24P011-O

Radical Flux Evaluation to Microcrystalline Silicon Films Deposited by Multi-Hollow Discharge Plasma CVD

マルチホロー放電プラズマで作製した微結晶シリコン膜へのラジカルフラックス評価

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To evaluate radical fluxes for microcrystalline silicon film deposition, Si films were deposited in a combinatorial way using a multi-hollow discharge plasma CVD method. Film crystallinity varied with the distance from the powered electrode. Based on the film deposition rate and Raman crystallinity, we proposed a method to estimate hydrogen flux. This evaluation of hydrogen flux is one of useful way to understand a process window of μc-Si:H films.

1. Introduction

Microcrystalline silicon(μc-Si:H) films have attracted much attention as materials of the bottom-cells in Si thin film tandem photovoltaics [1]. However, thedeposition rate of μc-Si:H is very low due to the low SiH_4 density under relatively low partial pressure conditions using Plasma Enhanced Chemical Vapor Deposition (PECVD). Even though high pressure depletion regime has been proposed as an alternative deposition method to overcome above difficulties [2], a narrow process window which determined by H and Si radical flux makes it difficult to fabricate device quality microcrystalline silicon films.

In this study, Si films were deposited in a combinatorial way using a multi-hollow discharge plasma CVD method. We present a new approach to flux evaluation, by which we discuss a process window of μc-Si film deposition.

2. Experimental

A multi-hollow discharge plasma CVD reactor consists of one powered electrode and a pair of grounded electrodes to generate plasma as shown in Fig. 1[3].

The silane dilution ratio R=([H_2]+[SiH_4])/[SiH_4] was kept at 300. The total pressure and gas flow rate were sustained at 2Torr and 150sccm, respectively. The substrate holder was set vertically on the powered electrode to obtain information on spatial distribution of deposition radicals. The substrate temperatures (T_S) were varied from RT to 250^{o}C.Crystallinity (X_C) was determined with a Raman spectroscope (Jasco, NRS-3100). Deposition rate (R_d) was obtained as film thickness divided by deposition time.

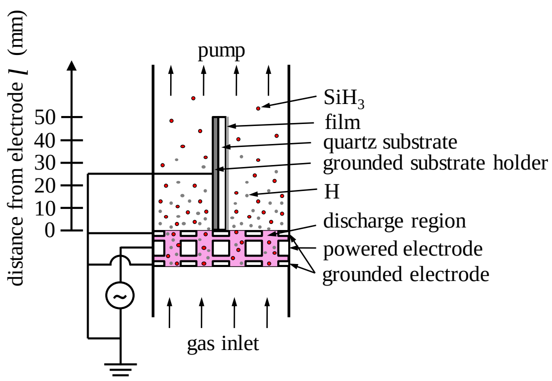

Fig.1. Schematic diagram of a multi-hollow discharge CVD reactor.

3. Experimental results and discussion

Figure 2 shows (a)R_d and (b)X_C as a function of the distance from the electrode l, respectively. The R_d of each film in Fig. 2 (a) decreases exponentially with l for l>5 mm because SiH_3 density, which is the main deposition precursor for high-quality films, decreases exponentially with l due to their loss at the surfaces of reactor wall. The X_C in Fig. 2 (b) is higher near the discharge region and drops sharply in the crystalline/amorphous transition region around l=20-40mm.

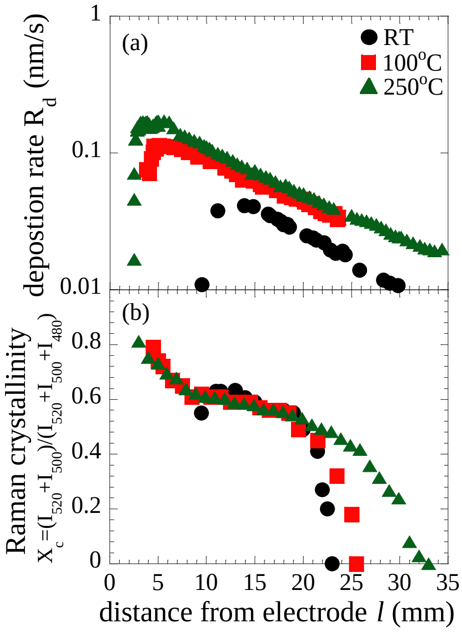

Fig.2. Dependence of (a) deposition rate and (b) crystallinity X_C on the distance from the electrode l.

