Materials Science in Semiconductor Processing 88 (2018) 86–92

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: www.elsevier.com/locate/mssp

All metal oxide-based transparent and flexible photodetector

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ARTICLE INFO

Keywords:
Metal oxide
Transparent
Flexible
Photodetector

ABSTRACT

We demonstrate the fabrication and characterization of flexible and transparent photodetector based on conformally grown $Ag_xO/TiO_2/ITO$ layers on polyethylene terephthalate substrate. Initially, the thickness of Ag_xO and ITO layers were optimized via deposition time to match layers crystallinity with fixed intermediate TiO_2 layer, for better optical and electrical properties. The highly transparent (> 70%) and flexible device presents the high-preforming responses over the broadband light. Under UV illumination, it showed a high responsivity (323 mA/W) and detectivity (4.2×10^8 Jones). Similarly, a noise equivalent power of 2.3×10^{-9} W/Hz$^{1/2}$ is obtained, which confirmed its capability to detect light at nanowatt level. Moreover, mechanical flexibility test was performed for the flexible and transparent $Ag_xO/TiO_2/ITO$ device. It was found that the device is robust to have good optoelectronic performances even after 500 no. of bending cycles. Thus, the utilization of earth abundant metal-oxides can facilitate the flexibility, durability and transparency of next generation optoelectronic devices.

1. Introduction

Photodetectors (PDs) are the optoelectronics devices, which work on the principle of the photoelectric effect formulated by Einstein in 1905. Generally, photodetector is composed of an active layer sandwiched between two contact electrodes. The active layer is a light sensitive material that absorbs illuminating photon energy and generates the electron hole pairs. The light-induced photogenerated carriers are collected to the contact electrode to provide light-induced current flow to the external circuit [1,2]. PDs have been widely applied in medical, military, space exploration, flame detection, advanced communication and environmental monitoring [2]. Therefore, the fabrication of low cost, light weight, flexible and transparent PDs at room temperature by exploring novel materials and using simple fabrication techniques have remained a huge interest. Further, transparent flexible PDs can revolutionize wearable electronics, skin sensors, and implantable biological devices [1]. However, the realization of high transparency and better mechanical flexibility along with efficient optoelectrical performance PD is still a big challenge.

The organic-inorganic perovskites and metal oxides are the potential candidates for the fabrication of transparent and flexible PDs. In general, perovskite-based PDs show better optical and electrical performances [3], however they have critical issues of instability and toxicity [4,5]. For example, the Jeon et al. employed the perovskite ($CH_3NH_3PbI_3$) nanoparticles to design the transparent (80%) and flexible high-speed, photodetectors [3]. Similarly, Hu et al. exploited the lead-based perovskite to design the flexibly robust photodetector to show excellent responses to the wide band spectral range with a high responsivity and external conversion efficiency of 3.49 A/W and 1.19×10^3%, respectively [6]. On the other hand, the metal oxide-based PDs are eco-friendly and can bear the environmental condition [7]. Nonetheless, the metal oxides usually require high temperature for crystallization with complex fabrication steps [8], which limit their applications for flexible electronics. However, some of the metal oxides like V_2O_5 [7], ZnO [9], Zn_2GeO_4 [10], AuO [11] and CdO [12], have been employed for flexible PDs. As Kim et al. utilized the V_2O_5 to design a broadband, transparent and flexible photodetector with a high detectivity of 1.45×10^{12} Jones and response speed of 4.9 ms [7]. The Willander et al. employed the n-type ZnO with p-NiO to design a flexible photodetector with the fine heterojunction having an ideality factor of 7. [9]. Interestingly, Liu et al. transferred the Zn_2GeO_4 nanowires on a PET substrate to design a flexible photodetector with a response time of 1/15 s while retaining the electrical properties even after 100 no. of bending cycles [10]. Liu et al. adopted the AuO-decorated graphene for a flexible and broadband photodetector, which demonstrated a high responsivity of 3300, 58 and 9 A/W with light wavelength of 310, 500 and 1550 nm, respectively [11]. Similarly, the Zheng et al. designed a broadband photodetector based on the CdO-ZnO nanowire array with

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https://doi.org/10.1016/j.mssp.2018.07.027
Received 9 May 2018; Received in revised form 30 June 2018; Accepted 17 July 2018

high flexibility and excellent transparency (95%) [12].

Moreover, TiO_2 is one of the great interest for metal oxide-based photoelectric applications, which have been demonstrated by using chemical and physical fabrication techniques [13,14]. The remarkable feature of TiO_2 is the ninth abundant natural material in earth crust [15] and has a large bandgap (E_g = 3.2 eV) [13,16]. Likewise, the optical and electrical properties of silver oxides are also widely studied but mainly utilized for photocatalysis and optical storage applications [17–19]. It exists in multivalent form as AgO, Ag_2O, Ag_3O_4 and Ag_2O_3, where the band gap can be tuned in a wide range (1.2–3.4 eV), [20] depending on the deposition time and oxygen concentration [21]. In magnetron sputtering, normally the biphase oxide e.g. Ag_xO (AgO + Ag_2O) is formed [22]. It has a high transmittance in visible wavelength region and reported to have *p*-type conductivity [19]. Thus, optimizing and designing simple fabrication steps for integrating *p*-type silver oxide with *n*-type material can extend the flexible optoelectronics application, which is yet to be explored.

