Influence of Surface Texturing on the Electrical and Optical Properties of Aluminum Doped Zinc Oxide Thin Films

Jaeh-Yeong Lee, Joong-Pyo Shim, and Hak-Kee Jung, *Member, KIMICS*

Abstract— **An aluminum doped zinc oxide (AZO) film for front contacts of thin film solar cells, in this work, were deposited by r.f. magnetron sputtering, and then etched in diluted hydrochloric acid solution for different times. Effects of surface texturing on the electro-optical properties of AZO films were investigated. Also, to clarify the light trapping of textured AZO film, amorphous silicon thin film solar cells were fabricated on the textured AZO/glass substrate and the performance of solar cells were studied. After texturing, the spectral haze at the visible range of 400 ~750 nm increased substantially with the etching time, without a change in the resistivity. The conversion efficiency of amorphous Si solar cells with textured AZO film as a front electrode was improved by the increase of short-circuit current density (J_{sc}), compared to cell with flat AZO films.**

Index Terms— **Aluminum doped zinc oxide (AZO) film, surface texturing, optical properties, electrical properties, thin film solar cell.**

I. INTRODUCTION

THE current density of thin films solar cells can be substantially increased by enhancing the absorption of the incident light in the absorption layer [1]. In the ideal case, the scattered light is confined within the multilayered thin film solar cell and almost completely absorbed after multiple passes through the photocurrent generating layer. In particular, silicon based solar cells rely on an effective light trapping either for stability reasons (amorphous silicon) or because of a low absorption coefficient (thin crystalline or microcrystalline silicon) [2]. Currently it becomes a standard approach to increase optical absorption in the active layer of solar cell by the light trapping and the reduction of reflection losses at the front surface. Especially, an adequate surface texturing is necessary to provide an efficient light scattering and subsequent light trapping inside the absorber material of the solar cell [3]. Doped zinc oxide (ZnO) films have attracted much interest in recent years as a transparent conductive oxide (TCO) material because of their good electro-optical properties in combination with large band gap, low cost than indium tin oxide (ITO), absence of toxicity, and high stability in hydrogen plasma [4-8]. Surface texturing of ZnO films is achieved by formation of rough surface, by means of surface etching [9]. Kluth et al. [2] reported a etching technique to smooth r.f. magnetron sputtered ZnO:Al films and described different surface textures developed upon etching in weak hydrochloric acid (HCl). However, the electrical and optical properties of the films are critically dependent on their microstructure, texture and surface morphology, on the presence of defects and the structure of the film–substrate interface [10].

In this paper, we investigated the influence of surface texturing on the electrical and optical properties of aluminum doped zinc oxide (AZO) films. AZO films were prepared by r.f. magnetron sputtering and subsequently textured by a wet-chemical etching in diluted hydrochloric acid process. In addition, amorphous silicon thin film solar cells were fabricated on the textured AZO/glass substrate and effect of surface texturing on the performance of solar cells were studied.

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II. EXPERIMENTAL DETAILS

The AZO films were deposited by a conventional r.f. magnetron sputtering system. Corning 1737 glass, in this work, was used as a substrate. The glass substrates were ultra-sonically cleaned in acetone and methyl alcohol, rinsed in deionized water, and subsequently dried in flowing nitrogen gas before deposition. A sintered ceramic target with a mixture of ZnO (99.99% purity) and Al_2O_3 (99.999 purity) was employed as source materials. The amount of Al_2O_3 added to the target was 2.5 wt.%. The diameter of the target was 4 inch and the target-to-substrate distance was kept at 7 cm. The substrate was loaded in the load-lock chamber and then carried into the main chamber. A turbo-molecular pump coupled with a rotary pump was used to achieve the ultimate background pressure of 9.33×10^{-5} Pa before introducing argon gas. The pre-sputtering was applied for 15 min to clean the target surface and remove any possible contamination. During deposition process, the r.f. power was maintained at 170 Watt and the frequency is 13.56 MHz. The pressure of Ar was controlled at 2.7 Pa. The substrate temperature was measured by using a K-type chromal-alumal thermocouple and the substrate was kept at room temperature. The AZO films with various thicknesses were obtained by varying the deposition time. Surface textured AZO films were created by anisotropically wet etching in diluted hydrochloric acid (0.5 wt% HCl (50%) in H_2O). The etching time was varied from 10 to 80 second.

