様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科研費
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研究課題名（和文）トンネリング酸化物パッシベーション層を用いたヘテロ接合シリコン太陽電池の開発

研究課題名（英文）Development of silicon heterojunction solar cells with tunneling oxide passivation layers

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交付決定額（研究期間全体）：（直接経費）　　2,300,000円

研究成果の概要（和文）：ヘテロ接合型結晶シリコン太陽電池の新規正孔コンタクト層の開発を行った。
従来構造に使われるアモルファスシリコンは耐熱性が低く、光学損失を有するため、それぞれ低コスト製造プロセス導入や変換効率向上の妨げになっている。これらの問題を解決すべく、代替正孔コンタクト構造の開発を行った。トンネル効果を利用した厚さ2 nm以下の酸化膜でシリコン表面の欠陥を終端し、正孔選択層として種々の材料を検討した結果、変換効率15.4%を達成した。

研究成果の学術的意義や社会的意義
本来絶縁体である酸化膜を厚さ2 nm以下にすることでキャリアの輸送が可能となり、またそれを適切な材料と組み合わせることでこれまでにないコンタクト形成に新たな可能性を示した。さらに、提案する簡易な作製方法はデバイス製造の低コスト化に貢献できる。
本研究の成果は太陽電池だけでなく多様な半導体デバイスへの応用が期待できる。

研究成果の概要（英文）：Alternative p-type contact structure was developed for application to silicon heterojunction solar cells.
The amorphous silicon material used in the conventional structure has a low thermal tolerance and show parasitic absorption losses that, respectively, restrict the use of low-cost fabrication processes and limit further efficiency improvement. An alternative hole selective contact structure is proposed to overcome these issues. By using an oxide passivation layer thinner than 2 nm that allows tunneling effect, and after evaluating several hole selective materials, an efficiency of 15.4% was achieved.

研究分野：太陽電池

キーワード：シリコン太陽電池　キャリア選択層　パッシベーション　ヘテロ接合

様　式　C－19、F－19－1、Z－19、CK－19（共通）

１．研究開始当初の背景

The concern about environmental issues and energy sustainability in recent years has resulted in an emerging demand for renewable energies around the world. Among of the large variety of renewable energy sources, solar energy has attracted considerable attention because of its several advantages such as infinite source of energy, low maintenance cost, low geographical restrictions, etc. To further spread the solar power technology, reductions in generation cost are required.

Crystalline silicon (c-Si) solar cells are the most common type of solar cell, representing almost 90% of the market. Therefore, to further reduce the generation cost of solar power, it is essential to reduce the fabrication cost and increase the efficiency of c-Si solar cells.

The most efficient c-Si solar cell reported up to now has a heterojunction structure that utilizes undoped and doped amorphous silicon to form the passivation layer and p-type emitter layer, respectively. However, amorphous silicon shows parasitic absorption losses that limit the short circuit current density (J_{sc}). Additionally, there is less tolerance in the highest temperature that can be used during the fabrication, restricting the use of cheap electrode metallization that is usually performed at temperatures higher than 800°C. These factors limit the efficiency and restrict the use of low-cost fabrication processes.

２．研究の目的

The objective of this research is to further enhance the efficiency and lower the fabrication cost of c-Si heterojunction solar cells by developing an alternative p-type contact structure that overcome the limitations of the conventional structure. To achieve this objective, alternative materials are used for the passivation layer and p-layer, aiming in combining high passivation quality, low parasitic absorption loss, and high thermal tolerance.

３．研究の方法

To fabricate a p-type contact, or hole selective contact, that combines high passivation quality, low parasitic absorption loss, and high thermal tolerance, oxides such as silicon oxide (SiO_x) and alumina (Al_2O_3) were applied as the passivation layers. Although these materials are usually insulators, the use of thicknesses lower than 2 nm creates a tunneling effect that allows the carrier flow.

The thin SiO_x layer was fabricated by simply dipping the silicon wafer into a diluted peroxide solution at room temperature. The Al_2O_3 was fabricated by atomic layer deposition (ALD). Several materials were considered for the hole selective layer, such as p-type microcrystalline silicon (p-μc-Si:H), p-type microcrystalline silicon oxide (p-μc-SiO_x:H), and molybdenum oxide (MoO_x). p-μc-Si:H was fabricated by plasma-enhanced chemical vapor deposition (PE-CVD) method, using a mixture of hydrogen, silane, and diborane as source gases. Carbon dioxide was added to the same mixture to fabricate the p-μc-SiO_x:H material. MoO_x was deposited by thermal evaporation using MoO3 powder (99.97%, Sigma Aldrich) as source material. The MoO_x thickness was controlled using a quartz oscillator. The native oxide on the c-Si wafer was removed by dipping into diluted HF before the deposition or growth of any layer. The n-a-Si:H back-surface field layer applied for solar cell structures was fabricated by PE-CVD using a mixture of silane, hydrogen, and phoshine gases.

