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# Toward Industrial Production of a High-Performance Self-Powered Ultraviolet Photodetector Using Nanoporous Al-Doped ZnO Thin Films

Manh Hoang Tran, Thi My Huyen Nguyen, and Chung Wung Bark\*



**ABSTRACT:** Al-doped ZnO (AZO) thin films are effective n-type semiconductors for ultraviolet (UV) detection because of their low cost, high electron mobility, and high sensitivity to UV light, especially in the UVA spectrum. However, a reasonable compromise between performance (such as sensitivity, detectivity, and response time) and fabrication ease remains an obstacle to the practicability of AZO-based UV photodetectors. To address this issue, we propose an efficient strategy to achieve a large AZO photoactive area for outstanding performance, along with a facile sol–gel method. Consequently, the device exhibits a superb on/off ratio of >10<sup>4</sup>, a high detectivity of 1.85 × 10<sup>12</sup> Jones, and a fast response speed under 365 nm UVA illumination without external energy consumption. Hence, this study suggests a self-powered and high-performance nanoporous AZO-based UVA detector with an environmentally friendly scalable process that satisfies industrial production requirements for numerous practical UV-detection applications.



# **1. INTRODUCTION**

Ultraviolet (UV) detection has gathered much attention due to its plentiful applications such as fire detection, biological sensing, environmental sensing, and secret wireless communication, civil, and astronomy systems.<sup>1</sup> Consequently, UV photodetectors, which enable the conversion of UV lightderived photon energy to electrical signals, have been intensively developed over the past decades, achieving not only high sensitivity and fast response times but also high stability, low power consumption, and light weight.<sup>2</sup> Nowadays, Ga-based semiconductors possessing wide bandgaps and high electron mobility (such as Ga<sub>2</sub>O<sub>3</sub>, GaN, and AlGaN) have been regarded as the benchmark photoactive materials for UV and deep-UV detection.<sup>3–7</sup> For example, a high-electronmobility transistor based on the AlGaN/GaN heterojunction could perform the giant responsivity to UVA (1.0  $\times$  10  $^{6}$  mA  $W^{-1}$  at 340  $\mu$ W/cm<sup>2</sup>) and UVC (3.6 × 10<sup>7</sup> mA W<sup>-1</sup> at 12  $\mu$ W/ cm<sup>2</sup>).<sup>4</sup> However, the preparation for a high-quality Ga-based thin film requires complicated processes. Moreover, the scalability of Ga-based devices is hindered due to the fact that gallium constitutes only 0.0005% of the Earth's crust.<sup>8</sup> In constrast, ZnO is regarded as an excellent candidate for practical and scalable optoelectronics owing to a wide bandgap (~3.37 eV), high thermal and chemical stability, and abundance in the Earth's crust.<sup>9,10</sup> Because of its n-type nature, ZnO is widely utilized as either a photoactive or an electron-transfer component for UV photodetectors. To improve the performance of ZnO-based detectors, several studies have focused on introducing trivalent metal cations

such as Al and Ga to increase the electron mobility and concentration of ZnO, thus enhancing the device performance.<sup>11–13</sup> In this study, we chose Al-doped ZnO (AZO) as the n-type photoactive constituent for the UV detector.

In addition to the nature of the active material, its morphology significantly affects the performance of the device. Several strategies for synthesizing complex nanostructures, such as nano-pencils, nanowalls, and nanocolumns, have been employed to increase the surface-to-volume ratio, aiming for a larger active area exposed to light.<sup>14–16</sup> For instance, Agrawal et al. recently grew an AZO nanowall network growing on Al foil for an on/off ratio of 349 and a high specific detectivity of  $4.5 \times 10^{10}$  Jones at an applied voltage of only 0.1 V under 350 nm UV radiation with light intensity as low as 396  $\mu$ W cm<sup>-2.15</sup> However, the device suffered from a slow response time (rise/ decay time of 150/210 s). Considering the concept of using nanostructured AZO films for photoactive region expansion, we prepared a continuous AZO thin film with a nanoporous structure using a facile sol-gel spin-coating process. To overcome the sluggish response speed, a p-type poly [9,9dioctylfluorene-co-N-[4-(3-methylpropyl)]-diphenylamine] (abbreviated as TFB) possessing a high hole mobility of  $1 \times$ 

