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著者(和文)	GEKKOPATRIABUDIUTAMA
Author(English)	Gekko Patria Budiutama
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論文審査の要旨及び審査員

報告番号	甲第　　号		学位申請者氏名		GEKKO PATRIA BUDIUTAMA	
論文審査 審査員		氏　名	職　名		氏　名	職　名
	主査	伊原　学	教授	審査員	多湖　輝興	教授
	審査員	Manzhos Sergei	准教授			
		山田　明	教授			
		平山 雅章	教授			

論文審査の要旨（2000 字程度）

In this thesis, the passivation of the TiO_2/Si interface using a natively formed SiO_x interlayer was studied using experimental and atomistic modeling approaches for application in perovskite/Si tandem solar cells.

In Chapter 1, the perovskite/Si tandem solar cell, the importance of the interface between the perovskite top cell and Si bottom cell, and Si passivation technologies were introduced. Here, it was pointed out that, although Si passivation technologies have been thoroughly studied, the lack of band alignment between the current state-of-the-art Si surface passivation materials such as Al_2O_3 and SiO2 with perovskite materials leads to the need for a new material passivation method that accommodates electron transport in perovskite/Si tandem solar cells. Furthermore, the concepts of Scanning Zone Annealing, Density Function Theory, and Density Functional Tight Binding were introduced.

The scope of the thesis was introduced in Chapter 2. Here, the study aims to maximize the efficiency of perovskite/Si tandem solar cells based on a new way to treat metal oxide/Si interface (Scanning Zone Annealing) and to better the understanding of the Si/TiO_2 interface using atomistic modeling approaches.

Chapter 3 describes the methodologies used in the experimental and calculation parts of the study. In the experimental part, the followings are described: the fabrication of perovskite solar cell and perovskite-on-silicon solar cell, Scanning Zone Annealing to passivate the TiO_2/Si interface, and estimation of the change in solar cell efficiency using solar cells simulation software PC1D. In the calculation part, the following are described: an overview of Density Functional Theory (DFT) and Density Functional Tight Binding (DFTB). Calculations of TiO_2, SiO_x, and Si interfaces using the above methods are performed.

In Chapter 4, the bottleneck of the perovskite/Si tandem solar cell using single-junction perovskite solar cells grown on Si substrate (PoSiSC) is addressed. Here, it was found that although the correct open circuit voltage of around 1.1 V was achieved, the short circuit current density for the PoSiSC devices was very low. Two main reasons for this were proposed: the lack of transparency of the electrode used in our PoSiSC device and high carrier recombination at the TiO_2/Si interface. Here, we found that the latter significantly reduced the carrier lifetime of the carriers and therefore limit the number of extractable carriers. Furthermore, a very thin SiO_x interlayer was found at the TiO_2/Si interface. This interlayer is

unavoidable for most metal oxide/Si interfaces. Then it was hypothesized that an increase in carrier lifetime might be achievable by changing the properties of the interlayer SiO_x.

The idea of using unavoidable natively formed interlayer SiO_x was then explored using DFT and DFTB in Chapter 5. A suitable band alignment between TiO_2 and c-Si was first confirmed by DFT. From the larger-scale DFTB calculation, it was found that reducing the thickness of the layer and increasing the oxygen content of the SiO_x interlayer led to the expansion of its band gap. As the capacitance of metal oxide materials can be approximately calculated from the band gap and thickness, here it was argued that the reduction in SiO_x layer thickness might increase the capacitance of the layer. This led to a possible increase in passivation effectiveness, especially field-effect passivation.

Based on the result above, Scanning Zone Annealing (SZA) was proposed to alter the properties of the SiO_x interlayer at TiO_2/Si interface in Chapter 6. Here, a significant increase in carrier lifetime from less than 5 μs to up to 355 μs was observed on TiO_2/Si interface using p-type and n-type Si wafers. This improvement allows the perovskite/Si tandem solar cell to achieve an efficiency of up to 29.7% based on the simulation using PC1D software. Furthermore, different SZA conditions (scanning rate and lamp output) led to a different level of improvement in carrier lifetime. To investigate this phenomenon further the thickness of the SiO_x interlayer was directly observed using Tunneling Electron Microscope. Here, it was found that an optimum SiO_x interlayer thickness that maximizes carrier lifetime was 2.4-2.6 nm. Furthermore, as the SiO_x interlayer thickness increases, increasing then decreasing trends were observed on the carrier lifetime. While the increasing trend can be explained based on the calculation in Chapter 5 as well as the literature, the decreasing trend was specific to the TiO_2/a-SiO_x/Si system.

In Chapter 7, the change in trap-states with the increase in SiO_x thickness was investigated using novel atomistic modeling of TiO_2/a-SiO_x/Si interface models. Here, DFT calculation of the models was found to be too costly. Moreover, DFTB cannot be directly implemented as a suitable parametrization for Si and Ti pair-wise interaction is currently missing. To solve this problem, a novel hybrid approach that combines DFTB and force-field-based molecular mechanics was developed. Here, it was shown that the accuracy of the DFTB/MM was comparable to the original DFTB and DFT calculations. This novel DFTB/MM approach was then used to calculate the TiO_2/a-SiO_x/Si interface. Here, it was found that the number of trap states at TiO_2/a-SiO_x and a-SiO_x/Si interfaces was increased with the increase in a-SiO_x interlayer thickness. Thus, the declining trend in carrier lifetime observed in the experiment can be explained by the increase in the number of the recombination centers, in other words, the reduction in chemical passivation as the thickness of the interlayer increases.

In summary, this thesis explored the possibility of using a natively formed SiO_x interlayer to passivate the TiO_2/Si. Atomistic modeling approaches were utilized to inspire and explain, while an experimental approach using SZA led to real-world improvement in the carrier lifetime. These results therefore contribute to the fields of photovoltaic and computational materials science. Thus, the thesis was deemed of sufficient quality to complete the Ph.D. in engineering.