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論文 / 著書情報
Article / Book Information

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Title(English)	Passivation of TiO2/Si by Interlayer SiOx studied with Scanning Zone Annealing and Atomistic Modelling for Perovskite/Si Tandem Solar Cells
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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

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（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	応用化学 系 エネルギー コース	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of （ 工学 ）
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要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Tandem solar cells that combine metal organohalide perovskite and crystalline Si materials have attracted much intention for next-generation photovoltaic devices as they promise high theoretical energy conversion efficiency combined with low fabrication cost potential. However, the current efficiency of these devices is almost equivalent to the single-junction Si and perovskite solar cells. Therefore, these perovskite/Si tandem solar cells (PSTSCs) do not present economic benefits today based on the increase in materials and technological costs required to fabricate these solar cells.

One way to increase the efficiency of solar cells is to increase the carrier lifetime of the devices. Longer carrier lifetime leads to the enhancement in the number of extractable carriers from the device which directly increases the maximum current achievable by the photovoltaic device. For Si-based devices, carrier recombination at the surface of the Si is widely known to be the bottleneck for the carrier lifetime of the devices. To reduce carrier recombination at the Si surface, methods to passivate the surface of Si have also been extensively studied for single-junction Si solar cells. These passivation methods mostly adopted the deposition of oxide materials with very large band gaps such as Al_2O_3 and crystalline SiO_2 among other materials. Therefore, these methods cannot be directly applied to the PSTSCs, as band alignment between the perovskite and electron transporting layer is required for maximum carrier extraction.

For the reasons stated above, this thesis aims to maximize the efficiency of PSTSCs by effective passivation of the TiO_2/Si interface. As one of the most widely used electron transporting layers, TiO_2 promises high carrier extraction for the perovskite top cell, however, its effectiveness to passivate the Si surface is very limited.

Firstly, the bottlenecks of the PSTSCs were investigated by fabricating single-junction perovskite solar cells on Si substrates. Here, it was found that the low carrier lifetime at the crystalline Si substrate with ETL materials, such as TiO_2, is one of the bottlenecks that reduce the efficiency of the PSTSCs.

To better understand the role of the interface and to be able to rationally design TiO_2/Si interface optimal for PSTSC performance, we used atomistic models of the Si, amorphous SiO_x, and TiO_2, as well as the Si/a-SiO_x interfaces studied using Density Functional Theory (DFT) and Density Functional based Tight Binding (DFTB) methods. Using DFT calculation of the c-Si, SiO_x, and TiO_2 models, it was found that the band gap of the SiO_x changes with different layer properties, particularly in the composition of the material (ratio between Si and O atoms). Using DFTB calculations, it was found that the band gap of the SiO_x layer decreases as the thickness of the layer increases. This may lead to an increase in the ability of the interlayer to maintain fixed charges as the thickness of the SiO_x layer increases. This phenomenon can

be useful to alter the recombination dynamics at the TiO_2/Si if adjustment on the thickness of the natively formed amorphous SiO_x layer can be performed after the deposition of the TiO_2 layer.

Based on the findings above, Scanning Zone Annealing (SZA) was proposed to directly treat the TiO_2/Si interface and alter the properties of the natively-formed interfacial SiO_x layer. Here, it was found that the TiO_2/Si interface treated with SZA showed an increase in the carrier lifetime. The level of improvement by SZA was in direct correlation with the amount of heat transferred during the SZA process with the highest carrier lifetime achieved at 355 μs. This carrier lifetime was one of the highest reported for the TiO_2/Si system at the time the work was reported. Furthermore, it was found that the optimum thickness of a-SiO_x that maximizes carrier lifetime was 2.4-2.6 nm. This phenomenon can be explained by the trade-off between field-effect and chemical passivation as estimated in previous findings by the atomistic models as well as from literature. Through numerical simulation PC1D, the improvement in carrier lifetime was expected to increase the efficiency of the PSTSCs by at least 0.8%.

Moreover, modeling Si/a- SiO_x/TiO_2 interfaces might provide deeper insight into the recombination dynamics between Si and perovskite subcells. However, direct calculation of Si/a-SiO_x/TiO_2 was impossible using the semi-empirical method DFTB due to the lack of parameterization of the pair-wise interaction between Si and Ti atoms. To solve this problem, a novel atomistic modeling method that combines the *ab-initio* DFTB with potential-based molecular mechanics was proposed. This Hybrid DFTB/MM method was proven to be effective to perform structural optimization as well as the calculation of the electronic interaction of different materials that include TiO_2-C_3N_4 and covalently bonded Si-Ti-O systems. The method was then used to investigate the Si/a-SiO_x/TiO_2 interface models. Here, it was found that the interaction at the interface of TiO_2/a-SiO_x as well as a-SiO_x/c-Si leads to the existence of states located at the band gap of each material. At Si/SiO_x interface, the band states near CBM were found with models with thicker SiO_x. Thus, the increase in the number of recombination centers can be expected with an increase in the thickness of the SiO_x interlayer.

備考 ： 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。
Note : Thesis Summary should be submitted in either a copy of 2000 Japanese Characters and 300 Words (English) or 1copy of 800 Words (English).

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