

A Novel Atomic Layer Deposited Al_2O_3 Film with Diluted NH_4OH for High-Efficient c-Si Solar Cell

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Abstract—In this paper, Al_2O_3 film deposited by thermal atomic layer deposition (ALD) with diluted NH_4OH instead of H_2O was suggested for passivation layer and anti-reflection (AR) coating of the p-type crystalline Si (c-Si) solar cell application. It was confirmed that the deposition rate and refractive index of Al_2O_3 film was proportional to the NH_4OH concentration. Al_2O_3 film deposited with 5 % NH_4OH has the greatest negative fixed oxide charge density (Q_f), which can be explained by aluminum vacancies (V_{Al}) or oxygen interstitials (O_i) under O-rich condition. Al_2O_3 film deposited with NH_4OH 5 % condition also shows lower interface trap density (D_{it}) distribution than those of other conditions. At NH_4OH 5 % condition, moreover, Al_2O_3 film shows the highest excess carrier lifetime (τ_{PCD}) and the lowest surface recombination velocity (S_{eff}), which are linked with its passivation properties. The proposed Al_2O_3 film deposited with diluted NH_4OH is very promising for passivation layer and AR coating of the p-type c-Si solar cell.

Index Terms—Solar cell, Al_2O_3 , NH_4OH , passivation layer, anti-reflection coating

I. INTRODUCTION

In the field of c-Si solar cells, the reduction of recombination losses at the surface of c-Si has become increasingly important in order to improve its power conversion efficiency [1-3]. In order to mitigate recombination losses at the surface of c-Si, many kinds of materials had been used [1-4]. In high-efficiency laboratory cells, traditionally, thermal SiO_2 grown in a high-temperature (≥ 900 °C) oxidation process have been applied as passivation layer with excellent chemical passivation property. However, due to the degradation of the c-Si bulk lifetime at high-temperature, thermal SiO_2 cannot be applied to the industrial solar cell processes. Hence, for industrial high-efficiency c-Si solar cells, alternative passivation layers are required, which can be synthesized at low temperature.

One of the alternative passivation layer is amorphous SiN_x (a- SiN_x) synthesized by plasma-enhanced chemical vapor deposition (PECVD) at ~ 400 °C. However, at the rear of passivated emitter and rear cell (PERC)-type solar cell, it can cause ‘parasitic shunting’ which can lead to a loss in the short-circuit current density due to its high positive Q_f . a-Si deposited by PECVD at 200~250 °C is also considered as alternative passivation layer for c-Si solar cell. In the case of PERC-type solar cell passivated with a-Si at rear, no parasitic shunting occurs. However, it is reported that a-Si is highly sensitive to thermal processes.

Recently, a- Al_2O_3 synthesized by ALD has attracted strong interest as a promising passivation layer on lowly and highly doped p-type c-Si due to its high level of negative Q_f [1-5]. a- Al_2O_3 film can provide not only

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chemical passivation but also outstanding field-effect passivation on p-type c-Si surfaces. Moreover, a-Al₂O₃ has some outstanding properties such as good thermal and chemical stability, and good adhesion to various surfaces [6]. In order to improve the light collection and the stability of Al₂O₃ passivation layer, a-SiN_x film is usually deposited above Al₂O₃ film by PECVD [3]. Due to the optical properties which can be altered with wide window, a-SiN_x is the standard for AR coating in solar cell. Moreover, N-rich a-SiN_x shows the excellent thermal and chemical stability.

In this work, diluted NH₄OH was used instead of H₂O to deposit Al₂O₃ film by thermal ALD. Electrical and material properties of Al₂O₃ film were studied as a function of the NH₄OH concentration to confirm the applicability for not only passivation layer but also for AR coating as a substitute for Al₂O₃/SiN_x stack.

