



Fabrication of solution-processed nitrogen-doped niobium zinc tin oxide thin film transistors using ethanolamine additives



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ARTICLE INFO

Article history:

Received 7 June 2017

Received in revised form

5 September 2017

Accepted 14 September 2017

Available online 18 September 2017

Keywords:

Solution process
Monoethanolamine
Thin film transistor
Nitrogen
Oxide

ABSTRACT

In this study, we fabricated the nitrogen-doped NbZnSnO channel layers by using the sol-gel method. Monoethanolamine (MEA) was used as a nitrogen additive. From the XPS results, the concentration of oxygen vacancies changes as a function of MEA/Nb ratio. A NbZnSnO film with MEA/Nb of 0.2 shows the lowest amount of oxygen vacancies. TFT electrical performance also shows a device with an MEA/Nb ratio of 0.2 possesses a high carrier mobility ($7.4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and good bias stress stability. In addition, we also investigated the effect of the aging time of precursor solution on the electrical characteristics of the TFT. After adding MEA, the annealing temperature of the NbZnSnO channels can be reduced, pertaining to the acceleration of the hydrolysis and condensation reaction.

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1. Introduction

In recent years, ZnSnO films have received much attention in the application for channels of thin film transistors (TFTs) [1]. The devices show a high carrier mobility in ambient air, a large on-to-off current ratio, and low production cost in comparison with Si-based TFTs [2,3].

In addition, solution-derived thin films have been widely investigated, which possesses many merits such as cost reduction, low-temperature fabrication, the simplicity of the process, the precise control of the stoichiometry, and high yield as compared with vapor deposition techniques. Therefore, solution processed ZnSnO has been utilized as a channel material in microelectronic. It is well known that the bias stress instability of TFTs is related to the amounts of oxygen vacancies in channel layers [4]. To improve the bias stress instability during device operation, some works have investigated ZnSnO channels added with various transition metal cations [5,6]. Our previous study [7] showed that introducing Nb into the ZnSnO channel materials could enhance the electrical performance of TFTs. Some literature [8,10] indicated that nitrogen anions can successfully substitute to O and eliminate the amounts

of oxygen vacancies in metal oxide channel layers because the ionic radii of N^{2-} (0.129 nm) and N^{3-} (0.132 nm) are similar to that of O^{2-} (0.126 nm). The effective substitution of oxygen ions by nitrogen ions is based on the Hume-Rothery rules [9] where the ionic radii difference between the solute and solvent atoms should be no more than 15%. However, the N-doped metal oxide channel layers in the previous studies were fabricated by vacuum deposition techniques instead of solution processes [10,11]. In addition, monoethanolamine (MEA) was often used as a stabilizing agent for fabrication of solution-processed metal oxide channel layers [12,13]. In this study, we attempt to use MEA as a nitrogen additive for adding N into the host NbZnSnO channel layers to enhance the electrical performance of solution-processed NbZnSnO TFTs. We demonstrate the production of NbZnSnO device with MEA/Nb of 0.2 shows a mobility of $7.4 \text{ cm}^2 \text{ VV}^{-1} \text{ s}^{-1}$, an on-to-off current ratio of 10^8 and a small subthreshold swing ($\sim 0.3 \text{ V/decade}$) by using a solution process technique. At the same time, we try to adjust the oxygen vacancies of the N-doped NbZnSnO TFTs using different amounts of MEA additives. In addition, the effect of MEA on the aging time of NbZnSnO precursor is also investigated. Finally, the correlation among the MEA-doping concentration, aging time, and device performance is discussed.

2. Material and methods

NbZnSnO channel layers with adding different amounts of

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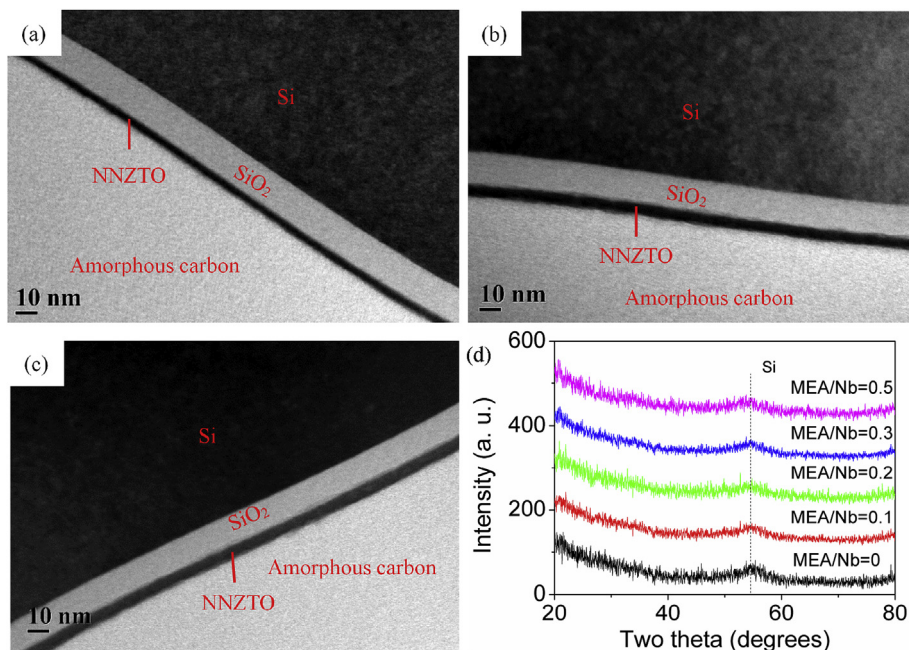


