Development of apparatus supplying hydrogen radical remotely to decompose $SiCl_4$ source

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Introduction: Siemens process, in which $SiHCl_3$ source is reduced in H_2 ambient, has been most commonly used for obtaining high-pure silicon production. Drawbacks of this process are a low yield of ~25% and formation of a by-product of $SiCl_4$ that is chemically more stable. We have proposed the use of hydrogen radical instead of H_2 gas to improve the yield of Si in Siemens process. The effect of H-radical on decomposing $SiHCl_3$ and $SiCl_4$ has been confirmed under 20 mTorr of low pressure [1]. For practical process, however, H-radical must be generated in the condition higher than atmospheric pressure. Also, it must be remotely supplied into a reaction chamber. In this study, we have developed the apparatus to generate hydrogen radical by using tungsten filament in higher pressure. The effect of H-radical on the decomposition of $SiCl_4$ source has been studied during supplying H-radical remotely into the reaction chamber.

Experiments: Our experiment equipment is composed of two main parts: the main reaction chamber and the H-radical generator with W filament (0.5mmø, 1m). The two parts are connected by tube covered inside with an alumina pipe (200 mm, 16 mmø). The main chamber was heated at 1000°C using Zr filament, and the inside was mostly filled with Ar gas to avoid the generation of H-radical in the main chamber. The W filament in the H-radical generator was heated from RT to ~1500°C using AC power supply. A Q-mass spectroscopy connected to the main chamber was used for detecting the decomposition of $SiCl_4$. Ar gas with 60 sccm of flow was used as carrier gas for supplying $SiCl_4$. H-radical was supplied from the generator chamber to the reaction chamber though 3 mmϕ of small aperture. The pressure in main chamber was kept at 100 Pa, and H-radical generator was kept at 2.0 kPa with 1.2 L/min of H_2 gas flow.

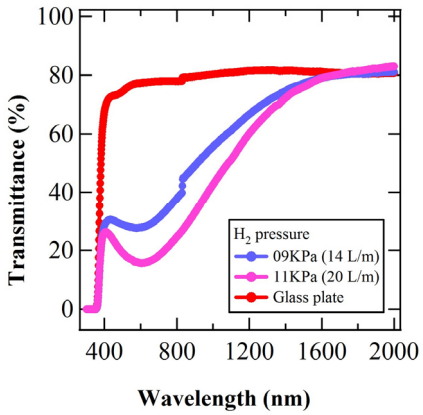

Fig. 1 Transmittance of WO_3 placed in the main chamber when W-filmanet in the H-radical chamber was heated at 1500 °C with H_2 flow of 14 and 20 L/min.

Results and discussion: To confirm the introduction of H-radical into the main chamber, the phosphate glass doped tungsten (WO_3) was used. Transparent WO_3 becomes darker, when the H-radical reacts with its surface. Figure 1 shows the dependence of the optical transmittance spectra for WO_3 placed on the alumina tube in the main chamber, when W filament was heated at 1500 °C with H_2 gas flow of 14 L/min and 20 L/min. From the ratio of transmittance at 600 nm [2], H-radical density can be estimated at 0.7 and 1.1 x 10^{12} cm^{-3}, respectively. It was confirmed that H-radical can be supplied remotely into the reaction chamber. Figure 2 shows the variation of partial pressure of m/z 170 and 133 corresponding to $SiCl_4$ and $SiCl_3$, respectively, detected with the Q-mass spectroscopy, when $SiCl_4$ source was supplied into the main chamber. At the temperatures of W-filament of H-radical chamber higher than 1200 °C, the signals related to source material are decreased, indicating that gas must be decomposed by H-radical supplied remotely. We need further development of apparatus so that it can be operated at atmospheric pressure.

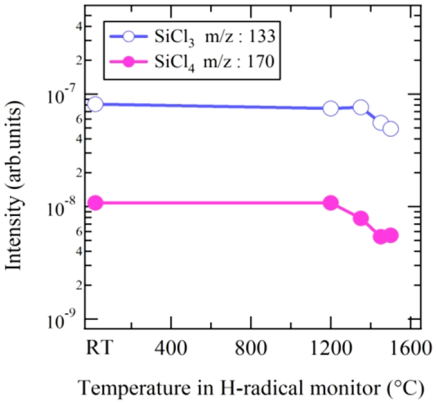

Fig.2. Variation of partial pressure of m/z 170 and 133 corresponding to $SiCl_4$ and $SiCl_3$, respectively, as a function of W-filament temperature.

References:

[1] Sumiya et al., J. of Phys. Conf. series **441**, 012003 (2013). [2] Takashi et al., Jpn J. of Appl. Phys. **44**, 732 (2005).