as the annealing temperature, exciton peaks in the AZO thin films post-annealed at high temperatures relatively increase. This is believed to be because as the annealing process temperature increases, activation energy allowing oxygen gas to penetrate into the thin film by the influence of the oxygen atmosphere gas is sufficiently supplied to the thin film structure that has many defects before annealing, and oxygen vacancies (VO), zinc vacancies (VZn), zinc interstitials (ZnI), oxygen interstitials (OI) disappear a lot while a stoichiometric 1:1 bond between Al-Zn and oxygen increases a lot [53-54]. The results of the annealing indicated that the NBE peak of the AZO thin film annealed at 600°C showed a stronger intensity than that of the AZO thin films annealed at other temperatures. This suggests that a highly crystalline thin film having less defects and impurities was formed and showed strong NBE emission. This is believed to be because the composition of the AZO thin film was controlled by controlling the oxygen gas atmosphere in the post-deposition annealing process. In fact, a study conducted by Shi indicated that emission in the NBE region was improved by controlling the annealing atmosphere to reduce the green emission of ZnO thin films, thus supporting the results of the present study [55].

In this experiment, it is believed that annealing at 600°C is the optimum condition that prevents the introduction of impurities and eliminates other various interference factors, and is also a thin-film growth condition that can provide only high-quality green and UV emission. It is believed that the AZO thin film annealed at a temperature of 600°C has the nearest stoichiometric composition and that the activation energy of Al, Zn and O atoms in the AZO thin film increases and contributes to the surface energy of AZO crystals.

It is thought that through either the change in crystalline structures by annealing in the oxygen atmosphere or activation of the thin film surface by annealing without influencing residual stress, defects are minimized and the thin film density is increased while the crystallinity is improved, thereby improving the PL characteristics.

However, in the case of the sample annealed at 700°C, NBE emission decreased. This is believed to be because of the influence of grain precipitation and recrystallization. From PL measurement, it is expected that in the case of the sample annealed at 700°C, focus would be made on small AZO grains that precipitated upon irradiation with an He-Cd laser, and on recrystallized grain boundaries, and that charge trapping would occur a lot due to the small grains. It is believed that due to these effects, weak NBE emission in the exciton region of ZnO thin films appears. The reason why the UV emission intensity decreased is believed to be because of non-stoichiometry caused by defects and formation of isolated excitons and many oxygen vacancies due to a large lattice constant as the post-growth annealing temperature increased [56].

These results are consistent with the XRD and SEM results and suggest that recrystallized grains and grain boundaries influence NBE emission.

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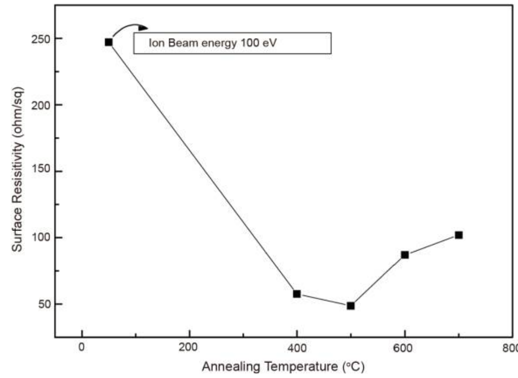

Fig. 9. Change of the resistance of the ZnO films with changing Annealing heat treatment temperature (°C) In O_2 gas ambient.

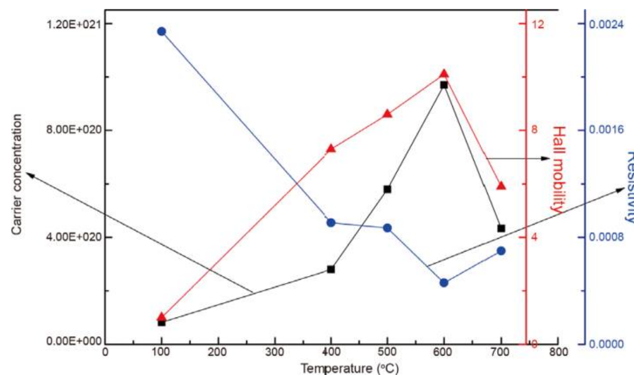

Fig. 10. Change of the Mobility and the Carrier concentration, and Resistivity of the ZnO: Al films with changing heat treatment Temperature in O2 gas.

