



## Dependence of aluminum-doped zinc oxide work function on surface cleaning method as studied by ultraviolet and X-ray photoelectron spectroscopies

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### ABSTRACT

The dependence of aluminum-doped zinc oxide (AZO) work function on surface treatment, i.e. acetone solvent cleaning and ultraviolet (UV)-ozone cleaning, was studied by ultraviolet photoelectron spectroscopy and X-ray photoelectron spectroscopy. Comparing different cleaning methods, UV-ozone treated AZO achieved relative higher work function of 4.26 eV; whereas acetone solvent treated AZO had relative lower work function of 3.94 eV. Two factors, stoichiometry ratio of  $[O_{ZnAl}]/([Zn] + 1.5[Al])$  and carbon contamination on AZO surface, affected the work function and, moreover, the stoichiometry ratio was supposed to be the controlling factor. It was concluded that AZO with high stoichiometry ratio and reduced carbon concentration possessed high work function.

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### 1. Introduction

Transparent conductive oxide (TCO) thin films are widely used in organic optoelectronic devices as anodes [1–4]. Indium tin oxide (ITO) films are the most popular TCO materials in organic light emitting diodes and organic photovoltaic devices due to their high conductivity, high transparency over the visible range, and large area homogeneity [5–7]. However, the indium is extremely rare and highly toxic and, moreover, it can diffuse into the organic layer, leading to the efficiency degradation [8,9]. Therefore, alternate new anode materials are currently under study. Among the candidate TCO films, aluminum-doped zinc oxide (AZO) films have been considered as suitable anode materials, because they are more stable in reducing ambient, more abundant, and less expensive in comparison with ITO films [10–12].

To achieve optimal device performance, the work function of TCO layer, which directly affects the efficiency of hole injection to or collection from an organic layer, is of great importance [13,14]. It is well known that TCO work function is sensitive to the surface cleaning method [15–20]. Several studies focused on the relation between TCO surface treatment and its surface properties in order to clarify the effect of TCO surface treatment on its work function. For the case of ITO films, it has been reported that ultraviolet (UV)-

ozone treatment or plasma treatment in different atmospheres can influence their work function, attributed to the surface chemical composition, i.e. carbon-related contamination, O/In and In/Sn ratio [17–20]. However, few investigations have been reported on the effect of cleaning method on AZO work function and surface properties. Though it has been pointed out that the UV-ozone treated AZO film had a work function of 0.5–0.6 eV higher than that of the untreated film [10], further intensive study on the relation between work function and surface properties is needed.

In this work, organic solvent cleaning and UV-ozone cleaning were used as cleaning methods, the relation between work function and surface properties of AZO film under various cleaning methods were investigated using ultraviolet photoelectron spectroscopy (UPS) and X-ray photoelectron spectroscopy (XPS). The important factors that affect AZO work function were tentatively discussed.

### 2. Experiment

The AZO films grown on 7059 corning glass were prepared by radio-frequency magnetron sputtering using AZO ceramic with Al content of 4 at% as a target. The films were deposited at a constant argon pressure of 0.21 Pa and a fixed substrate temperature of 250 °C. The thickness and resistivity of the obtained AZO films were ~300 nm and  $\sim 1 \times 10^{-3} \Omega \text{ cm}$ , respectively. Herein, three samples subjected to different cleaning methods were used for comparison. One sample without any cleaning treatment was used as a reference. The other two samples were firstly immersed in ultrasonic

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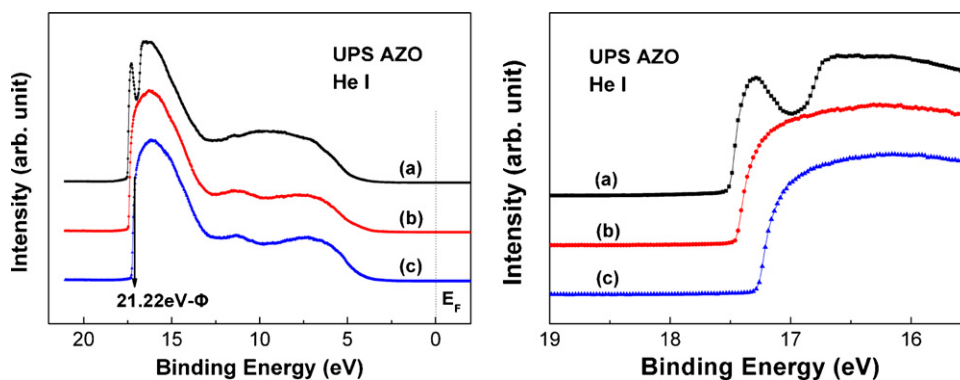


Fig. 1. He I UPS spectra of AZO samples without and with different surface treatments. (a) Without any treatment, (b) acetone solvent cleaning, and (c) UV-ozone cleaning.

baths of acetone and distilled water, each for about 15 min, then they were blown dry by  $N_2$  gas. Subsequently, one was reserved without any further cleaning treatment; the other was exposed to a 150 W lamp for UV irradiation for 20 min. In this case, the ambient oxygen was converted to ozone by UV irradiation, which reacted rapidly with residual organic molecular on sample surface, as a result, a clean surface can be produced. Following the cleaning procedure, all the samples were immediately transferred to an ultrahigh vacuum chamber equipped with UPS and XPS analysis.

