Study on Deposition Mechanism of SiOx Films Produced by Silicone Oil and Ozone Gas

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1. Introduction

Low temperature SiOx films have been widely used for gate insulator and passivation layer of, e.g., thin film transistors (TFTs). Previously, we proposed APCVD method for that, using silicone oil (S.O.) and ozone (O_3) gas. S.O. is much cheaper and safer material compared to TEOS (tetraethoxysilane). However, SiOx films produced by this method contain lots of residual -OH bonds, which degrades electrical insulation property severely.[1)] To reduce them, it is necessary to understand the deposition mechanism more in detail. In this meeting, we report the results to clarify and discuss them.

2. Experimental

N-type (111) oriented Si wafers were used as substrates to deposit SiOx films. The Si substrate was heated up to 200 ˚C. The S.O. solution was heated to 50 ˚C and bubbled by N_2 carrier gas with the flow rate from 70 to 330 ml/min, which was supplied with S.O. vapor to the reaction chamber. The O_3 was generated by ozone generator with ozone concentration (C_{O3}) from 100 to 220 g/m^3, and the O_2 flow rate (F_{O2}) to the generator was from 200 to 500 ml/min. Film thickness and refractive index were measured by the ellipsometry method. Chemical bonds and the -OH content were estimated by Fourier transform infrared spectroscopy (FT-IR) method.

3. Results

Figure 1 shows the dependences of deposition rate Rd (left) and Si-OH bond content (right) on the silicone oil flow rate $F_{S.O.}$ (S.O. flow rate). It can be seen that the Rd increases linearly with increasing $F_{S.O.}$ and is getting saturated for $F_{S.O.}$ more than ~0.4 ml/min. The linear increasing means that the deposition rate is limited or controlled by $F_{S.O.}$ supply, and the tendency of the saturation indicates the transition region from $F_{S.O.}$ supply control to the F_{O3} (ozone flow rate) one. Also, the OH integrated peak ratio increases with $F_{S.O.}$ from the lower end and gets saturated for $F_{S.O.}$ more than ~0.3 ml/min.

Figure 2 shows the results of the low (~950 cm^{-1}) wavenumber (LW) case of Fig. 1 and the high (~3300 cm^{-1}) wavenumber (HW) case, both of which are related with OH bonds. The OH contents in both HW and LW increase with Rd initially, probably because the dehydration reaction between the OH bonds on the precursors becomes more and more imperfect with increasing Rd. The OH content in the HW increases with Rd without saturation behavior, which is different from that of the LW. Since the peak integrated intensity can be considered as a kind of averaged density of OH content in the film, the density of broken Si-O-Si network maybe increase with the deposition rate or thickness initially but get saturated in the high deposition rate for the LW case while that of absorbed H_2O and/or neighbor OH maybe increase constantly within the experimental one for HW case.

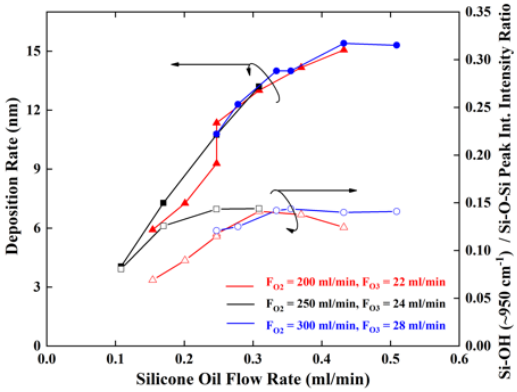

Figure 1. Silicone oil flow rate dependences of deposition rate and OH content.

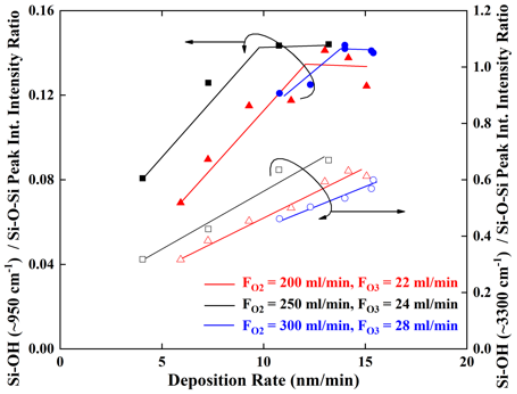

Figure 2. Deposition rate dependences of OH content.

4. Summary

Both deposition rate and OH content increase with $F_{S.O.}$ initially but get saturated in the high $F_{S.O.}$ Also, the OH content depends on the deposition rate. In this meeting, the details will be shown and discussed.

Reference: (1). S. Horita and P. Jain: Jpn. J. Appl. Phys. 57 (2018) 03DA02.