The passivation quality was evaluated by measuring the minority carrier effective lifetime of the samples using a quantum-steady-state photo-conductance apparatus. The performance of the solar cells were evaluated by measuring the current density-voltage (J-V) characteristics of the samples under AM1.5G illumination.

４．研究成果

Initially, the combination of oxide passivation layers (SiO_x and Al_2O_3) and p-layer deposited by PE-CVD (p-μc-Si:H, p-μc-SiO_x:H) was considered. Figure 1 shows the relationship between resistance and effective lifetime of samples with symmetrical structures. p-type c-Si wafer was used in this experiment. Lifetime measurements were carried out before the electrode deposition.

The lower resistance values and the highest lifetimes were obtained with p-μc-SiO_x:H layer. The higher lifetimes can be attributed to the higher amorphous ratio in this material compared to the p-μc-Si:H film. The higher is the amorphous ratio, the higher is the hydrogen content in the film that can contribute to the passivation of dangling bonds. Additionally, the high crystalline ratio in the p-μc-Si:H film suggests that its bandgap is close to that of c-Si, which result in a lower barrier to repel the minority carriers. The crystallinity tended to increase with thickness for both p-type materials.

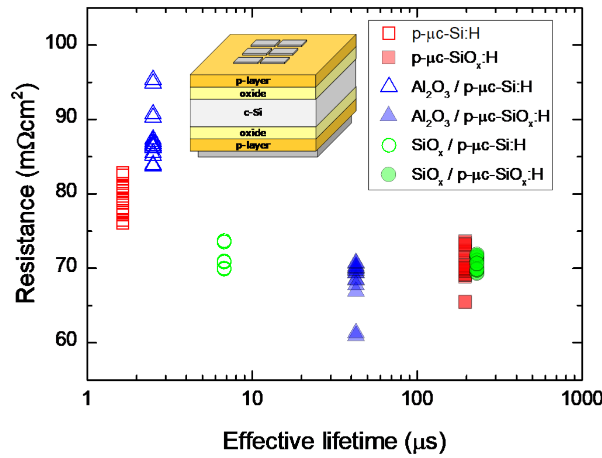

Fig. 1. Relationship of resistance and effective lifetime of samples with different p-layers.

Although the insertion of SiO_x between the silicon wafer and the p-μc-SiO_x:H (p-μc-Si:H) layer was effective in hindering interfacial recombination, the passivation quality was inferior than that required for a high efficiency solar cell. This occurs because the high hydrogen dilution required to deposit a microcrystalline film with the PE-CVD method can damage the c-Si surface.

To avoid the deterioration of passivation quality, MoO_x was applied as an alternative material for the hole selective contact. The large work function of MoO_x creates a band bending on the c-Si surface that repels the electrons, allows the hole flow toward the contact, and creates an inversion layer at the c-Si/p-type contact interface. As a result, rectifying properties similar to those obtained with conventional pn-junctions can be realized. Addtionally, MoO_x has a wide bandgap that is beneficial to avoid parasitic absorption losses.

The passivation effect achieved in MoO_x/n-c-Si/MoO_x structures without passivation layers has been attributed to both band bending and unintentional silicon oxide formation at the MoO_x/c-Si interface. [Gerling et al., J. Mater. Res. **32**, 260 (2017)] To clarify the effect of interfacial SiO_x layer on the passivation properties, the effective lifetime of solar cell precursors with and without intentionally grown SiO_x layers was investigated. Figure 2 shows the minority carrier effective lifetime dependence on carrier density for samples with MoO_x / SiO_x / n-c-Si / SiO_x / n-a-Si structure. Samples without the SiO_x layer on the MoO_x/c-Si interface were also prepared by etching the SiO_x after the n-a-Si:H layer deposition and before the MoO_x evaporation. Double-sided-polished n-c-Si wafers (FZ, <100>, 280 μm, 1–5 Ω·cm) were used as substrates. The effective lifetimes for samples with and without SiO_x at the minority carrier density of $1x10^{15}cm^{-3}$ were 1.2 ms and 740 μs, respectively. As a result, insertion of the SiO_x layer results in implied-Voc improving from 653 to 667 mV. The intentional insertion of the SiO_x passivation layer is effective to improve the passivation properties, presumably by separating the nonstoichiometric and defective MoO_x surface from the c-Si surface.