II. EXPERIMENTAL

The experimental process flow for this work is summarized in Fig. 1. A p-type Si substrate was treated with RCA cleaning. Al₂O₃ film (10 nm) was deposited by thermal ALD at 250 °C with trimethylaluminum (TMA) and diluted NH₄OH (0~10 %). Ti film (100 nm) was deposited as a top electrode on the Al₂O₃ film by RF magnetron sputter. After patterning the top electrode by photolithography and wet etching, Al film (100 nm) was

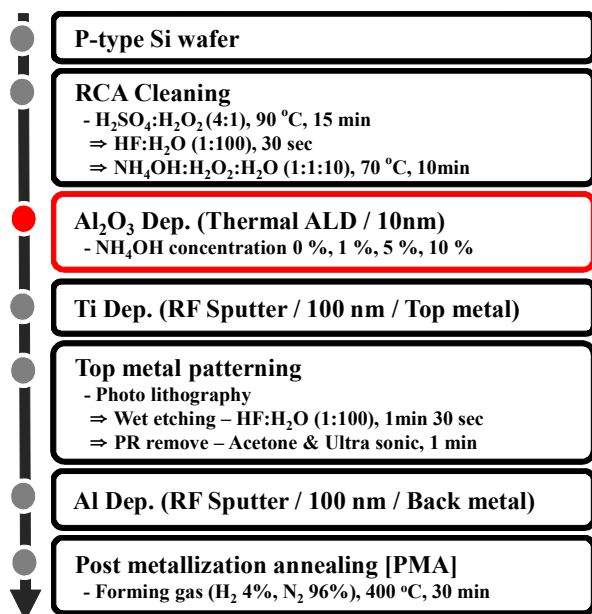


Fig. 1. Process flow for the experiments.

deposited on the backside of the substrate by RF magnetron sputter. Finally, post-metallization annealing (PMA) was carried out in a furnace with forming gas ambient at 400 °C for 30 min.

Deposition rate and refractive index of the Al₂O₃ film deposited at different NH₄OH concentration were confirmed with Ellipsometry measurement. High frequency capacitance (C_{HF}) was measured by an Agilent 4284A precision LCR meter. Quasi-static capacitance (C_{QS}) and leakage current were measured by an Agilent 4156A semiconductor parameter analyzer. Al₂O₃ film deposited with diluted NH₄OH with different concentration was analyzed by X-ray photoelectron spectroscopy (XPS) to find out the origin of the negative fixed charge, Q_f. To confirm the passivation properties of Al₂O₃ film deposited at different NH₄OH concentration, Quasi-Steady-State Photoconductance (QSSPC) measurement was carried out.

III. RESULTS AND DISCUSSIONS

Before the fabrication of Al₂O₃ MIS (Metal-Insulator-Semiconductor) capacitor, deposition rate and refractive index of Al₂O₃ film were confirmed by Ellipsometry measurement as shown in Fig. 2. Deposition rate is proportional to the concentration of diluted NH₄OH, which might be attributed to an increased density of hydroxyl (-OH) surface groups which is supplied from diluted NH₄OH. It was reported that -OH surface groups are essential for high-quality Al₂O₃ film growth in H₂O-based ALD process [7]. Refractive index was also increased from 1.76 to 2.00 in the wavelength range of 400~800 nm with the increasing of the NH₄OH concentration. Higher refractive index of the AR coating means that a relatively lower critical thickness is needed for the minimum reflection as explained in Eq. (1) [8]:

$$d_j = \frac{\lambda_0}{4 n_j} \quad (1)$$

where λ_0 is the light wavelength in the air, n_j and d_j are the refractive index and the critical thickness of the coating material, respectively, and j is equal to 1 for single layer AR coating.

