Investigation of Si paste for coated solar cell application

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論 文 内 容 の 要 旨
Thesis Summary

Energy crisis has increasingly become a bottleneck restricting the economic development of the international community. Among all kinds of renewable and clean energy sources, solar power is considered to be one of the most promising energy sources. A solar cell is the core device of photovoltaic power generation system. Among all kinds of solar cells, the crystalline Si wafer based solar cell is the most matured and commercialized photovoltaics device. However, the cost of Si wafer which mainly comes from the costly Czochralski Si ingot growth and the kerf loss remains the major barrier that prevent further large-scale installation. In the previous researches, utilizing Si pastes fabricated by planetary ball milling the n- or p-doped Si fragments was one of the simple kerf-less approaches that could skip the ingots growth and sawing process. The Si paste consists of Si nanocrystals owned some extraordinary properties due to the size effect. However, Si paste was found to be oxidized easily even in high purity Ar gas atmosphere due to the high surface to volume ratio of the Si nanocrystals. Hence, this PhD research intended to investigate the oxidation reduction technologies employed on Si paste, while continuing to further improve the Si paste electronic device performance, especially the photovoltaic performance.

There were three topics investigated in this dissertation:

(1) In Chapter 2, the first attempt was to lower the annealing temperature. p- and n-type Si paste films coated on Al and Al-sputtered Fe substrates were annealed at low temperatures to fabricate pn homo-junction devices for development of low-cost solar cell devices. This process based on aluminum-induced-crystallization (AIC) which enabled one to recrystallize Si paste films with reduction of residual tensile stress while suppressing oxidation even at low temperatures (400 to 550°C). The current-voltage characteristic of the pn homo-junction device using the Si paste films showed rectification with the on/off current ratio of about 3200 and the reverse current density of the order of 10^{-9} A/cm^2 at room temperature. The photocurrent of 0.058 μA/cm^2 in the pn homo-junction diode under AM1.5 illuminations was observed.

(2) In Chapter 3, the second attempt was rapid thermal annealing (RTA). RTA was conducted in order to recrystallize the Si paste. It was possible to minimize the oxidation during the melting process of Si nanoparticles with RTA even at 1200°C in 1 s. Lowering of the melting temperature appeared to be due to the size effect and release of surface energy from the Si nanoparticles. RTA was conducted in an infrared furnace with temperatures varying from 1150 to 1300°C. Si pn homo-junction structure was also fabricated by coating p-type followed by n-type Si pastes on a carbon substrate. Typical rectifying characteristics and slight photo-induced current of 10 μA/cm^2 was observed.

(3) In Chapter 4, the third attempt was to introduce a layer of material that was more

susceptible to oxidation. In order to reduce the oxidation of Si paste during the RTA, Ti was deposited on the p-type Si paste since the Gibbs' free energy of Ti oxidation was smaller than Si oxidation. It was found that the RTA condition of 1200°C/2 s enabled the Si paste to obtain the least oxidation and the best crystallization conditions as well as the transformation of the Ti layer into rutile TiO_2 phase. It was observed that some molten Si grain lumps appeared on the surface of the sample annealed at 1200°C for 2 s. This phenomenon was due to the melting process of the surface Si particles promoted by the Ti layer. Typical rectifying characteristic with a photocurrent density of 36 $\mu A/cm^2$ was observed with the TiO_2/p-Si paste heterojunction device annealed at 1200°C for 2 s.

In conclusion, this dissertation presented significant progress in oxidation reduction technologies as well as the advancement of photovoltaic performance for Si paste. Together with detailed explanations of the underlying fundamentals, the results presented in the dissertation are expected to shorten the gap between c-Si wafer and Si paste. It is expected that after adopting the oxidation reduction methods introduced in this investigation, the Si paste will be regarded as an alternative raw material for manufacturing the low-cost coated solar cells which will be beneficial to the large installation of photovoltaic system in the future human society.