Figures 9 and 10 show the results obtained by measuring the electrical properties of thin films after annealing. The experimental results indicate that the AZO thin film irradiated with the ion beam (100 eV) showed a high resistivity value or had unstable electrical properties, and thus needs to be improved for use as a transparent electrode or light-emitting material. However, in the case of samples obtained by annealing the AZO film in the oxygen atmosphere while increasing the annealing temperature from 400°C to 700°C by 100°C, the carrier concentration increased with increasing temperature, and the resistivity showed to a tendency to decrease with increasing temperature. In particular, the AZO sample annealed at 600°C showed the highest carrier concentration value, the lowest resistivity value and the highest mobility value.

This is believed to be because atoms were sufficiently diffused by providing energy necessary for crystallization through the annealing process, and the doped Al atoms were all activated by transition from interstitial sites to substitutional sites , and at the same time, lattice motion actively occurred, and thus a uniform and stable thin-film surface was formed and the concentration of electrons on the surface increased [52]. However, in the case of the sample annealed at 700°C or higher, the mobility and carrier concentration values showed a tendency to decrease, and the resistivity showed a tendency to increase.

Generally, AZO thin films have oxygen vacancies in which a donor produces two electrons. As the number of these oxygen vacancies increases, the carrier concentration increases and the resistivity decreases. However, in the case of the AZO thin film annealed at 700°C in the oxygen atmosphere, it is believed that due to the O_2 atmosphere gas and the increase in the annealing temperature, the oxygen vacancies in the thin film were replaced with oxygen gas, the tendency of Al atoms into Zn sites increased, and thus the carrier concentration decreased, leading to an increase in the

resistivity. At the same time, it is believed that since the annealing process suppressed transposition occurring in the AZO thin film, the effect of reducing the effective concentration of electrons is dominant, and thus scattering of ionized impurities and factors that increase the temperature cause partial evaporation of Zn, thus reducing the crystallinity of the AZO thin film and increasing defects, thereby relatively increasing the electrical resistivity.

In addition, in the case of the AZO thin film annealed at 700°C or higher, the different lattice constants between the substrate and the AZO thin film affect the atom spacing. This difference in the atom spacing causes stress in the thin film [54]. Thus, in the case of the AZO thin film deposited on the glass substrate, the interface between the glass and the AZO thin film will receive stress caused by the difference in the lattice constant, and for this reason, there is a high possibility that the lattice is split or a grain boundary is formed. Accordingly, in this experiment, it is believed that when annealing is performed at 700°C or higher in an oxygen atmosphere, the interfacial stress affects and reduces the crystallinity of the thin film throughout the thin film. This belief is supported by existing studies on substrate/ZnO thin film stress models [57, 58].

IV. CONCLUSION

n-type Al (2 wt%)-doped ZnO (AZO) thin films were deposited by ion-beam-assisted molecular beam epitaxy, and the applicability of the AZO thin films to light-emitting devices was examined. An Al-doped ZnO thin film was deposited on glass and annealed at each of 400°C, 500°C, 600°C and 700°C for 30 minutes in a high-purity (99.99%) atmosphere, and the electrical, optical and structural characteristics of the AZO thin films were observed. As the post-deposition annealing temperature increased, the resistivity decreased and the light emission characteristics increased gradually. In particular, the thin film annealed at 600°C for 30 minutes showed a visible transmittance of 85% or higher in the transmittance experiment and showed an good resistivity of 4.6 x 10 Ω.cm in the electrical experiment. This thin film exhibited the best blue emission in the visible region, indicating that it can be applied to electronic devices, including LED devices and organic EL devices.

However, as the annealing temperature increased to 700°C, the resistivity increased again and the light emission characteristics decreased. This is believed to be because the post-deposition annealing at this high temperature reduced grain boundaries and caused grain recrystallization, resulting in charge trapping that affects the electrical and optical characteristics of the thin film.

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