Due to the increase of deposition rate and the decrease of the critical thickness for the minimum reflection,

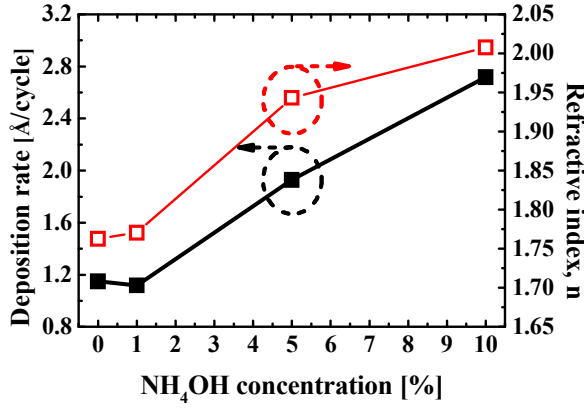
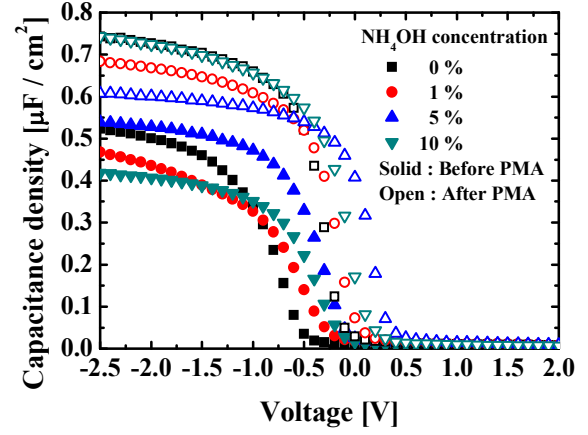


Fig. 2. Deposition rate and refractive index of Al₂O₃ film as a function of NH₄OH concentration.

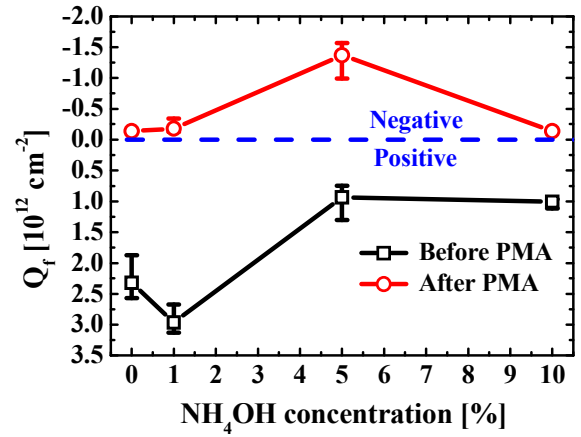
proposed Al₂O₃ film can be applied as AR coating layer of c-Si solar cell. And the process efficiency of c-Si solar cell can be improved by using diluted NH₄OH due to increased deposition rate of Al₂O₃ film. Moreover, because the refractive index is directly linked to material density, it can be said that NH₄OH affects the density of Al₂O₃ film deposited by thermal ALD [9, 10].

Fig. 3(a) shows the C-V characteristics of an Al₂O₃ MIS capacitor at 1 MHz. The flatband voltage (V_{FB}) of NH₄OH 5% condition moved largely toward positive bias direction, but then shifted toward the negative bias at NH₄OH 10% condition before PMA. After PMA, the V_{FB} of all conditions shifted abruptly toward the positive bias region and showed the same trend with those before PMA. Fig. 3(b) shows the fixed oxide charge density, Q_f as a function of the concentration of NH₄OH. To extract Q_f , a reference V_{FB} (-0.53 V) calculated from the dependence of V_{FB} on Al₂O₃ thickness was used. Before PMA, Al₂O₃ film at all conditions showed positive Q_f in which the lowest positive Q_f was shown at NH₄OH 5% condition. After PMA, Q_f of Al₂O₃ film was negative for all conditions and it became the greatest at NH₄OH 5% condition. Therefore, NH₄OH 5% condition is desirable from the view point of negative Q_f .

To confirm the trend of interface trap density, D_{it} with the NH₄OH concentration, quasi-static C-V (QSCV) measurement was performed. From the QSCV curve, the surface potential as a function of voltage was extracted by Berglund's integral [11] and D_{it} was calculated with high-frequency capacitance (C_{HF}) and quasi-static capacitance (C_{QS}) from Eq. (2) [11]:



(a)



(b)

Fig. 3. (a) C-V characteristics of an Al₂O₃ MIS capacitor. (1 MHz), (b) Fixed oxide charge density, Q_f of an Al₂O₃ MIS capacitor as a function of NH₄OH concentration before and after PMA.

$$D_{it} = \frac{C_{QS} - C_{HF}}{q} \left(1 - \frac{C_{QS}}{C_{ox}} \right)^{-1} \left(1 - \frac{C_{HF}}{C_{ox}} \right)^{-1} \quad (2)$$

where C_{ox} is the oxide capacitance and q is electronic charge.