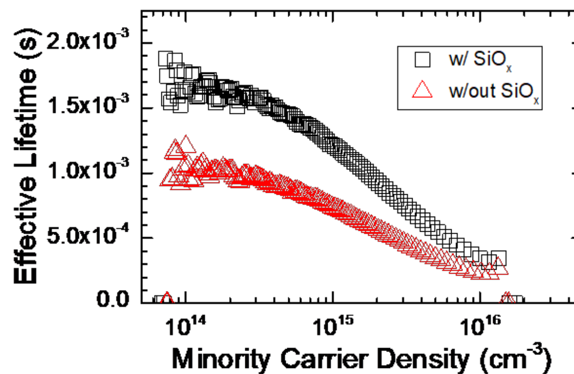

Fig. 2. Effective lifetime dependence on minority carrier density for samples with and without SiO_x layer.

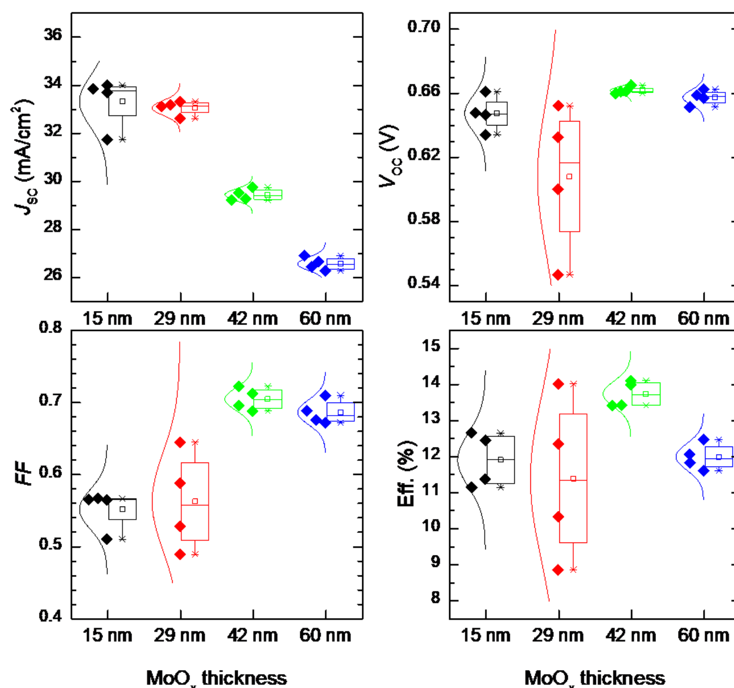

Fig. 3. Dependence of *J-V* parameters on the MoO_x hole selective emitter thickness.

Next, to clarify the relationship between carrier selectivity and optical losses, the dependence of the heterojunction solar cell performance on MoO_x hole selective emitter thickness was investigated. For this purpose, an Al/Ag/ITO/MoO_x/SiO_x/ n-c-Si/SiO_x/n-a-Si:H/Ag/Al structure was fabricated. Figure 3 shows the J-V parameters of the fabricated solar cells as a function of the MoO_x layer thickness. The thicker was the MoO_x layer, the lower was the J_{sc}, suggesting that the MoO_x film contain defects that cause absorption losses. On the other hand, the thicker was the MoO_x layer, the higher was the fill-factor (FF). These results indicate that a fine control of the MoO_x film thickness is required to obtain an optimum performance. The highest efficiency in this experiment was 14.1% [open circuit voltage (V_{oc}) = 665 mV, FF = 71.3%, J_{sc} = 29.8 mA/cm^2] with a 42-nm-thick MoO_x layer, while similar efficiency of 14.0% was obtained with 29-nm-thick MoO_x (V_{oc}= 652 mV, FF= 64.5%, J_{sc}= 33.3mA/cm^2).

The MoO_x optimum thickness would largely depend on the film stoichiometry since it affects the work function. Moreover, the band bending is also affected by the ITO layer that is deposited on the MoO_x layer. After further optimization of ITO sputtering, a higher efficiency of 15.4% (V_{oc} = 651 mV, FF = 69.9%, J_{sc} =33.9 mA/cm^2) was achieved with a 29-nm-thick MoO_x layer. Comparing the with the previous result using the same MoO_x thickness, the FF was remarkably improved. This indicates that careful device optimization should be carried out not only at the passivation layer/MoO_x interface, but also at the MoO_x/ITO interface.

As a summary, c-Si heterojunction solar cells using ultrathin tunneling oxide passivation layer and an alternative hole selective layer were successfully developed. Considering that the solar cells were fabricated using polished wafers without texture and without anti-reflection coating, the obtained efficiencies are encouraging and demonstrate that the proposed SiO_x/MoO_x stack is a promising hole selective contact.

５．主な発表論文等

〔雑誌論文〕（計 0 件）

〔学会発表〕（計 4 件）

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〔図書〕（計 0 件）

〔産業財産権〕
○出願状況（計 0 件）

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権利者：
種類：
番号：
出願年：
国内外の別：

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名称：
発明者：
権利者：
種類：
番号：
取得年：
国内外の別：

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