Thin Solid Films 587 (2015) 126–131

Contents lists available at ScienceDirect

Thin Solid Films

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

Effects of cluster incorporation into hydrogenated amorphous silicon films in initial discharge phase on film stability

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ARTICLE INFO

Available online 25 February 2015

Keywords:
Plasma chemical vapor deposition
Solar cells
Light-induced degradation
Clusters
Amorphous silicon

ABSTRACT

We studied the effects of incorporation of hydrogenated amorphous silicon (a-Si:H) nanoparticles (clusters) generated in the initial discharge phase on light induced degradation of a-Si:H films. The amount of clusters incorporated into the films in the initial discharge phase is 15 times larger than that in the steady state. To evaluate the effects of such initial cluster incorporation on stability of a-Si:H films, we fabricated a-Si:H Schottky cells with and without initial cluster incorporation using a multi-hollow discharge plasma chemical vapor deposition method with a shutter and compared cell stability against light exposure. The degradation ratio of the cell without initial cluster incorporation is less than 1% even after 100 hour light soaking of 2.7 suns. Our results show that suppressing initial cluster incorporation into a-Si:H films is a key to stable a-Si:H cells. Moreover, Si–H_2 bonds in films can be reduced down to 1/10 using a cluster eliminating filter.

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1. Introduction

A-Si:H/μc-Si:H (hydrogenated microcrystalline silicon) tandem solar cell is one of the promising low-cost solar cells for large-scale power generation [1–4]. Light-induced degradation (LID) of a-Si:H films causes a significant reduction of the efficiency of the a-Si:H/μc-Si:H tandem cells [5–10]. Incident photons modify nanostructures in the a-Si:H films and generate the light-induced metastable defects. Several models have been proposed to understand generation mechanisms of the defects [11–13], but none of them has succeeded in showing the complete picture of the degradation. Some methods have been developed to measure the nanostructure in the films such as electron spin resonance (ESR) spectroscopy [14,15], ellipsometry [16,17] and doppler broadening positron annihilation spectroscopy [18–20]. Besides these methods, the most popular method is Fourier transform infrared (FTIR) spectroscopy which provides information on configurations of bonds such as Si–H and Si–H_2 bonds in films [21]. Using the FTIR spectroscopy, Matsuda et al. have pointed out that a-Si:H films with the lower density of Si–H_2 bonds show higher stability [22]. They have developed a triode discharge method to reduce the Si–H_2 bond density in the films [23,24]. Single-junction a-Si:H pin solar cells with an efficiency of over 10% have been fabricated by using the triode discharge method, while their light-induced degradation still remains 10% [25].

In SiH_4 discharges employed for a-Si:H deposition, there are three size groups of deposition species: SiH_x ($x \leq 3$) radicals, higher order silane (HOS) molecules Si_mH_n ($m \leq 4$), and a-Si:H particles in a size range below 10 nm (clusters) [26–30]. SiH_3 radicals are main deposition precursors, which dominate the film deposition rate. HOS molecules are generated due to polymerization between the SiH_2 radical and SiH_4 molecule. Clusters are nucleated from HOS molecules of Si_nH_x ($n \sim 4$) and grow due to coagulation between two clusters and deposition of SiH_x radicals to the surface of the clusters [26]. In a-Si:H film deposition, clusters are amorphous. HOS radicals, which are generated from HOS molecules due to electron collision, and clusters are minor deposition precursors and are pointed out to lead to the light-induced degradation [31,32]. Clusters and SiH_3 radicals contribute to the formation of SiH_2 bonds in films, whereas HOS radicals contribute little [33]. We have previously reported that suppression of clusters incorporation into films is the key to realizing highly stable a-Si:H films [34]. Based on the results, we have deposited highly stable a-Si:H films of 4.7×10^{15} cm^{-3} in stabilized defect density by using a multi-hollow discharge plasma chemical vapor deposition (CVD) method, by which the volume fraction of clusters can be reduced significantly [35,36]. Although ESR measurements show high stability of our bulk a-Si:H films of 0.6 μm thickness, Schottky cells using such "stable a:Si:H films" still show modest light-induced degradation. We also have developed a real-time monitor of the cluster volume fraction by employing three quartz crystal microbalances (QCMs) [37,38]. The real time measurements revealed large amount of cluster deposit in the initial discharge phase even in the multi-hollow discharge plasma CVD method [38].

