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Reduction of surface recombination velocity by rapid thermal annealing of p-Si passivated by catalytic-chemical vapor deposited alumina films

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ABSTRACT

Excellent electrical-passivation of p-type Si (p-Si) in Si solar cells has been achieved by post-deposition rapid annealing of aluminum oxide (AlO_x) films prepared by catalytic chemical vapor deposition (Cat-CVD) using trimethyl aluminum (TMA) and O_2. Extremely small surface recombination velocity of below 0.1 cm/s has been obtained at post-deposition annealing temperatures in the range of 350–400 °C for an annealing time of 2 min. The reduction of surface recombination velocity has been attributed to band bending induced by a fixed negative charge density of 5 × 1011 charges/cm^2 and an additional small interface trapping density of around 10^{10} cm^{-2} eV^{-1}.

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1. Introduction

Electrical passivation of p-Si surfaces to reduce surface recombination in crystalline Si solar cells is very important for increasing their conversion efficiency. It is becoming all the more important as solar cells are becoming thinner in order to reduce their cost [1,2]. Recently, there have been reports on Alumina (AlO_x) film-passivation techniques which can reduce surface recombination velocity (S_0) mainly by dint of band bending effect attributed to fixed charges induced by AlO_x films deposited on Si [3]. The creation of fixed charges is attributed to precursor gases, annealing of AlO_x films, and method depositing the films. Fixed charges have been created in AlO_x films deposited on Si using the following deposition methods at various annealing temperatures, and annealing times: remote plasma enhanced chemical vapor deposition of tri-ethyldyaluminum and tri-sec-butoxide (400 °C, 30 min) [4], pyrolysis of aluminum tri-isopropoxide (510 °C, 15 min) [5], atomic layer deposition (ALD) of TMA and H_2O (300 °C) [6], plasma-assisted atomic layer deposition (PA-ALD) of TMA and O_2 (425 °C, 30 min) [7–9], plasma-enhanced chemical vapor deposition (PE-CVD) of TMA, CO_2 and H_2 (around 400 °C) [10], sputtering of an aluminum target with O_2 ambient 500 °C, 30 min) [11], spatial atomic layer deposition of TMA and H_2O (350–425 °C, 15 min) [12], and Cat-CVD of TMA and O_2 (from 230 °C, cooled for 300 min) [13]. The surface recombination velocity obtained has been very small in all deposition methods mentioned above. It has been reported that post-deposition annealing is very effective in reducing surface recombination velocity [14,15]. We have already obtained extremely small surface recombination velocity (S_0) of below 0.1 cm/s in p-Si passivated by Cat-CVD deposited alumina films which have been cooled down slowly for several hours in a vacuum [13]. The extremely small surface recombination velocity of below 0.1 cm/s in these films has been kept intact for over one year [13]. However, the duration of the thermal treatment of several hours is too long for industrial applications.

In this study, the post-deposition annealing process has been investigated to achieve a decrease of a surface recombination velocity by dint of the AlO_x film passivation. Film samples have been prepared by Cat-CVD deposition at varying annealing temperature (T_a) and time (t_a). The surface recombination at the interface between the AlOx film and Si surface has been characterized by photoconductivity decay (PCD) curves and evaluated also by direct measurement of surface recombination velocities (S_0). Furthermore, fixed charge density (N_f) and interface trapping densities (D_{it}) have been obtained from capacitance voltage (C–V) curves measured for metal-oxide-semiconductor (MOS) diodes fabricated using AlO_x films. The annealing temperature and time which achieve the smallest surface recombination velocity due to the AlO_x passivation films have been determined from PCD curves, measured surface recombination velocities, and fixed charge density. The mechanism for reduction of surface recombination velocity has been studied in terms of the behaviors of measured fixed charge density and interface trapping density.

2. Experimental

Aluminum oxide films were deposited on p-Si wafers by Cat-CVD using TMA and O_2 as the precursor gas. The experimental apparatus used in this study is shown in Fig. 1. Si substrates used for the samples were Czochralski (CZ) grown crystalline p-Si (100) wafers having a resistivity of 10 Ωcm with a mirror-polished front surface and a chemically-polished back surface. 2 × 2 cm^2 samples were scribed

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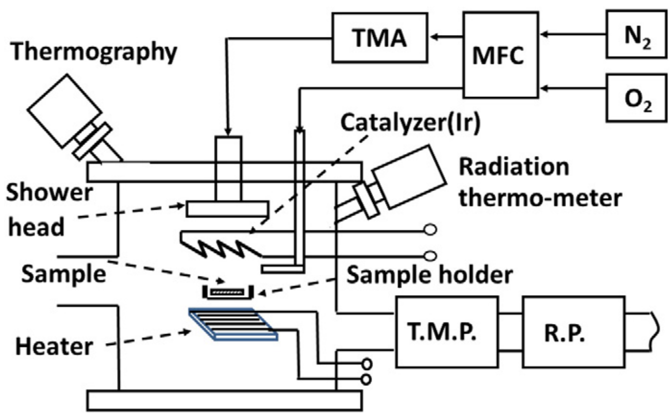

Fig. 1. Schematic illustration of apparatus used for film deposition and post-deposition rapid thermal annealing.

from the wafers. The samples were then slightly etched by a 2.5 wt.% hydrofluoric acid (HF) rinse to remove the natural oxide to obtain an H-terminated surface, and rinsed in deionized (DI) water prior to deposition. The samples were mounted on the sample holder 7 cm away from the catalyzer. The samples were heated from the back side by radiation from a Chromel alloy-wire heater mounted in a zigzag manner on a ceramic plate as shown in Fig. 1. The sample temperature (T_{sub}) was measured by thermography. An oxidation resistant 25 cm long, a 0.2 mm diameter iridium wire was used as the catalyzer. The wire was wound around the holder in a W-shaped bend, and the catalyzer temperature (T_{cat}) was measured by a radiation thermometer. The deposition of AlO_x films on the p-Si samples were carried out using N_2 gas as a carrier gas for the TMA which was introduced through the shower head and O_2 gas was independently blown around the catalyzer.