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Figure 1. (a) XRD patterns of ZO and AZO (1.5 and 2 mol % Al) thin films. (b) UV-vis spectra, (c) Tauc plots, and (d) PL spectra (insets show illustrations of the corresponding band diagrams) of ZO and AZO (1.5 mol % Al) thin films.

 $10^{-2}$  cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> was coupled with the as-prepared AZO film to fabricate a p-n photodiode.<sup>17</sup> Two primary reasons are motivating this work to construct the UV photodetector based on the organic—inorganic p-n heterojunction. The first is that an organic polymer can easily penetrate the AZO film in addition to fully covering the porous oxide surface to form a large-area and compact p-n junction in which a huge amount of photogenerated electron—hole pairs upon UV illumination can be efficiently collected at the corresponding electrodes. The second is that the p-n photodiode possessing an intrinsic built-in potential is able to operate at 0 V with a fast response speed. For instance, Chen et al. proposed a p-type PANI/ntype ZnO-based UV detector performing a rapid rise/decay time of 0.11/1.45 ms at 0 V.<sup>18</sup>

Thereby, our device exhibited an outstanding performance with an on/off ratio of  $>10^4$ , a specific detectivity of 1.85  $\times$  10<sup>12</sup> Jones, and a fast response speed of ~0.1 s under 365 nm UVA illumination without external energy consumption. Hence, this study proposes a new approach for efficiently utilizing nanoporous thin films in practical photodetectors.

# 2. EXPERIMENTAL SECTION

**2.1. Preparation of AZO Thin Films.** The AZO thin films were fabricated using a facile sol-gel spin-coating method. First, 1 mmol Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O (99% Aldrich), 15  $\mu$ mol Al(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O (99.99% Aldrich), and 2.4 mmol citric acid (C<sub>6</sub>H<sub>8</sub>O<sub>7</sub>, Aldrich) were dissolved into a solution consisting 1 mL of 2-methoxyethanol (99.8% Aldrich) and 10  $\mu$ L of acetic acid (99.7% Aldrich). Then, the mixture was stirred at 500 rpm at 25 °C for 1 h to obtain a uniform precursor sol with 1.5 mol

% of the Al-to-Zn ratio. Finally, the sol was spin-coated at 4000 rpm for 30 s on the substrate, followed by annealing at 500 °C for 3 h to form a nanoporous AZO thin film. For comparison, undoped ZnO (denoted as ZO), 1 mol % Al-doped ZnO, and 2 mol % Al-doped ZnO thin films were prepared by replacing 15  $\mu$ mol of Al(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O with 0, 10, and 20  $\mu$ mol of Al(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O, respectively.

2.2. Preparation of Photodiode. A UV photodiode with a vertical p-n structure was fabricated by layer-by-layer spincoating on FTO-coated glass substrates. First, the substrates were cleaned in an ultrasonic bath for 5 min with acetone, deionized (DI) water, and ethanol, followed by drying using a nitrogen gun. Then, 100  $\mu$ L of a solution consisting of titanium diisopropoxide bis(acetylacetonate) (75 wt % in isopropanol, Aldrich) and ethanol (1:10 v/v ratio) was spin-coated on the substrate at 2000 rpm for 40 s, followed by annealing at 500 °C for 3 h to form a hole-blocking TiO<sub>2</sub> layer. After the porous AZO thin film was applied on the TiO<sub>2</sub>-coated substrate, a ptype layer was deposited onto the AZO film by spin-coating 100  $\mu$ L of TFB ( $M_w$  > 30,000, Aldrich) solution in chlorobenzene (30 mg mL<sup>-1</sup>) at 3000 rpm for 40 s. After drying the TFB film on the hot plate at 120 °C for 5 min, the aqueous PEDOT:PSS solution (1.1 wt % in H<sub>2</sub>O, Aldrich) was spin-coated at 1000 rpm for 60 s, followed by heating on the hot plate at 120 °C for 5 min to form the top electrode. In this work, the photoactive area of the vertical-structured device was defined as the region where the FTO, TiO<sub>2</sub>, AZO, TFB, and PEDOT: PSS layers overlapped (fixed at  $0.5 \times 0.5 \text{ cm}^2$ ).