To demonstrate superior optical properties of textured AZO film, amorphous Si thin film solar cells with p-i-n structure were fabricated by plasma enhanced chemical vapor deposition (PECVD). The solar cell structure was glass/AZO/p-type a-Si:H/intrinsic a-Si:H/n-type a-Si/Ag.

The thickness of as-deposited AZO films was measured by using a surface profiler (α-step, Tencor Co.) and ranged between 1 μm and 1.4 μm, depending on etching time. A structural change of AZO films was evaluated by X-ray diffraction (XRD) measurements in the 2θ mode. The surface morphology of the films was studied by field effect scanning electron microscope (FE-SEM). The crystal structure and preferred orientation was investigated by x-ray dffracttion (XRD). The electrical properties, such as a resistivity, carrier concentration and Hall mobility, were obtained from Hall-effect measurement by the Van der Pauw technique. Optical transmittance and reflectance, both total and diffuse, was recorded at the range of 300~1100 nm by using a UV-Vis spectrophotometer (Sinco, U3100) with an integrating sphere. The haze was calculated by the ratio of diffuse to total transmittance. The photovoltaic characteristics of amorphous Si thin film solar cells were measured under illumination of 100 mW/cm^2 intensity and AM1.5 spectrum.

III. RESULTS AND DISCUSSION

Fig. 1 shows the XRD patterns of AZO film etched in diluted HCl solution for different times. As-deposited AZO film have a strong peak near 2θ=34.42°, and two weak peaks at 62.88° and 72.52°, which is very close to the standard ZnO crystal (2θ=34.422°, 62.864°, 72.562°. These peaks are associated with (0 0 2), (1 0 3), and (0 0 4) planes of hexagonal phase, suggesting that the AZO film has the polycrystalline structure with preferred orientation of the c-axis perpendicular to the substrate. No Al or Al_2O_3 related phase was detected from the XRD pattern, suggesting that Al substitutionally replaces zinc in the hexagonal lattice or Al segregates into the non-crystalline region in the grain boundary [11]. As the etching time was longer, the intensity of peaks corresponding to (0 0 2) and (0 0 4) plane was highly increased. This means that the degree of c-axis orientation becomes more preferred by wet etching. When the etching time was longer over 70 second, however, the intensity of these peaks was reduced.

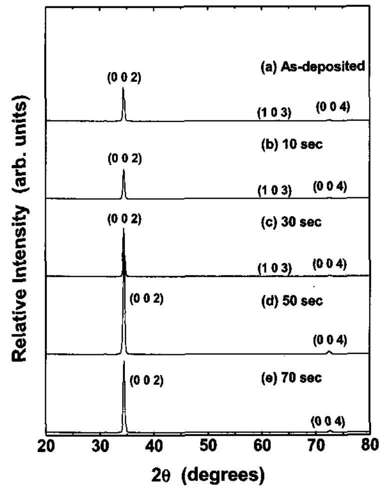

Fig. 1. XRD patterns of AZO films etched in diluted HCl solution for different times.

Fig.2 shows a surface micrographs for AZO films etched for various times. The morphology of as-deposited film is found to be smooth, continuous and dense. After anisotorical etching, the film exhibits a pyramid-like rough surface with small angle. This textured surface can be useful for effective light trapping in silicon thin film solar cells [12]. As the etching time surface is longer, a lunar-landscape like surface appearance develops, consisting of statically spread flat craters [13].

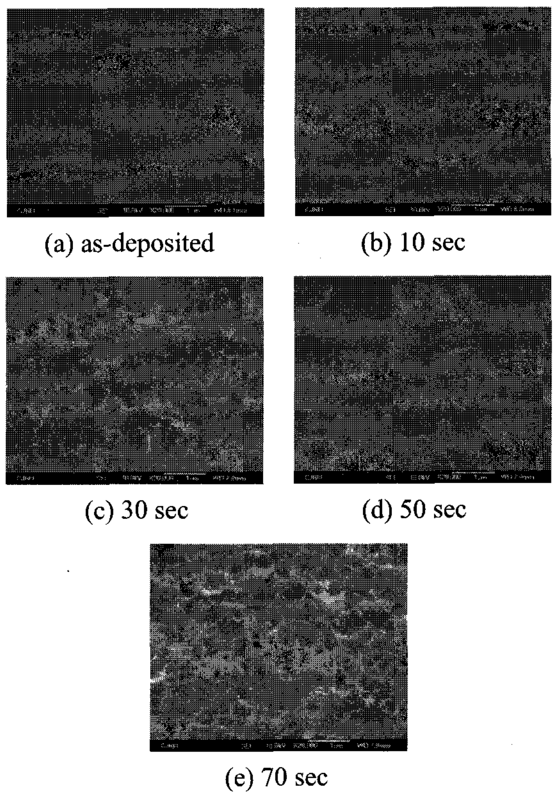

Fig. 2. SEM micrographs of AZO films etched in diluted HCl solution for different times.

