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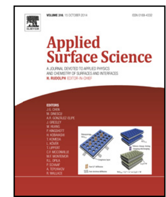

Investigation of H_2/CH_4 mixed gas plasma post-etching process for ZnO:B front contacts grown by LP-MOCVD method in silicon-based thin-film solar cells

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ABSTRACT

A new plasma post-etching method, H_2/CH_4 mixed gas plasma, is introduced to modify ZnO:B films grown by LP-MOCVD technique, successfully relaxing the double trade-offs, i.e., transparency/conductivity trade-off and surface texture/V_{oc} and FF trade-off. To deeply evaluate the post-etching process, optical emission spectroscopy technique is applied to diagnose the plasma condition. Upon different etching power, three distinct possible etching mechanisms are identified by analyzing the evolution of H_α^*, H_β^*, CH^* emission species in the plasma space. It is demonstrated that H_β^* and CH^* species are responsible for the physical etching process and chemical etching process, respectively, from which a new "soft" surface morphology is formed with a combination of micro- and nano-sized texture. Additionally, H_α^* species can bond with ZnO and also passivate the grains boundaries, thereby making both the carrier concentration and hall mobility increase. This process is defined as chemical bonding process. Finally, pin-type a-Si:H single-junction solar cells with an optimized device structure is grown on the etched ZnO:B substrate. The corresponding electrical parameters, such as J_{sc}, V_{oc} and FF, are simultaneously improved compared with the solar cell deposited on as-grown ZnO:B substrate with the same fabrication process. As a consequence, a noteworthy 8.85% conversion-efficiency is achieved with an absorber layer thickness only 160 nm.

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1. Introduction

Silicon-based multijunction thin-film solar cells, a well-established technology based on the use of cheap, abundant, and non-toxic raw materials, are a promising candidate for the future large-area photovoltaic systems operating in the gigawatt scale [1–4]. A significant issue for such photoelectric device is to keep the absorber layer (amorphous silicon or microcrystalline silicon) as thin as possible aiming to both reduce the production time and lessen the impact of light induced degradation [5]. Based on this consideration, advanced light trapping, making the solar cell "optical thick" despite the limited physical thickness, has gained tremendous interest [6,7]. Transparent conductive oxide (TCO) having a textured surface, commonly used as front contacts, is extensively applied to couple the incident light into the absorber layer, resulting in an increased photo-generated current [8,9]. Of the various TCO candidates, boron doped zinc oxide (ZnO:B) films grown by low pressure metal–organic chemical vapor deposition (LP-MOCVD) method has drawn great attention because of its self-textured surface, i.e., pyramidal features, which can demonstrate excellent light scattering capability [10–12]. Unfortunately, similar as other TCOs, ZnO:B films is still trapped in the constraints of inherent transparency/conductivity trade-off. As it is the first layer of solar cells crossed by the incoming light, its transparency is fundamental, meanwhile, it also has to be adequately conductive to extract carriers without significant resistive losses. As conductivity is improved, transparency tends to decrease requiring a compromise to be found [13]. In addition, although the steeply V-shape surface texture formed by the adjoining pyramidal grains can realize the strong light harvesting, it, however, can promote the formation of voids (shunts) and microcracks (ore pores) in the absorber layer, which can act as local current drains [14] and thereby cause significant deterioration in the open-circuit voltage (V_{oc}) and fill factor (FF) compared with the flat substrates [15,16]. Additionally, the absorber layer with porous structures can suffer

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from fast oxidation, thereby further weakening the device stability [17]. Here, the relationship between the surface texture and the performance of solar cells can be defined as another trade-off, i.e., surface texture/V_{oc} and FF trade-off. Therefore, in order to achieve a higher conversion-efficiency of solar cells grown on ZnO:B substrates, the above-mentioned double trade-offs need to be relaxed simultaneously.

Some post-etching method, which can be summarized into two categories including wet etching (acid etching) and dry etching (mainly plasma etching), has already been proposed aiming to resolve such problems. However, ZnO:B films grown by LP-MOCVD technique are very sensitive to acid solution, thus limiting the application of wet etching. On the other hand, although special attention has been focused on the plasma dry etching method, the reported results show that it is very difficult to balance the double trade-offs at the same time, e.g., hydrogen gas plasma indeed can effectively relax the transparency/conductivity trade-off [18], but can not eliminate the negative effect of V-sharp surface texture on the performance of solar cell. Argon gas plasma can tailor the surface texture from V-shape to "soft" U-shape through a long-time etching process, which is apparently beneficial for the recovery of V_{oc} and FF [19], but the light trapping capability and also the sheet resistance inevitably become worse. Very recently, we propose a new plasma post-etching technique, H_2/CH_4 mixed gas plasma, successfully relaxing the double trade-offs. Not only a novel surface morphology consisting of micro- and nano-sized textures is formed, but also both the transparency and conductivity are improved [20].

In order to correlate the formation of new surface morphology, together with the optical and electrical properties, of ZnO:B films to the plasma condition of H_2/CH_4 mixed gas, in this contribution, we introduce the optical emission spectroscopy (OES), an easy-to-implement process monitoring tool for plasma space, to collect reliable evidences to shed light on the possible plasma etching mechanisms. Upon different etching power, the evolution of different emission species in plasma space and the corresponding changes of surface texture/transparency/conductivity are detailed discussed. The possible etching mechanisms are summarized. Finally, hydrogenated amorphous silicon (a-Si:H) single-junction thin-film solar cells with an optimized device structure grown on ZnO:B front contacts (etched and non-etched) are used to check the application functions.