2.2.1. Analysis. The crystal structures of ZO and AZO thin films were determined by X-ray diffraction (XRD, Rigaku DMAX 2200, Japan) using Cu K $\alpha$  radiation ( $\lambda$  = 1.5406 Å) at



Figure 2. (a) SEM images and (b) elemental distributions of AZO (1.5 mol % Al) thin film. (c) Schematic structure and energy band diagram of the AZO-based photodiode.

a scan rate of 1  $^{\circ}$  min<sup>-1</sup>. The ultraviolet–visible (UV–vis) spectra and steady-state photoluminescence (PL) spectra of the thin films were measured using an Agilent 8453 (USA) instrument and PTI QuantaMaster TM50 (USA), respectively. The morphologies and elemental compositions of the AZO films were analyzed using scanning electron microscopy (SEM, Hitachi S-4700, Japan). Electrical resistivities of as-prepared films were measured at 25 °C by the four-probe resistivity method (CMT-SR2000N, South Korea).

Current-voltage (I-V), current-time (I-t), and voltagetime (V-t) plots were obtained using a Keithley sourcemeter (2410, USA) equipped with a UV lamp (365 nm, Vilber Lourmat VL-6, France). Spectral responsivity was measured at 300-800 nm using a solar cell quantum efficiency test system (McScience K3100, Korea).

## 3. RESULTS AND DISCUSSION

Figure 1a shows the XRD patterns of the as-prepared ZO and AZO films. Both undoped and doped thin films exhibited characteristic peaks at  $2\theta = \sim 31.8$ , 34.4, 36.3, 47.5, 56.6, 62.9, 66.4, 67.9, and 69.1° corresponding to (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes, respectively, of hexagonal ZnO (ICSD, 79-0206). Compared to the ZO film, there were no impurity diffraction peaks for the AZO film, indicating that Al<sup>3+</sup> was uniformly distributed in the original ZnO structure without the formation of secondary crystalline phases, such as  $Al(OH)_3$  or  $Al_2O_3$ . However, Al incorporation led to broadening of the peak intensity (Figure 1a) and a slight shift in the peak position (Figure S1a), implying a change in lattice stress as  $Zn^{2+}$  sites were substituted by  $Al^{3+}$  sites in the ZnO lattice.<sup>8</sup> Besides, the atomic dislocations and strains in the ZnO lattice as Zn-substituted Al sites were confirmed by the increase in lattice strain with the dopant concentrations (more detailed calculation is provided in Figure S1b).