To investigate D_{it} as a function of surface potential, D_{it} was plotted as a function of $E-E_v$ as shown in Fig. 4. Al₂O₃ films with NH₄OH 5% condition showed the lowest D_{it} at midgap (0.56 eV) and comparable to the without NH₄OH condition.

To find out the origin of the negative Q_f in Al₂O₃ film deposited with NH₄OH, XPS analysis was carried out as shown in Fig. 5. The atomic percentage of the Al2p, O1s, and N1s peaks of the Al₂O₃ and the ratio of Al2p to O1s was calculated in Table 1. It is notable that nitrogen is

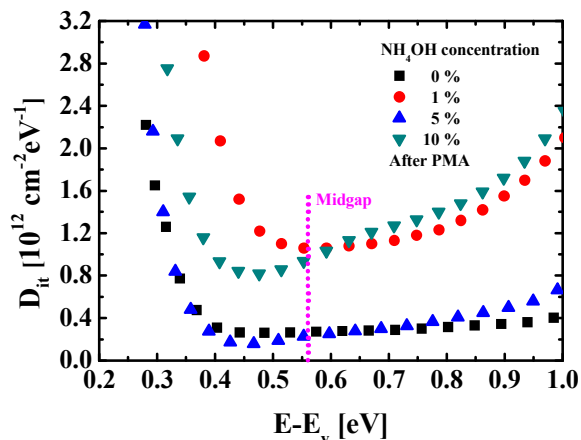


Fig. 4. The interface trap density, D_{it} as a function of surface potential after PMA.

rarely incorporated in the Al_2O_3 films despite the use of NH_4OH . Therefore, it can be said that any other material such as AlN_x or AlO_xN_y is not formed while Al_2O_3 film is synthesized by thermal ALD with diluted NH_4OH . In a Al_2O_3 , V_{Al} and O_i form oxygen dangling bonds (O DBs), which contribute to negative Q_f . Hence, if Al_2O_3 film becomes O-rich, it exhibits greater negative Q_f [12, 13]. Al_2O_3 film deposited with 5 % diluted NH_4OH has lower percentage of Al2p and higher percentage of O1s than those deposited with other concentration as in Table 1. That is, the least positive Q_f and the greatest negative Q_f before and after PMA with NH_4OH 5 % condition might be the result of great O DBs.

Fig. 6 shows the leakage current of Al_2O_3 MIS capacitor before and after PMA. As diluted NH_4OH was applied in Al_2O_3 deposition, the leakage current of Al_2O_3 MIS capacitor was decreased before PMA. Especially, Al_2O_3 film synthesized with 5 % NH_4OH showed the lowest leakage current. Decrease of leakage current of Al_2O_3 MIS capacitor means the improvement of bulk oxide quality [7] and/or the reduction of oxygen vacancies (V_{O}) which can introduce leakage current of Al_2O_3 film [14, 15]. From the results of Ellipsometry and XPS, it can be said that diluted NH_4OH improve Al_2O_3 quality and decrease of V_{O} by increasing the mass density and the oxygen concentration in the Al_2O_3 film. After PMA, leakage current of MIS capacitor with NH_4OH 0 and 5 % condition was decreased and that with NH_4OH 5 % condition exhibited the lowest leakage current. At NH_4OH 1 and 10 % condition, however, leakage current was increased and higher than other

