Formation of nanocrystalline silicon thin-films through magnesiothermic reduction of quartz-substrates
Tsukuba Univ.1, Muhammad Monirul Islam1, Takeaki Sakurai1, and Katsuhiro Akimoto1
E-mail: islam.monir.ke@u.tsukuba.ac.jp

Introduction: Silicon (Si) is the most abundant semiconductor-material found in form of silica (SiO2) that contributes to the large-scale to microelectronics, and optoelectronics-field etc. In particular, nanocrystalline Si (nc-Si) is promising for future optoelectronics, since it shows material properties which combine advantages of both amorphous-Si and polycrystalline-Si. However, current methods of obtaining nc-Si thin-films involve re-crystallization of amorphous-Si, or direct deposition of nc-Si through plasma enhanced chemical vapor deposition (CVD), hot wire CVD, glow discharge CVD etc. Various complex-steps, requirement of higher process-temperature, and application of toxic materials in the above techniques limit its potential applications.

We propose a simple method for the formation nc-Si thin-films on low cost substrate (silica: glass or quartz etc) through metallothermic reduction. Particularly, in this study, we have focused on the formation of nc-Si-film through magnesiothermic reduction of quartz (SiO2). According to thermodynamic properties of SiO2, magnesiothermic reduction occurs around melting temperature of magnesium (Mg), which is around 650°C following the basic equations:

\[
\begin{align*}
\text{SiO}_2 \text{(Solid)} + 4\text{e}^- & \rightarrow \text{Si (reduced)} + 2\text{O}^2- \quad \text{(1)} \\
\text{Mg (s)} & \rightarrow \text{Mg}^+2 + 2\text{e}^- \quad \text{(2)} \\
\text{SiO}_2 (s) + 2\text{Mg} & \rightarrow 2\text{Si (s)} + 2\text{MgO} \quad \text{(3)}
\end{align*}
\]

Experimental: Mg thin-films with thickness of ~1 μm were deposited through RF magnetron sputtering on quartz-substrates. Then, Mg/quartz structure was transferred to a thermal annealing system, where it was annealed at 700°C in the vacuum; thereby, going through a magnesiothermic reduction, where surface of the quartz-substrate reduced to become Si-film. Several samples were prepared by changing the annealing time ranging from 1 ~ 9 hours. Obtained Si-films were characterized by Raman spectroscopy, photoluminescence, scanning electron microscopy (SEM), and optical absorbance measurement etc.

Results and Discussions:

Fig.1 (a) Raman spectra of magnesiothermic reduced Si-film on quartz-substrate. Inset of the figure shows fitting of the Raman spectrum; (b) Band-gap determination from data obtained by UV-visible measurement.

Shown in Fig. 1(a) is the Raman spectroscopy of the Si-thin film on the quartz-substrate, obtained through magnesiothermic reduction at 700°C with 3-hours of reaction time. Raman spectrum of the oriented single crystal Si-wafer has been shown as a reference. An asymmetric peak at ~507 cm⁻¹ has been apparent for the Si-film on the quartz-substrate. Fitting of the Raman spectra (inset of the figure) shows Gaussian-deconvolution of the two peaks centered around ~ 480 cm⁻¹, and 506 cm⁻¹, respectively. Peak at 506 cm⁻¹ roughly suggests formation of nc-Si with evolution of small amorphous phase at 480cm⁻¹. Optical band-gap (E_g), of the Si-film obtained by absorption measurement was found around 2.8 eV, which is characteristic for the nc-Si thin-films. Annealing time was varied during reduction to study the effect of reaction time on the quality of the nc-Si film. Optical and structural properties were discussed in relation to the various annealing time at 700°C.