The effect of Al doping on the optical properties of ZnO was examined by UV-vis spectroscopy of ZO and AZO prepared on quartz substrates (Figure 1b). The ZO and AZO films exhibit a strong UVA absorbance peak centered at ~350 nm with a sharp absorption edge below 400 nm, reflecting the feasibility of UVA detection. The optical bandgaps  $(E_{\sigma})$  of ZO and AZO were determined by Tauc plots, which can be expressed as  $\alpha h\nu = D(h\nu - E_g)^{1/2}$ , where  $\alpha$  is the absorption coefficient,  $h\nu$  is the photon energy, and D is a constant.<sup>19</sup> After Al doping, the optical bandgap of ZnO slightly increased from 3.28 to 3.3 eV (Figure 1c), which was consistent with previous studies.<sup>11,20,21</sup> This phenomenon can be explained by the Burstain-Moss effect when the electron carrier concentration exceeded the conduction band (CB) of ZnO due to the high doping with Al.<sup>20</sup> The enhancement of electron concentration of ZnO due to Al doping was also confirmed by the decrease in the electrical resistivity from 7672  $\Omega$  cm (for the ZO film) to 3145  $\Omega$  cm (for the AZO film at 1.5 mol % Al content). To determine the energy states present in AZO, PL spectra of ZO and AZO (1.5 mol % Al) at the excitation wavelength of 325 nm were analyzed. As shown in Figure 1d, both the PL spectra showed the emission peaks at 3.30-3.28 eV, 2.98-3.01 eV, 1.94-2.00 eV, and 1.74-1.75 eV assigned to the transitions from the ZnO conduction band (CB) to the ZnO valence band (VB), Zn-vacancy ( $V_{Zn}$ ), O-interstitial (O<sub>i</sub>), and O-vacancy ( $V_O$ ), respectively.<sup>21,22</sup> Notably, the PL of AZO additionally showed a prominent emission peak at 3.22 eV, corresponding to the transition from the Zn-substituted Al  $(Al_{7n})$  state to VB.<sup>21,22</sup> This confirms that the doping Al would introduce Zn-substituted Al sited in the ZnO system, agreeing with the XRD results.

The morphology and elemental distribution of AZO films were observed by SEM (Figure 2a). At the large scale (scale bar of 5  $\mu$ m), the AZO film appeared to be smooth and continuous without micro-cracks and voids. Interestingly, the film possesses a nanoporous structure with numerous nano-



Figure 3. (a) Linear and (b) semi-logarithmic I-V curves and (c) multi-cycle and (d) individual cycle I-t plots at 0 V of a photodiode in dark and under 365 nm UV light (340  $\mu$ W cm<sup>-2</sup>).

scale cavities (high-magnification scale bar of 500 nm). The same morphology was observed in the ZO film (Figure S2), demonstrating that the Al dopant did not change the appearance of the ZnO film. It is thought that the nanoporosity present in the continuous films is due to the ease of releasing volatile solvents (e.g., acetic acid and 2-methoxyethanol) from the spin-coated thin films during annealing. Another reason is the out-diffusion of gases produced by the thermal decomposition of metal citrate complexes (e.g., N<sub>2</sub> and  $CO_2$ ), as follows:<sup>23–25</sup>

$$Zn(NO_3)_2 \cdot 6H_2O + C_6H_8O_7$$

$$\rightarrow Zn_x(C_6H_{8-y}O_7)_z$$

$$\xrightarrow{500^{\circ}C \text{ in } O_2} ZnO + H_2O + N_2 + CO_2$$
(1)

In addition, EDX elemental mapping showed a uniform distribution of Zn and Al on the AZO film (Figure 2b) with an Al/Zn ratio of 1.31% (Figure S3), which is in good agreement with the theoretical loading (Al/Zn ratio of 1.5%).

To apply the AZO thin film to a self-powered UVA photodetector, we constructed a p-n photodiode in the vertical structure of  $FTO/TiO_2/n$ -type AZO/p-type TFB/PE-DOT:PSS, in which FTO and PEDOT:PSS were used as the bottom and top electrodes, respectively, for the collection of electrons and holes. The schematic structure and energy band diagram of the device are illustrated in Figure 2c.<sup>19,26,27</sup> Here, a compact TiO<sub>2</sub> ultrathin film was used as the interfacial layer because the nanoporosity of the AZO film could lead to undesired contact between the TFB and FTO layers. The valence band (VB) of the compact TiO<sub>2</sub> layer was lower than that of the TFB, which hindered hole transport from the TFB to the FTO. Furthermore, the CB of TiO<sub>2</sub> is located between that of AZO and the Fermi level of FTO, facilitating electron extraction from AZO to FTO.<sup>27</sup>

