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論文審査の要旨 (2000 字程度)

In this thesis, the electrochemical passivation of porous silicon (PSi) for tandem solar cells based on experiment and simulation are investigated.

Chapter 1 introduces the concept of all-silicon tandem solar cell, which has the potential to surpass the theoretical efficiency limit of a single-junction silicon solar cell; it also eliminates the lattice mismatch issue found in hetero-junction structures. The porous silicon (PSi) is selected as a candidate for a top cell of the tandem due to its adjustable band gap, which is reported to be in a range of 1.2 - 2.2 eV depending on its porous structure, and possibly originated from the quantum confinement effect. It also has an anti-reflective property, which leads to the enhancement of light absorption. With these benefits, it provides the opportunity to use the PSi to improve the open circuit V_{oc} , short circuit current J_{sc} , as well as the efficiency η of the solar cell. However, the surface area and defects greatly increase compared to crystalline silicon (c-Si), which increases the surface recombination rate. Thus, a suitable passivation method for the nano-structured Si is necessary to minimize the surface recombination.

Chapter 2 addresses the statement of purpose, where a low-cost electrochemical passivation technique is proposed to achieve a uniform passivated layer on PSi. The PSi fabrication and the passivation process are varied in order to find the optimal fabrication condition that yields the best solar cell performance, which are compared with the conventional atomic layer deposition (ALD) method. Finally, in order to confirm the true electronic structure of PSi/c-Si system, various PSi models are created and calculated.

Chapter 3 describes the experimental PSi fabrication and the passivation conditions as well as the characterization process. The detail for the simulation part is also described, which includes the calculation methods, the models creation, the evaluation of the band structure.

Chapter 4 is divided into 2 main parts: the experimental and the simulation part.

For the experimental part, the structure of PSi in this study was controlled to have 25% porosity with a p-n junction at 200 - 250 nm deep where the PSi was introduced in the n layer of the p-n junction (so-called Type III structure). After passivation with the three methods, the results of carrier lifetime measurements showed that the performance of each method was

similar; all of the lifetimes are short (in a few μs range) compared to common Si solar cell (in ms range). However, while ALD and supercritical fluid methods passivated both sides of the PSi wafer, the electrochemical method only applied to the front surface. Finally, the solar cell performance of electrochemical passivated and ALD-passivated samples was measured. With ALD passivation, all the solar cell parameters (V_{oc} , J_{sc} , η) were poor, as well as diode characteristics due to the formation of a resistive layer on the top surface. The electrochemical passivation with varied reaction times was explored. The passivated PSi improved the quantum efficiency (QE) especially at the shorter wavelength compared to c-Si, while the unpassivated PSi sample showed poor QE due to significant recombination. The solar cell efficiency result showed that the optimized passivation time is 30 seconds, at which the recombination is minimized, while the passivated oxide layer is not too resistive. When compared to the c-Si solar cell and an unpassivated sample, it showed the improvement of J_{sc} and efficiency of electrochemical passivated PSi solar cell, while all V_{oc} values are in the range of 0.51-0.54 V, i. e., not improved. Therefore, in order to verify the possibility to improve V_{oc} by using PSi, the electronic structure of the PSi/c-Si system needed to be clarified by simulation.

In the simulation part, the band structures induced by porosity and passivation of Si were computed by the Density Functional Tight Binding (DFTB) method and compared. We created 4 types of models as a representative of PSi: pillar, crater, internal sphere, and internal cube models. The results showed that the only component that determines the degree of expansion is the size of Si framework itself, not the pore. The diameter of the pillar, the wall of the crater, and the thickest Si skeleton part were varied including sizes which induce quantum confinement. The pillar height and crater depth have little to no effect on the bandgap as long as the Si base is thicker than 1 nm. Also, only when two pillars are connected physically and turn to crater that the decrease of band gap is observed; the more in contact, the more the band gap converges to bulk c-Si value. The opposite is also observed when the crater diameters are large enough that the Si wall part is isolated, and the band gap drastically increases. Quantum confinement induced gap expansion was only observed when Si features approached 1 nm scale. To connect to the experiment to the simulation result, it is found that, with the PSi wall thickness of more than 3.5 nm that is observed in TEM, the band gap expansion effect is minimal (1.2-1.4 eV). Thus, the PSi framework must be made smaller in future fabrication.

In summary in chapter 5, this thesis explored the possibility to use PSi as a top cell of the all-silicon tandem solar cell by experiment and simulation. By using the electrochemical passivation, the uniform and high coverage oxide layer could be formed on PSi layer, resulting in the improvement of solar cell performance. The simulation on the electronic structure of relatively-large PSi/c-Si system also suggests the PSi structure that can further improve the performance of solar cell, thus providing a guideline for the future PSi fabrication. Therefore, these results largely contribute to the research field in the photovoltaics and experimental and computational material science, and the thesis has a sufficient quality to be permitted as a PH.D. in engineering.

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