To obtain further information on the contribution of each flux to film deposition near the discharge region, SiH_3 flux (Γ_{Si} = $nv_{th}/4$) was deduced using the following equation [4]:

$$R_d(SiH_3)=\left(\frac{M_{Si}}{N_A\rho_{Si}}\right)\left(\frac{nv_{th}}{4}\right)\left(\frac{\beta}{1-\beta/2}\right)\left(\frac{s}{\beta}\right) \quad (1)$$

where R_d is deposition rate, M_{Si} = 28.09 g/mol of the atom molar mass, N_A = 6.02×10^{23} /mol of the Avogadro number, and ρ_{Si} = 2.2 g/cm^3 the film specific gravity. The sticking probability s and the surface reaction probability β of SiH_3, being the predominant deposition precursor, were 0.1 and 0.28, respectively.

To determine Γ_{Si}, we considered R_d of only a-Si:H region, and extrapolated towards the μc-Si:H region as Γ_{Si} of μc-Si region as shown in Fig.3 (a) because the R_d of μc-Si region is determined by the balance between deposition due to Γ_{Si} and etching due to Γ_H whereas a-Si:H region is predominantly by deposition due to Γ_{Si}. Figure 3 (b) shows a flux ratio of H atoms to SiH_3 radicals estimated using the relationship between Γ_H/Γ_{Si} and X_c reported by S. Nunomura, et al.[5]. Figure 3 (c) shows l dependence of H atom flux estimated using Γ_{Si} in Fig. 3(a) and Γ_H/Γ_{Si} in Fig. 3(c). Γ_H decreases with increasing l and the gradient of Γ_H becomes sharper with decreasing T_S.

At high substrate temperature Γ_H is high near the electrode region resulting in widening the high crystallinity region. These results show that the increase of T_s is one possible way to obtain a wide process window of μc-Si:H films.

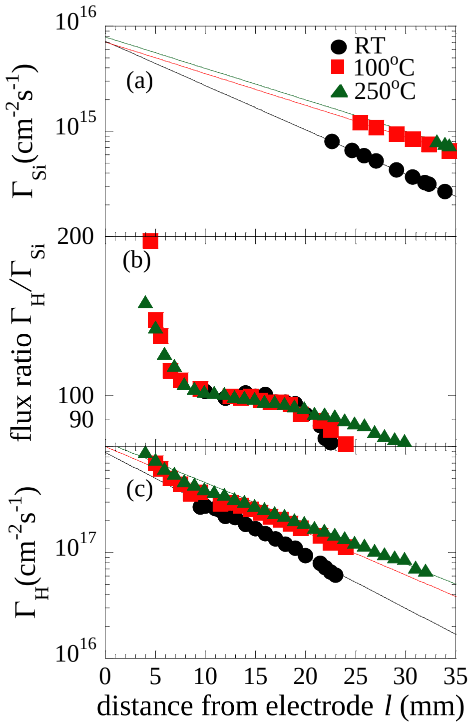

Fig.3. Dependence of (a)Γ_{Si},(b) Γ_H/Γ_{Si} and (c)Γ_H on the distance from the electrode l.

4. Conclusion

Si thin films were deposited in a combinatorial way using the multi-hollow discharge plasma CVD method. Based on the film deposition rate and Raman crystallinity, we proposed the method to estimate hydrogen flux. This evaluation of hydrogen flux is one of useful way to understand a process window of μc-Si:H films.

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