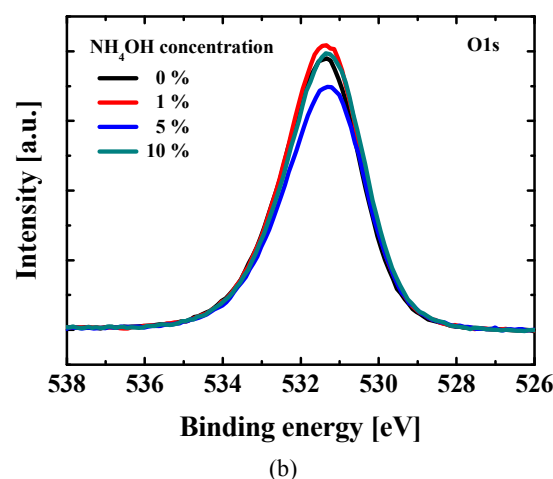
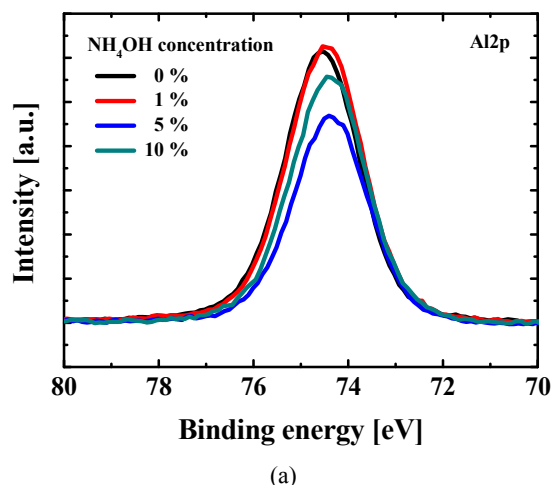


Fig. 5. XPS peaks of (a) Al2p, (b) O1s as a function of NH_4OH concentration.

Table 1. Atomic percent of Al2p, O1s, and N1s in as grown Al_2O_3 film as a function of NH_4OH concentration

NH_4OH	Al2p	O1s	N1s	Al2p/O1s ratio
0 %	35.69	54.14	-	0.66
1 %	35.25	54.38	0.32	0.65
5 %	30.51	56.51	0.31	0.54
10%	32.39	56.02	0.29	0.58

conditions. These higher leakage currents might be explained by great D_{it} distribution as shown in Fig. 4(b) which can increase leakage current of the Al_2O_3 film.

Fig. 7 exhibits the QSSPC measurement results before and after forming gas annealing (FGA). Surface recombination velocity, S_{eff} was extracted from Eq. (3) with excess carrier lifetime, τ_{PCD} which is the result of QSSPC measurement [16, 17]:

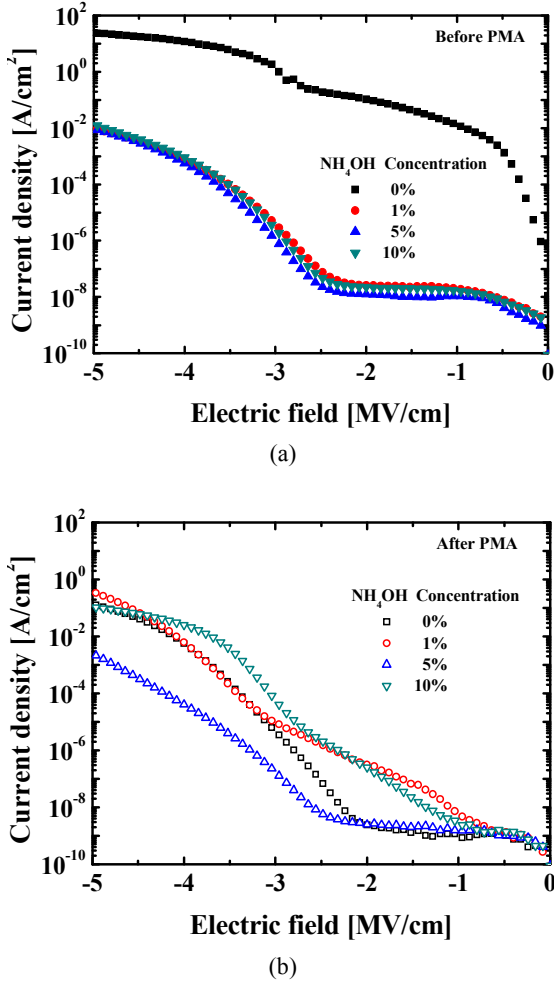


Fig. 6. Leakage current of an Al₂O₃ MIS capacitor (a) before, (b) after PMA.