To assess the influence of the Al concentration on the photoresponse of the device, diodes using p-n junctions of ZO/TFB and AZO/TFB (at Al concentrations of 1, 1.5, and 2 at%) were fabricated for I-t measurements at 0 V under the on and off states of 365 nm UV light (as shown in Figure S4). The current in light  $(I_{\rm L})$  of the AZO-based devices was higher than that of the ZO-based devices and achieved the maximum  $I_{\rm L}$  at a concentration of 1.5%. The dark current  $(I_{
m D})$  remained almost constant with Al doping. The enhanced I<sub>L</sub> for the AZObased devices compared to that for the ZO-based devices originates from three factors. The first is the expansion of the p-n depletion width for more incident photons absorbed during the depletion, leading to more photogenerated electrons and holes.<sup>28</sup> Notably, the p-n depletion region is considered the photoactive layer of a p-n photodiode. The second reason is the higher electrical conductivity of the outof-depletion AZO region, which facilitated electron transfer to the FTO electrode. The third reason is relevant to altering energy mismatch at TiO<sub>2</sub>/ZnO interfaces due to Al doping. As shown in Figure S5, after the individual FTO, TiO<sub>2</sub>, and ZO (AZO) layers contact, their Fermi levels would align and create an energy mismatch between ZO (AZO) CB and TiO<sub>2</sub> CB with the theoretical value of 0.67 eV (0.12 eV). Owing to the significantly lower mismatch, the photogenerated electrons could be easy to move from AZO to TiO<sub>2</sub> as compared to the case of ZO. However, when the concentration of Al was increased to 2 at%, the significant distortion in the ZnO structure caused more lattice defects as carrier traps, leading to the degradation of the electrical conductivity of the AZO film.<sup>11,29</sup> As a result, the  $I_{\rm L}$  decreased as the Al concentration increased from 1.5 to 2% (Figure S4). Therefore, the optimum Al concentration chosen for subsequent measurements was 1.5 mol % Al.

Figure 3a,b shows the linear and semi-logarithmic I–V curves of the optimized device under dark conditions and 365

Table 1. Performance Comparison of Recent	AZO-Based UV Photodetectors
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active material	process	bias (V)	$\lambda$ (nm)	$D^* \times 10^{10}$ (Jones)	on/off ratio	response time (s)	ref.
AZO-Si	pulsed laser deposition	1	368		4.9		36
AZO film	sol—gel		365	130		30	37
AZO nanowire	sol—gel	10	365		56	15/52	38
AZO film	chemical vapor deposition	2	365		10 <sup>6</sup>	$\sim 20$	39
AZO/ZnO/PVK	sputtering	5	365	0.35	<10	0.11/0.2	40
AZO/CuSCN	sol—gel	0	365		<10	>10	41
AZO film	spray pyrolysis	5	365		<5	>1000	42
AZO film	sputtering	4	380		800	0.2/0.3	37
AZO film	sputtering	1	385	1.0	24	2.61/2.85	43
AZO/CuCrO <sub>2</sub>	sputtering	1	385	352	$4 \times 10^{4}$	0.21/0.26	43
AZO nanowall	hydrothermal reaction	0.1	350	4.5	349	150/210	15
AZO/TFB	sol-gel	0	365	185	>10 <sup>4</sup>	~0.1	this work



**Figure 4.** (a) I-t plot in a short-circuit mode, (b) V-t plot in an open-circuit mode, (c) photocurrent ( $I_P$ ) and photovoltage ( $V_P$ ) as the functions of light intensity, and (d) responsivity (R), specific detectivity ( $D^*$ ), and external quantum efficiency (EQE) of the device at 0 V under 365 nm UV illumination in 80–370  $\mu$ W cm<sup>-2</sup>.