The photoelectron measurements were performed using Kratos AXIS ULTRA<sup>DL</sup>. The analysis chamber was maintained at  $5 \times 10^{-7}$  Pa during XPS measurement,  $5 \times 10^{-6}$  Pa during UPS measurement due to He. The UPS spectra were recorded using a He I radiation ( $h\nu = 21.22$  eV) from an unfiltered gas discharge lamp with step size of 0.025 eV. The work function  $\Phi$  was determined by the following reaction:

$$\Phi = h\nu - \Delta E$$

Where  $\Delta E$  is the energy difference between Fermi level and secondary electron cutoff [15]. During UPS measurement, the samples were biased at  $-7.5$  eV to observe the secondary electron cutoff and, moreover, the Fermi edge location of each sample was estimated using silver as a reference. Furthermore, the surface chemical composition of AZO samples was determined from XPS spectra using a monochromatic aluminum  $K\alpha$  ( $h\nu = 1486.6$  eV) excitation as a light source. It should be mentioned, the XPS sensitive factor used in calculation was derived from CasaXPS.

### 3. Results and discussion

#### 3.1. AZO work function under different cleaning methods

Fig. 1 shows the UPS spectra of AZO samples subjected to different treatments. It can be clearly observed that the AZO work function, obtained by subtracting the cutoff value from 21.22 eV, was dependent on the cleaning method. For the sample without any cleaning treatment, it was as low as 3.83 eV; while after acetone solvent cleaning, it slightly increased to 3.94 eV; moreover, as soon as the sample exposed to UV-ozone treatment, it significantly increased to 4.26 eV. Therefore, it was concluded that both the applied surface cleaning methods can increase AZO work function and, moreover, UV-ozone cleaning was more effective.

#### 3.2. AZO surface chemical composition under different cleaning methods

As illustrated in Fig. 1, the applied cleaning methods, i.e. acetone solvent cleaning and UV-ozone cleaning, have different influence on AZO work function, the reason for which may be ascribed to

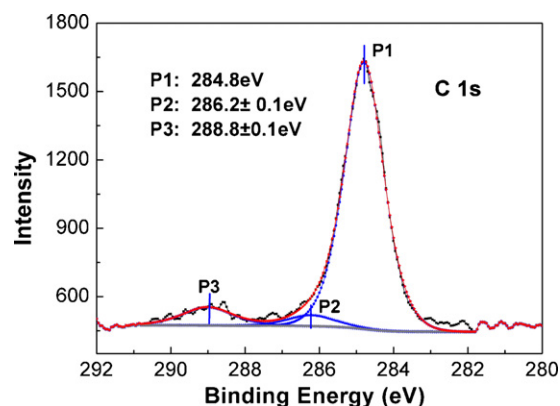


Fig. 2. The curve-fitted results of C 1s XPS spectrum of the AZO sample with UV-ozone cleaning.

surface chemical composition, especially  $O^{2-}$  concentration and Al concentration, as reported in previous papers [10,12,21]. Accordingly, Figs. 2–5 show the representative XPS spectra of C 1s, Zn  $2p_{3/2}$ , Al  $2p_{3/2}$  and O 1s peaks, and Table 1 summarizes surface chemical composition and work function of AZO samples under different surface cleaning treatments.

Based on the XPS spectra, carbon was found to be the dominant contaminant on sample surfaces, Fig. 2 shows the representative C 1s spectrum. As illustrated in Fig. 2, C 1s spectrum can be fitted into three peaks: C–H or C–C at 284.8 eV, C–O at  $286.2 \pm 0.1$  eV, and C=O or  $COO^-$  at  $288.8 \pm 0.1$  eV, apparently most carbon contaminant in surface was originated from C–H or C–C. Afterwards, the concentration of carbon-related contaminants was calculated, as shown in Table 1. It was found that the carbon concentration changed with

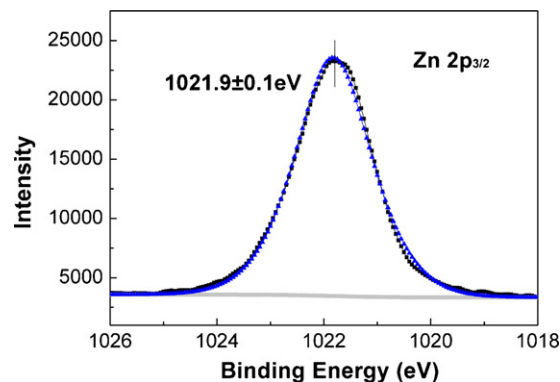


Fig. 3. The curve-fitted results of Zn  $2p_{3/2}$  XPS spectrum of the AZO sample with UV-ozone cleaning.

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