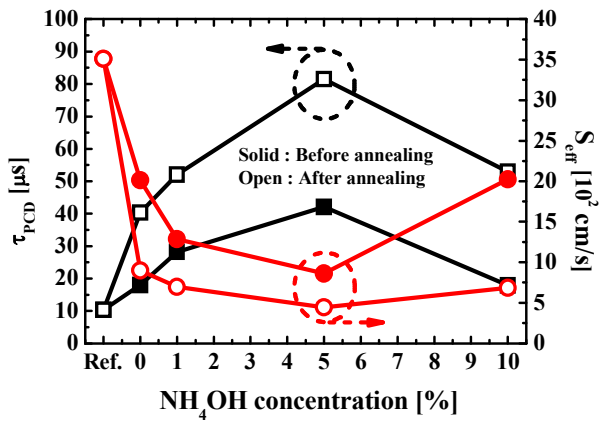


Fig. 7. Excess carrier lifetime, τ_{PCD} and surface recombination velocity, S_{eff} of Si wafers deposited with Al₂O₃ at different temperature conditions (Ref. : Bare wafer).

$$\frac{I}{\tau_{PCD}} = \frac{I}{\tau_b} + \frac{2S_{eff}}{W} \quad (3)$$

where W is the effective wafer thickness and τ_b is the bulk minority carrier lifetime which is usually assumed to be infinite as the best case.

The τ_{PCD} was increased and S_{eff} was decreased as the NH₄OH concentration increases up to NH₄OH 5 % condition both before and after FGA. Both the τ_{PCD} and S_{eff} was improved by FGA for all conditions. At NH₄OH 10 % condition, however, τ_{PCD} and S_{eff} became worse than NH₄OH 5 % condition. Because τ_{PCD} and S_{eff} of Si wafer are closely related to the passivation properties of Al₂O₃ film, it can be said that Al₂O₃ film deposited at NH₄OH 5 % condition with FGA has the best passivation effect, indicating the highest efficiency of c-Si solar cell, due to the outstanding chemical and field-effect passivation properties caused by not only the highest negative Q_f but also lower D_{it} distribution than those deposited at other conditions.

V. CONCLUSIONS

As the passivation layer and the AR coating of the p-type c-Si solar cell, the characteristics of Al₂O₃ film deposited by thermal ALD with diluted NH₄OH were studied in depth. Due to the increase of deposition rate and refractive index of Al₂O₃ film with the increase of the concentration of diluted NH₄OH, it is suitable for AR coating and can improve the process efficiency of c-Si solar cell. At NH₄OH 5 % condition, the Al₂O₃ film showed the outstanding chemical and field-effect passivation property in p-type c-Si due to not only the highest negative Q_f but also lower D_{it} distribution than those deposited at other conditions. The highest negative Q_f of Al₂O₃ film at NH₄OH 5 % condition is due to the greatest O-rich condition which may cause O DBs. From the result of Ellipsometry, XPS and leakage current measurement, diluted NH₄OH had an effect on the improvement of bulk oxide quality and/or the reduction of V_o of Al₂O₃ film which may induce the positive Q_f . The highest τ_{PCD} and lowest S_{eff} were appeared at NH₄OH 5 % condition. Therefore, Al₂O₃ film deposited with 5% NH₄OH shows the applicability as AR coating and the excellent field-effect passivation characteristic. In conclusion, it can be said that the Al₂O₃ film deposited

by thermal ALD with diluted NH_4OH is very promising for passivation layer and AR coating of p-type c-Si solar cell.

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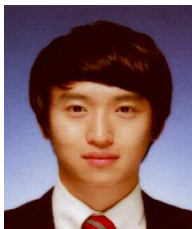
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