nm UV illumination (340  $\mu$ W cm<sup>-2</sup>). In the dark, the I–V curve shows diode-like rectifying characteristics with an asymmetrical shape, demonstrating good p-n contact formation. Under 365 nm light, the device performed a selfpowered operation, accompanied by an outstanding on/off ratio (defined as the  $I_{\rm L}/I_{\rm D}$  ratio) of >10<sup>4</sup> and a high opencircuit voltage of 0.38 V. The I-t plot of the device shows an unchanged  $I_D$  and a slightly increased  $I_L$  over 140 continuous on/off cycles (Figure 3c), suggesting good stability of the device. This gradual increment of  $I_{\rm L}$  can be explained by the oxygen desorption from oxide constituents of the device (e.g., AZO, TiO<sub>2</sub>, and FTO layer) under long-time UV light exposure, which decreased the total electrical resistance of the device.<sup>30</sup> Taking an individual on/off cycle into account (Figure 3d), our photodiode revealed a faster or comparable response speed (rise/decay time of 105/94 ms) as compared to those in previous research (Table 1). However, our

response speed (in the order of ms) is still much higher than the response times (only in the order of  $\mu$ s) reported elsewhere,<sup>31–33</sup> where the response times were measured with the pulsed laser. It can be acceptable because, herein, the rise time and decay time are defined as the periods in which photocurrent increases from 10 to 90% and drops from 90 to 10% of its saturation level, respectively.<sup>34</sup>

The dependence of the photocurrent ( $I_p$ , defined as  $I_L - I_D$ ) and photovoltage ( $V_p$ , defined as the difference between the open-circuit voltages with and without light illumination) on the incident light intensity (P) was determined using y-t (in a short-circuit mode, Figure 4a) and V-t (in an open-circuit mode, Figure 4b) plots. When P increased from 80 to 370  $\mu$ W cm<sup>-2</sup>, the gradual increase in  $I_p$  and  $V_p$  with light intensity reflected the efficient photovoltaic effect of AZO/TFB depletion (Figure 4c). By contrast,  $I_D$  remained ultralow and unchanged with increasing  $P_i$ , resulting in an on/off ratio



**Figure 5.** (a) Schematic illustration of the operating mechanism of the device upon UV irradiation. (b) I–t plots and (c) responsivity (R), specific detectivity ( $D^*$ ), and external quantum efficiency (EQE) of devices with different applied reverse biases.

proportional to *P* (Figure S6). The relationship between  $I_p$  and *P* could be fitted by a simple power law:  $I_p \propto P^{0.99}$ . The close-to-unity exponent (0.99) indicated the effective transfer of photogenerated charges to the electrodes with suppressed recombination. Consequently, the responsivity (*R*), specific detectivity (*D*\*), and external quantum efficiency (EQE) derived from eqs 2–34 were almost independent of the light intensity in the investigated range.<sup>34</sup>

$$R = \frac{I_{\rm P}}{P} = \frac{I_{\rm L} - I_{\rm D}}{P} \tag{2}$$

$$D^* = \frac{RA^{0.5}}{(2eI_D)^{0.5}}$$
(3)

$$EQE = R \frac{hc}{e\lambda}$$
(4)

where A is the active area of the device, e is the electron charge, h is the Planck constant, and c is the speed of light. In addition, the spectral responsivity photodetector (Figure S7) exhibited selective detection of UVA with a maximum response at ~385 nm, consistent with the bandgap of the prepared AZO thin film. The expansion of detectable wavelength reaching ~445 nm was assigned to the contribution of the relatively narrow-bandgap TFB constituent  $(E_g = 3 \text{ eV})^{35}$  in the photoactive AZO/TFB depletion.