The effect of surface texturing on light scattering properties can be observed by naked eye, changing transparent appearance to a milky one after etching in diluted HCl solution. Fig. 3 shows the total transmittance of the AZO films etched for different times in diluted HCl solution. The average transmittance in the range of 400~800 nm is inserted in the graph. The total transmittance and average transmittance at the visible region don't change, regardless of etching time, while the transmittance at the near infrared region was improved by texturing of AZO films. The optical loss by absorption by free charge carrier in near infrared region due to affects the current density of thin film solar cells, especially limiting the crucial long-wavelength response of amorphous silicon thin film solar cell [1]. Therefore, low free carrier absorption in the near-infrared region should be minimized by adequate surface texturing. The spectral haze serves as an indicator for the light scattering properties of the textured AZO films [2].

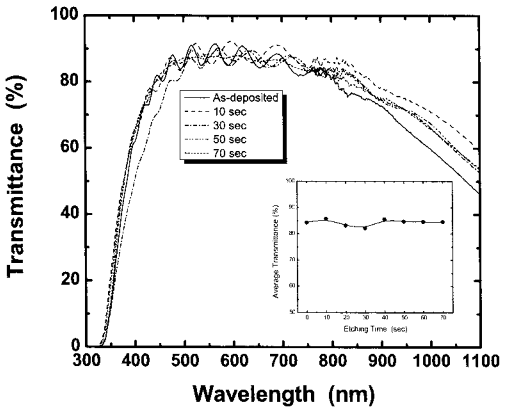

Fig. 3. Total transmittance of AZO films etched in diluted HCl solution for different times. An inserted graph is the average transmittance in the range of 400-800 nm.

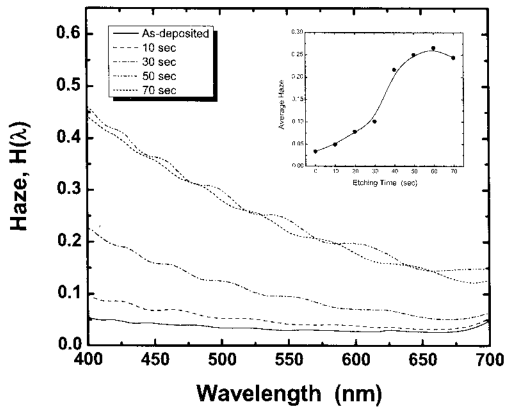

Fig. 4. Spectral haze of AZO films etched in diluted HCl solution for different times. The average haze in the range of 400-700 nm was inserted in the graph.

Fig.4 shows the spectral haze of the AZO films as a function of wet etching time. For as-deposited film, the haze is very low. The haze increases with the etching time, suggesting that the light scattering can be adjusted by etching time.

The electrical resitivity of as-deposited AZO films was 8.4×10^{-4} Ω-cm, and were little affected even after prolonged etching, as seen in Fig. 5. Thus, the etching process allows a separate optimization of the electro-optical and light scattering properties [2].