2. Experimental

ZnO:B films with a thickness of about 3 μm are deposited at low temperature (~160 °C) on the cleaned glass substrates with a size of 10 cm × 10 cm. Gas precursors employed are diethylzinc (DEZ) and water (H_2O) vapor with the flow rate of 11 and 16 sccm, respectively. Diborane (B_2H_6) diluted at 1% in argon is used as doping gas with a fixed flow rate of 16.5 sccm. The total growth pressure is kept at 0.5 Torr. The following H_2/CH_4 mixed gas plasma post-etching is performed in our in-house cluster plasma-enhanced chemical vapor deposition (PECVD) growth chamber with a changing etching power. The corresponding fixed flow rate of H_2 and CH_4 are set to 300 and 5 sccm, respectively. The etching pressure is kept at 0.6 Torr. Superstrate-type (p–i–n) a-Si:H single-junction thin-film solar cells grown on ZnO:B substrates (etched and non-etched) with a size of 0.25 cm^2 has the following structure: 11 nm p-type doped microcrystalline silicon oxide (p-μc-SiO:H)/10 nm intrinsic amorphous silicon oxide buffer layer (i-a-SiO:H)/160 nm intrinsic amorphous silicon (i-a-Si:H)/20 nm n-type doped amorphous silicon (n-a-Si:H)/500 nm aluminum back electrode. All the p- and n-type silicon-based layers are respectively doped by borontrimethyl ($B(CH_3)_3$, TMB) and phosphine (PH_3). Fig. 1 presents the corresponding schematic structure.

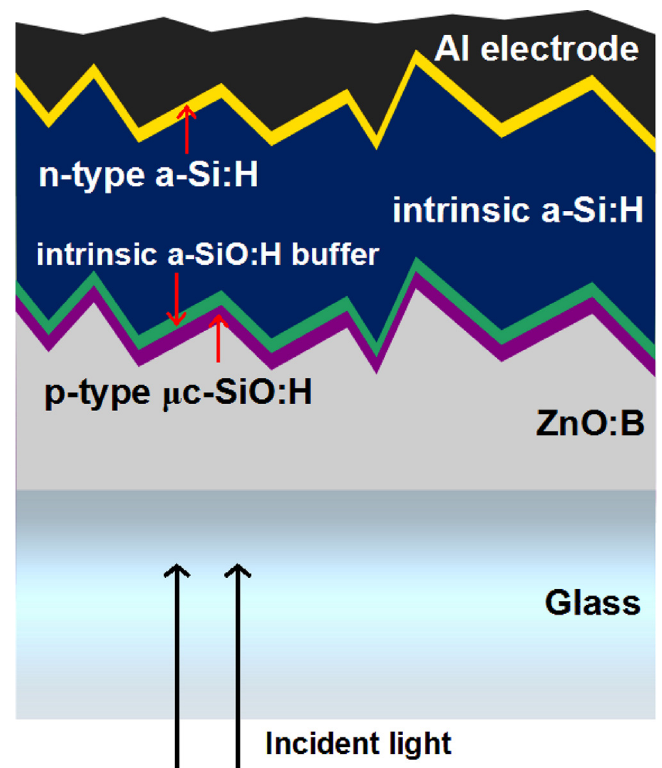

Fig. 1. Schematic structure of the a-Si:H solar cell architecture in superstrate (p–i–n) configuration. Only 500-nm thick aluminum reflector was used.

The electrical properties of ZnO:B, including carrier concentration and hall mobility (μ_{Hall}), are characterized by hall effect measurement. The surface morphology is deduced from atomic force microscopy (AFM). UV–vis/NIR spectrophotometer is employed to evaluate the total and diffuse optical transmittance (TT and DT) and, then haze factor defined as the light scattering capability, equal to DT/TT ratio, is calculated. In this contribution, Haze (600 nm) and Haze (1000 nm) are applied to represent the light scattering capability of short wavelength and long wavelength respectively. The sheet resistance is carried out by the four-probe method. OES is employed to in situ diagnose the plasma condition. A dual-lamp sun simulator is used to measure the light J–V curves of a-Si:H solar cells under standard test conditions (25 °C, AM1.5G, 1000 W/m^2). The external quantum efficiency (EQE) is introduced to analysis the spectral response of solar cells. Note that the J_{sc} mentioned in this contribution is calculated by EQE data.

3. Results and discussion

3.1. Possible etching mechanisms for H_2/CH_4 mixed gas plasma

It is well known that emission lines in an emission spectrum originate from the decay of electronically excited states of the atoms and molecules, i.e., the emission lines can be treated as the "fingerprint" of (excited) species [21–24]. Fig. 2 exhibits the schematic diagram of the OES measurement process. The optical emission spectra is measured through a quartz window. PR650 spectrometer is applied to detect the spectra in the range of 380–780 nm and SD-1000 spectrometer is used to record the wider spectra of 200–1000 nm but with lower sensitivity. Here, we focus on three emission species (H_α^*, H_β^*, CH^*) listed in Table 1, which are proved to be considerably correlated with the plasma etching behavior. Through analyzing such three emission species, some

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