

Trichloroethene Concentration Effect on Deposition Rate and Properties of Low-Temperature Grown SiO₂ Films by using Silicone Oil and Ozone Gas

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Introduction: Low-temperature (< 300 °C) grown silicon oxide (SiO₂) films are used for many purposes [1]. Previously, we have reported about the low-temperature deposition of SiO₂ films using atmospheric pressure chemical vapor deposition (APCVD) with silicone oil (S.O.) and ozone (O₃) gas along with an organic solution [2]. Advantages of S.O. are lower cost and safer to use, compared with TEOS (tetraethylorthosilicate) which is conventionally used. Adding organic solution or trichloroethene (TCE), the deposition rate has increased [2]. In these films, non-negligible amount of unreacted Si-OH bonds remains as much as conventional ones [1, 2]. Changing the TCE concentration from 0.08 to 0.65 mol/m³, we investigated the concentration dependence of deposition rate and material properties, e.g., -OH content, of the deposited SiO₂ films. In this meeting, the results are reported and discussed.

Experimental Procedure: SiO₂ films were deposited on Si substrates, using silicone oil (decamethylcyclopentasiloxane: C₁₀H₃₀O₅Si₅), and ozone gas (O₃). The silicone oil was heated around 50 °C and the vapor gas was introduced into the reaction chamber by bubbling with N₂ gas. The flow rate of nitrogen gas into the silicone oil, i.e., N₂(S.O.) was 0.35 and 0.25 lm (litres per minute). O₃ was produced by electric discharge of 99.999% pure O₂ gas with the flow rate of 0.5 lm. TCE was bubbled by N₂ gas flow and the TCE gas was added during the deposition along with the S.O. and O₃. The N₂ gas flow rate for TCE was kept at 0.1 lm for almost all the depositions. The concentration of TCE in gas was varied by controlling temperature of a bath where TCE solution bottle was kept in, using temperature dependence of the saturation vapor pressure. The films were deposited for 10 minutes at 200 °C. The molecular structure and thickness of as-deposited films were estimated using FT-IR (Fourier Transform Infrared) spectrometer, and ellipsometry method, respectively.

Results and Discussion: Figure 1 shows the FT-IR spectra of the SiO₂ films grown at 200 °C for 10 minutes for TCE concentrations of ~0.16 and ~0.21 mol/m³ for N₂(S.O.) of 0.35 and 0.25 lm, respectively. In the FT-IR spectra, the peaks at 800 and 1070 cm⁻¹ appear due to the Si-O-Si bonding,

while the peaks at 960 and 3400 cm⁻¹ appear due to Si-OH bonding [1]. The Si-OH bond is well known as a major problem at low-temperature grown SiO₂ films. Figure 2 shows the deposition rate dependence of TCE concentration for N₂ (S.O.) of 0.25 and 0.35 lm. It can be seen from Fig. 2 that the deposition rate increases with TCE concentration from about the low level of ~0.08 to ~0.65 mol/m³. This is because TCE enhances chemical reaction so that the deposition rate can be increased, as discussed in the last JSAP meeting [2]. Further, it is found that, after reaching a maximum deposition rate, the deposition rate decreases. This can be explained as follows: When the TCE concentration becomes sufficient enough for chemical reaction, gas phase reaction is prompted, which leads to the consumption of the reactant species near the substrate rather than forming a film on the substrate. It was also observed that the Si-OH content in the deposited films does not depend much on the TCE concentration.

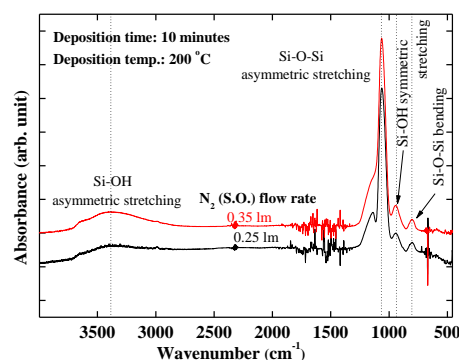


Fig. 1 FT-IR spectra of the SiO₂ films deposited with N₂(S.O.) = 0.25 and 0.35 lm.

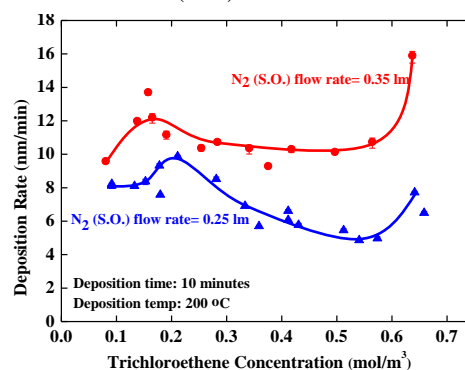


Fig. 2 Deposition rate dependence on organic solution concentration.

Summary: The deposition rate initially increases with TCE concentration, but excess of TCE concentration enhances gas phase reaction, which reduces deposition rate. In the meeting, we will show more detailed data under various deposition conditions and discuss them.

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References: [1] Horita *et al.*, *Jpn. J. Appl. Phys.*, **48**, (2009), 035502. [2] Puneet Jain *et al.*, Abstract JSAP 77th Autumn Meeting, 2016, E 16a-P4-3.