In this article, we fabricated the vertical stacks of metal oxides (Ag_xO/TiO_2/ITO) for the highly transparent photodetector. Due to the sputtering process at room temperature, a flexible polyethylene terephthalate (PET) was used as a substrate to realize the flexible electronics. The XPS and SEM were used to study the chemical and surface properties, respectively. The optical measurement of the device confirmed a high transmittance in visible region. In addition, the electrical properties were analyzed, which verified the device systematically responding to the varying light intensity. Similarly, the mechanical flexibility test (MFT) reveals the device robust mechanical flexibility by retaining both optical and electrical properties even after 500 no. of bending cycles. Furthermore, the important features of photodetectors like photoresponse speed, responsivity, detectivity, normalized photocurrent to dark current ratio and noise equivalent power were investigated for this flexible and transparent Ag_xO/TiO_2/ITO device.

2. Experimental section

2.1. Device fabrication

The flexible and transparent Ag_xO/TiO_2/ITO device was realized by the sequential 3-step sputtering depositions at room temperature, as shown in Fig. 1a. Prior to the sputtering process, the PET substrate was chemically cleaned by methanol and DI water, subsequently. A substrate was loaded in the sputtering system and deposited transparent layers in the sequence of ITO, TiO_2, and Ag_xO. Each layer deposition condition is summarized in the Table 1. The ITO layer works as a back contact and was partially covered by the kepton tape before the deposition of TiO_2 layer. The Ag_xO layer was deposited onto the TiO_2 layer, which also serve as the top contact. The optimum thickness of 4, 8 and 16 nm for Ag_xO was investigated for better optical and electrical properties by varying the deposition time for 30, 60 and 120 s under an identical sputtering condition.

2.2. Characterization

The oxides of the silver and titanium were confirmed with X-ray photoelectron spectroscopy (XPS-PHI 5000 Versa Probe II) and then scanning electron microscopy (SEM-JSM7800F, Jeol) was used to analyze the surface morphology. The optical analysis was performed in UV–Visible-NIR spectroscopy (Shimadzu, UV-2600) with air as baseline. The current voltage (I-V) characteristics were measured using linear sweep voltmetery (LSV) function of Potentiostat/Galvanostat (Zive SP1, ZIVELAB) and its chronoamperometry function was utilized to check the dynamic photoresponse with three LEDs light (365 ± 10 nm, 450 ± 10 nm, 560 ± 10 nm, LEDENGIN) powered from a functional generator (MFG-3013A). The varying intensity of the light is measured via photometer (TES-1333 solar power meter).

3. Result and discussion

3.1. Schematics, chemical composition and energy band diagram

The schematic diagram of the sequentially grown ITO, TiO_2 and Ag_xO layers is shown in Fig. 1a. The optimized ITO layer (570 nm) was deposited as electron transporting layer for *n*-type TiO_2 [23], while the holes were directly transported to the Ag_xO. In addition, the device photograph in Fig. 1b portrays the highly transparent and flexible nature of the as-fabricated device.

Fig. 1c shows the high resolution XPS results of the deposited transition metal oxides. In the binding energy range of 450 and 468 eV,

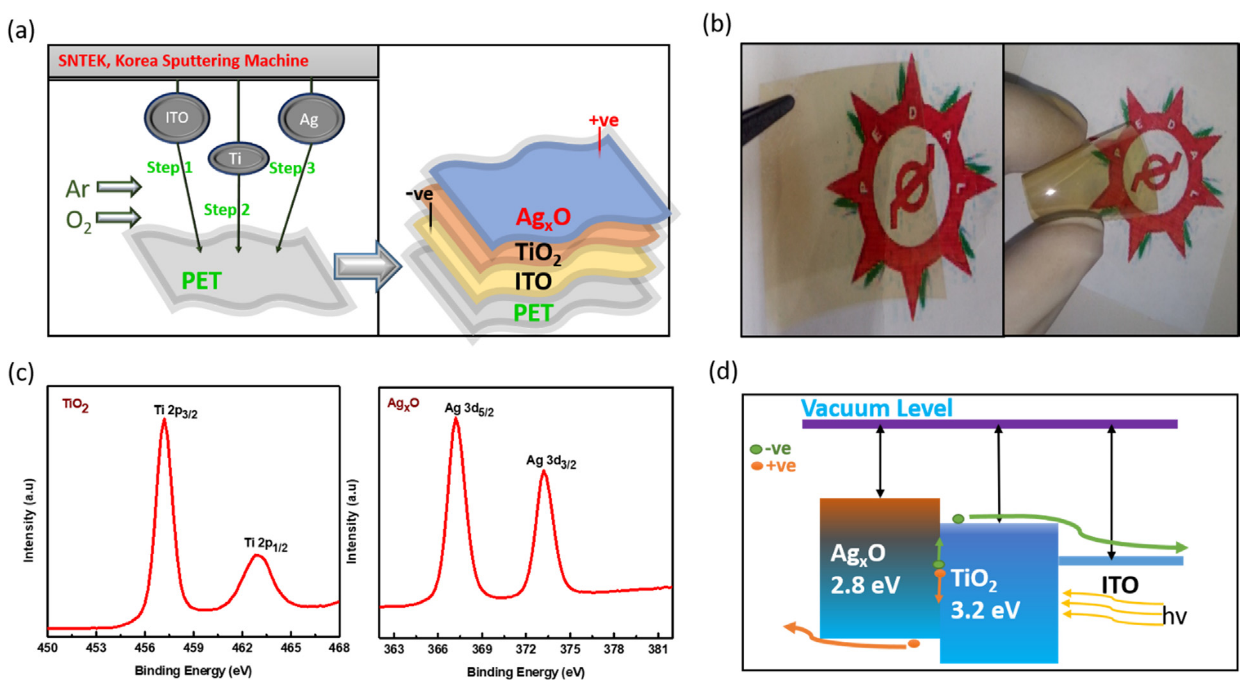

Fig. 1. (a) Fabrication steps and schematic diagram, (b) device photographs, (c) XPS spectra of Ti and Ag and (d) Energy band diagram.