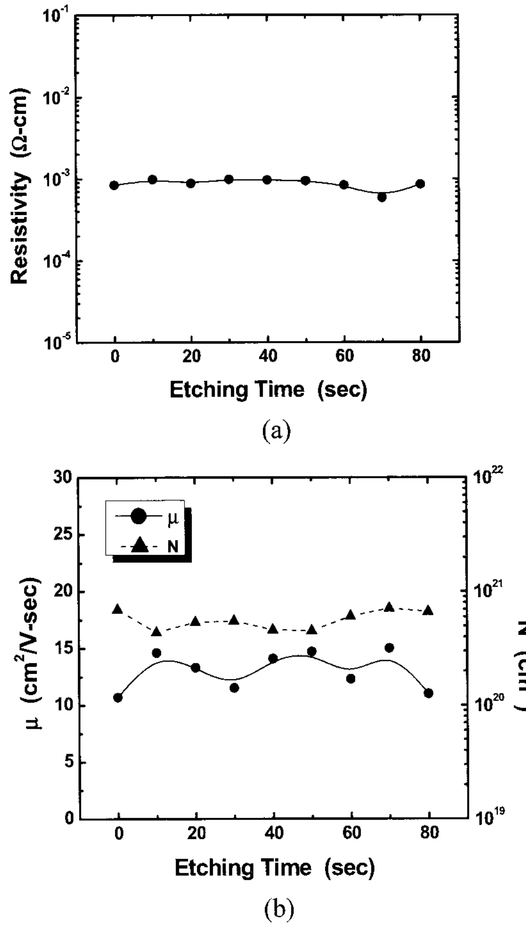

Fig. 5. Electrical properties of AZO films as a function of etching time: (a) resistivity, (b) Hall mobility (μ) and carrier concentration (N).

Fig. 6 shows the effect of texturing of AZO films on photovoltaic properties of amorphous silicon solar cell. The solar cell fabricated on textured AZO film gives larger short-circuit current density (J_{sc}) and also efficiency in comparison with a solar cell on as-deposited AZO film. This may be due to sufficient light trapping in textured AZO/glass substrate. Also, it is noted that the open circuit voltage (V_{oc}) deteriorated with increasing etching time. This is probably attributed to high recombination rate at the AZO/p-layer interface, leading to larger dark current density, as seen in Fig. 7 [12].

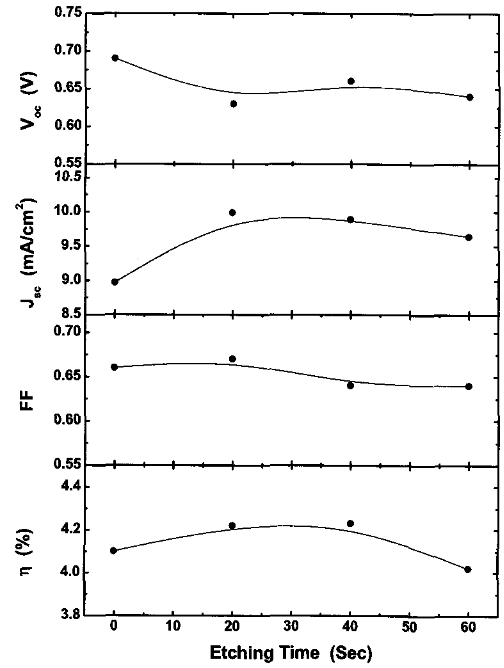

Fig. 6. Photovoltaic properties of amorphous silicon thin film solar cells fabricated on textured AZO films as a function of etching time.

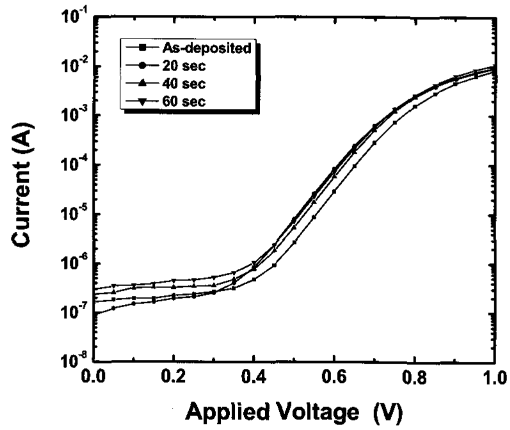

Fig. 7. Dark current-voltage characteristics of amorphous silicon thin film solar cells fabricated on textured AZO films as a function of etching time.

IV. CONCLUSIONS

The XRD patterns reveals that all of the obtained films are polycrystalline with the hexagonal wurtzite structure and have a preferred orientation with the c-axis perpendicular to the substrates, regardless of etching time. The location of the (0 0 2) peaks does not shift significantly with decreasing etching time, but the intensity become more intense and sharper. As the etching time in HCl solution was longer, the surface of AZO films became rougher and the diffused optical transmittance was increased, without a change in the resistivity. The enhancement of the optical properties of the films with increasing the etching time was associated with the surface structure of the films. The efficiency of amorphous silicon thin films solar cells was increased by texturing of AZO films. This improvement was attributed to increase of short-current of the solar cell.

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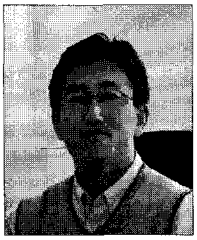

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