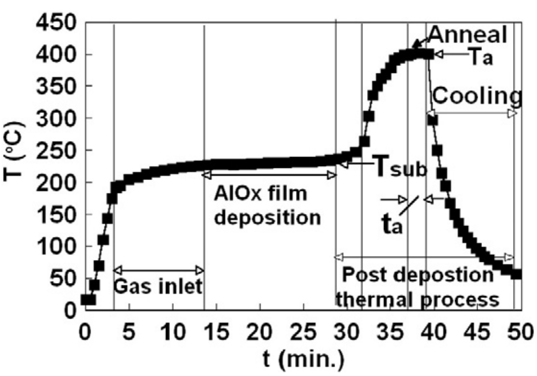

Fig. 2. Time chart used for film deposition and post-deposition thermal annealing.

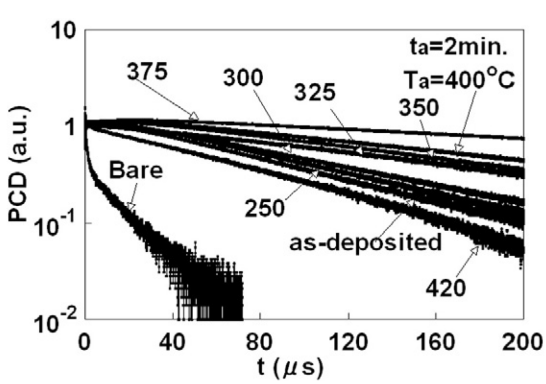

Fig. 3. Photoconductivity (PCD) curves observed by BS-τ for AlO_x/p-Si samples prepared by varying post-deposition annealing temperature, as-deposited and bare wafers (without AlO_x films).

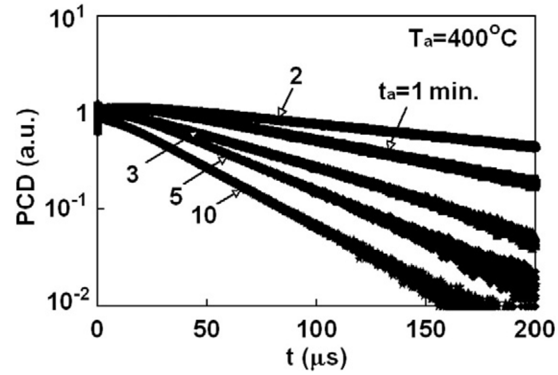

Fig. 4. PCD curves observed by BS-τ for AlO_x/p-Si samples prepared by varying post-deposition annealing time.

The films were deposited for 15 min at T_{sub} of 240 °C under O_2/TMA gas flow-rate ratio of 15 and a chamber pressure of 17 Pa. The flow rate of TMA was determined using the equivalent N_2 flow rate to satisfy the vapor-pressure of TMA. Subsequent to deposition, as seen in Fig. 2, the samples were annealed by elevating the sample temperature to the maximum annealing temperature (T_a) which was kept constant for the annealing time (t_a) of several minutes in N_2 ambient at a pressure of 10^5 Pa. The samples were prepared varying T_a in the range of 250–420 °C for t_a = 2 min, and then varying t_a in the range of 1–10 min with T_a = 400 °C. The AlO_x film thicknesses of the samples were measured to be in the range of 2.1–3.1 nm by a spectroscopic ellipsometer (ALPHA-SE, J.A. Woolam Co., Inc.) using two-layer model composed of an AlOx layer and a SiO_2 layer. The surface recombination at the interface in AlO_x/p-Si was characterized by the initial decay in photoconductivity decay curves measured and evaluated also by the surface recombination velocity (S_0) measured using contactless techniques [16–19]. In this experiment, the photoconductivity change was detected by a reflected 500 MHz electromagnetic wave. Excess carriers were created by impulse-irradiation of 904 nm laser diode with a photon flux density of 7.7×10^{12} cm^{-2}. Surface recombination velocities in the samples were directly determined by applying bi-surface photoconductivity decay (BS-PCD) method to two PCD curves measured by a BS-τ lifetime profiler equipment from Hemmi Inc. [16–19]. Fixed charge density (N_f) was obtained from the flat-band voltage shift of C–V curves measured at 1 MHz. Interface trapping density (D_{it}) was measured varying the signal frequency in the range of 10 Hz–1 MHz using the conductance method [20] with a signal amplitude of 20 mV for MIS diodes consisting of the AlO_x insulator film and an aluminum-gate electrode of 500 μm in diameter.

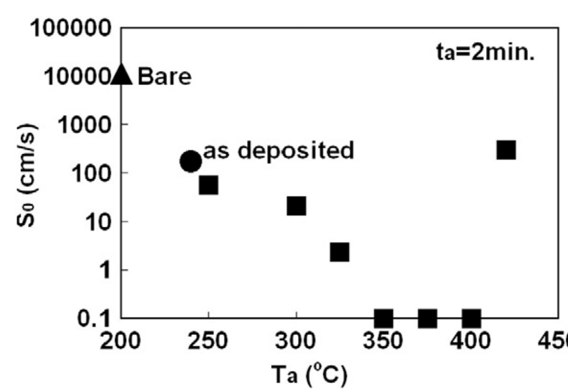

Fig. 5. Surface recombination velocity (S_0) measured as a function of post-deposition annealing temperature (T_a).

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