Fig. 1. Cross-section TEM photographs of the NbZnSnO films with MEA/Nb molar ratios of (a) 0, (b) 0.2, and (c) 0.5 on $\text{SiO}_2/\text{p}^+-\text{Si}$. (d) GIAXRD patterns of 10-layers NbZnSnO channel layers doped with various MEA/Nb ratios.

monoethanolamine (MEA) were fabricated via a combined sol-gel and spin-coating method. From our previous study [7], an optimum Nb-doping concentration of solution-processed NbZnSnO TFTs is 3%, which possesses the best characteristics of the device. Therefore, the molar ratio of Nb: Zn: Sn in NbZnSnO precursors was chosen as 0.03:1:0.97. All reagents, anhydrous niobium chloride (NbCl_5), hydrated Zinc nitrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) and hydrated tin chloride dihydrate ($\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$) were dissolved in 20 ml ethylene glycol monomethyl ether (EGME) for preparing the NbZnSnO precursors. For MEA dopants, different amounts of MEA used as starting materials were weighed according to the molar ratios of MEA/Nb (i.e., 0, 0.1, 0.2, 0.3, and 0.5). After stirring for 20 h at room temperature, the NbZnSnO precursors with and without adding MEA achieved chemical homogeneity. Additionally, the precursor solutions were stirred for 2, 6, and 20 h, based on the color variation of NbZnSnO precursor, to proceed with the aging experiment by Fourier transform infrared absorption spectroscopy (FTIR) analysis and TFT characteristics. The NbZnSnO precursors were spin-coated onto a heavily doped p-type Si wafer with a 100 nm thick SiO_2 layer, which functions as the gate electrode and gate dielectric. The spin-coated NbZnSnO active layers with and without MEA additives were rapidly baked at 110 °C for 3 min and subsequently annealed on a heating plate at 430 °C for 1 h under an air atmosphere. To verify the center of the plate surface controlled by the temperature controller, the temperature calibration of the heating plate was performed by a thermocouple. Finally, the 300-nm-thick Al source and drain electrodes were deposited on top of the active layer using an e-beam evaporation technique through a shadow mask to define transistor with the channel width (2000 μm) and length (100 μm).

The thickness of the NbZnSnO film with and without MEA additives was determined using a transmission electron microscope (TEM, PHILIPS CM-200). To prepare the TEM-specimen, a focused ion beam instrument (FIB) was used to thin the samples for electron transparency. A protective layer of carbon was deposited on the NbZnSnO films with and without MEA additives

to prevent beam-induced damage during the milling process. The chemical compositions and oxygen vacancies were carried out with the x-ray photoelectron spectroscopy (XPS, JEOL JAMP-9500F). A Hitachi U-2001 UV/Visible Spectrophotometer was used to measure the optical transmittance of the NbZnSnO composite films with and without adding MEA that were deposited on quartz glass substrates. Fourier transform infrared absorption spectroscopy (FTIR) was used to characterize the bonding configurations of NbZnSnO channels with and without MEA additives that were deposited on a KBr substrate. The current–voltage characteristics of the NbZnSnO TFTs were measured under air ambient in a dark box using an Agilent 4156 C semiconductor parameter analyzer.

3. Results and discussion

To determine the thickness of NbZnSnO films with and without adding monoethanolamine (MEA), the FIB-prepared thin samples were measured by transmission electron microscope (TEM). Fig. 1 presents the cross-section TEM images of solution-processed NbZnSnO films with different amounts of MEA (we designated it as NNZTO in Fig. 1). From Fig. 1(a)–1(c), the thickness of NbZnSnO films without and with adding MEA is around 5–7 nm. The crystalline characteristics of all NbZnSnO films were performed by glancing incidence angles x-ray diffraction (GIAXRD). 10-layers NbZnSnO films were fabricated to obtain sufficient pattern intensity. All NbZnSnO films, no matter with or without adding MEA, are amorphous, as shown in Fig. 1(d). A diffraction peak at around 54° is due to the Si(100) plane of the Si substrate.

The main idea of this study is to fabricate nitrogen-doped NbZnSnO films by using the sol-gel method and utilizing MEA additives. Therefore, the chemical composition and the chemical state of the NbZnSnO films with various ratios of MEA/Nb, prepared from precursor solution under the aging time of 20 h, were performed with X-Ray Photoelectron Spectroscopy (XPS). In order to avoid the surface contamination, sputter cleaning with 1 keV Ar

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