The operating mechanism of the AZO-based photodiode is illustrated in Figure 5a. Owing to the nanoporous structure of the AZO film, the TFB polymer can easily penetrate the AZO film in addition to fully covering the AZO surface to form a large area of AZO/TFB depletion. In other words, the nanoporosity of the AZO film can take advantage of the generation of an immense photoactive area, producing numerous photoexcited charges. Because their Fermi levels were aligned after contact, a built-in potential  $(E_{bi})$  was formed during the p-n depletion. Upon UVA irradiation, the AZO/ TFB junction was excited to generate electron—hole pairs. In the presence of  $E_{bi}$ , the electron—hole pairs are dissociated into electrons and holes and then rapidly swept toward the out-ofdepletion AZO and TFB regions, respectively. Eventually, the separated electrons and holes are transferred to the TiO<sub>2</sub> thin film (then the FTO electrode) and PEDOT:PSS electrode, respectively. Compared to AZO-based photodetectors from previous reports (Table 1), our device exhibited better performance with self-powered operation, fast response time, and excellent on/off ratio (>10<sup>4</sup>) and  $D^*$  (1.85 × 10<sup>12</sup> Jones) while having a more facile fabrication process.

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Finally, the effect of the external voltage on the performance of the photodiode was analyzed using I–t measurements at various reverse biases under 365 nm UV light (370  $\mu$ W cm<sup>-2</sup>). When a reverse voltage was applied, photogenerated electrons and holes were efficiently extracted from the depletion region toward the FTO and PEDOT:PSS sides, respectively. As a result, the more charges collected at the electrodes could contribute to a higher  $I_{\rm L}$  (Figure 5b), which enhanced R(EQE) from 30.8 mA W<sup>-1</sup> (10.4%) at 0 V to 42.8 mA W<sup>-1</sup> (14.6%) at 1 V, respectively (Figure 5c). However,  $I_{\rm D}$  also increased remarkably with the amplitude of the external bias (Figure 5b) owing to the Zener effect,<sup>28</sup> resulting in a significant decline in  $D^*$  from 10<sup>12</sup> to 10<sup>10</sup> Jones (Figure 5c).

#### 4. CONCLUSIONS

A nanoporous AZO thin film was prepared via a simple sol-gel spin-coating process and then applied to a vertical p-n photodiode. The fabricated device exhibited an excellent photoresponse to 365 nm UV light at zero bias with an outstanding on/off ratio (>10<sup>4</sup>), high detectivity ( $1.85 \times 10^{12}$  Jones), and fast response speed (105/94 ms) owing to its

large-area and high-quality p-n depletion region. This study provides new insights into the efficient utilization of nanoporous thin films for practical applications in futuristic photodetectors.

# ASSOCIATED CONTENT

# **Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.3c05266.

Fitted XRD peaks at (100) planes; lattice strain values of ZO and AZO (with 1.5 and 2 mol % Al); SEM images of the ZO thin film at scale bars of 5  $\mu$ m and 500 nm; EDX analysis of the AZO (1.5 mol % Al) thin film prepared on a quartz substrate; photoresponses of devices using ZO and AZO films with different ratios of Al at 0 V; individual band diagrams of constituents before contacting and band alignments after contacting; on/ off ratio of the AZO (1.5 mol % Al)-based device at 0 V under 365 nm UV light in the intensity range of 80–370  $\mu$ W cm<sup>-2</sup>; and wavelength-responsivity plot of the AZO (1.5 mol % Al)-based device at 0 V (PDF)

# AUTHOR INFORMATION

## **Corresponding Author**

Chung Wung Bark – Department of Electrical Engineering, Gachon University, Seongnam-si, Gyeonggi-do 13120, South Korea; orcid.org/0000-0002-9394-4240; Email: bark@ gachon.ac.kr

## Authors

Manh Hoang Tran – Department of Electrical Engineering, Gachon University, Seongnam-si, Gyeonggi-do 13120, South Korea

Thi My Huyen Nguyen – Department of Electrical Engineering, Gachon University, Seongnam-si, Gyeonggi-do 13120, South Korea

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.3c05266

## **Author Contributions**

M.H.T.: conceptualization, methodology, investigation, and writing—original draft. T.M.H.N.: conceptualization, analysis, and validation. C.W.B.: writing—review and editing, project administration, and supervision.

# Notes

The authors declare no competing financial interest.

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