Here, we have studied the effects of the incorporation of cluster generated in initial discharge phase into films on the stability of Schottky cells using the multi-hollow discharge plasma CVD method

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http://dx.doi.org/10.1016/j.tsf.2015.02.052
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equipped with a shutter to overcome the light-induced degradation in cell level. First, we measured time evolution of amount of cluster incorporated into film obtained using the QCMs to compare the amount of cluster incorporation without the shutter to that with the shutter. Then, we have evaluated the effects of a cluster eliminating filter to reduce Si–H_2 bonds in films. Finally, we fabricated Schottky cells with or without initial cluster incorporation to evaluate the effects of such initial cluster incorporation on the stability of a-Si:H cells.

2. Experimental details

Measurements of amount of cluster incorporation into films were carried out using a multi-hollow discharge plasma CVD reactor equipped with three QCMs as shown in Fig. 1 [37,38]. Three electrodes of 70 mm in diameter having 16 holes of 5 mm in diameter were placed in a stainless steel reactor. The holes were arranged in a lattice pattern. Gas of SiH_4 was supplied from a gas inlet and was pumped out through the electrodes with a turbo molecular pump. The flow rates of SiH_4 and H_2 were 30 sccm and 120 sccm, respectively. The total pressure was 66.5 Pa. Discharges were mainly sustained in the holes by applying a 60 MHz high frequency voltage to the powered electrode. The discharge power was set between 10 W and 90 W. Both SiH_3 radicals and clusters are generated in the discharges. Most clusters are transported to the downstream region because their diffusion velocity is less than the gas velocity. On the other hand, SiH_3 radicals are transported both toward the upstream region and the downstream region due to their fast diffusion velocity [39]. Therefore, incorporation of clusters into films deposited in the upstream region can be significantly suppressed compared with that in the downstream region.

To obtain information of a volume fraction of clusters V_f incorporated into a-Si:H films, three QCMs were placed 9 mm below the electrode in the upstream region of the multi-hollow discharge plasma reactor. The response of the QCM follows the Sauerbrey equation [40], in which the change in resonance frequency is inversely proportional to the mass change of films deposited on the QCM. Therefore the deposition rate of films was obtained from time dependence of the resonance frequency. Fig. 1 shows a schematic of QCMs for measurements of amount of cluster incorporation. The QCM of Channel A was used for measuring the deposition rate $DR_{w/}$ due to SiH_3 radicals and clusters. The microbalance of Channel B was applied to measure the deposition rate $DR_{w/o}$ due to SiH_3 radicals by using the cluster eliminating filter [41]. As shown in Fig. 2, the filter eliminates the contribution of the cluster deposition to films, because the sticking probability of the clusters and the surface reaction probability of the radicals are 100%

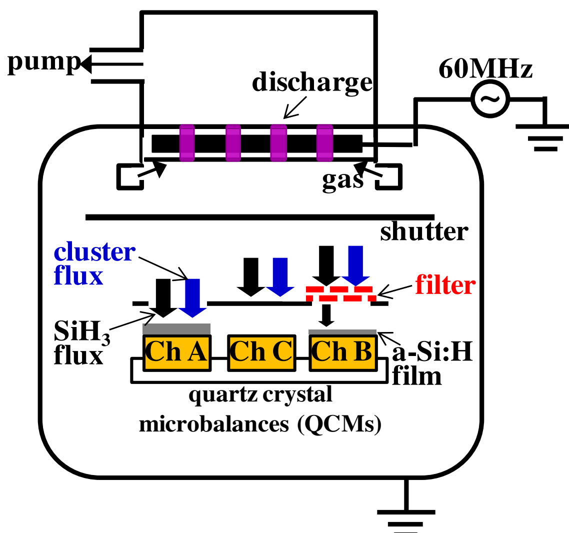

Fig. 1. Schematic of QCMs for measurements of amount of cluster incorporation into films.

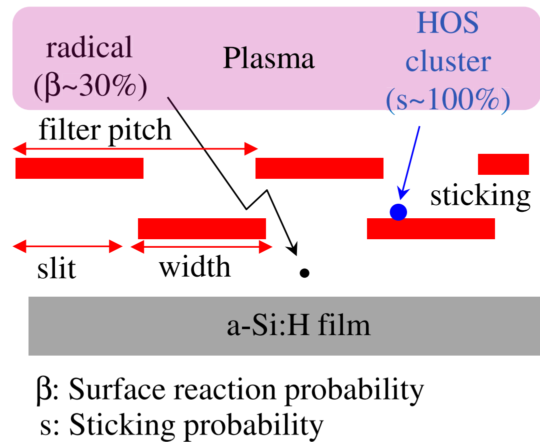

Fig. 2. Structure of cluster eliminating filter.

and 30%, respectively [41,42]. The QCM of Channel C was used as a reference sensor because the resonance frequency of quartz crystal is affected by experimental conditions such as temperature and pressure. The ratio of $DR_{w/}$ and $DR_{w/o}$ is given by

$$DR_{w/}/DR_{w/o} = 1/a\left(1-V_f\right) \quad (1)$$

where α is a coefficient of radical transmittance of the filter. In this paper, we employed the ratio $R = DR_{w/}/DR_{w/o}$ as an indicator of the amount of clusters incorporated into films. The temperature of the QCM was kept at 100 °C. In the measurement, the resolution of R was 1.

For deposition experiments, we employed another multi-hollow discharge plasma CVD reactor as shown in Fig. 3. Three electrodes of 35 mm in diameter having 8 holes of 5 mm in diameter were placed in a stainless steel tube of 38 mm in inner diameter. The flow rate of SiH_4 was 150 sccm and the total pressure was 66.5 Pa. Discharges were mainly sustained in the holes by applying a 60 MHz high frequency voltage to the powered electrode. The discharge power was 8 W. Substrates were located at 10 mm from the grounded electrode in the downstream side. The discharges were mainly sustained in the holes of the electrode, thus the ion bombardment from discharges to the substrates can be negligible.

To obtain information of Si–H_2 bond density in a-Si:H films, films were prepared on crystalline Si (111) wafers with a high resistivity (1000–5000 Ω cm) at a substrate temperature of 250 °C. An absorption intensity profile of a-Si:H film in a range of 1750–2350 cm^{-1} was measured with a FTIR spectroscope (JASCO FT/IR-620), and then it was deconvoluted into two gaussian profiles: one had a peak at

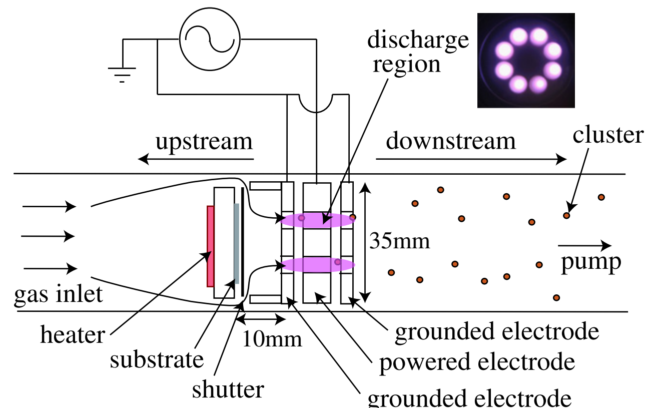

Fig. 3. Multi-hollow discharge plasma CVD reactor for Schottky cell fabrication.

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