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論 文 要 旨
THESIS SUMMARY

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学籍番号： Student ID Number	17D20606	指導教員（主）： Academic Supervisor(main)	Prof. Manabu Ihara
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論文題目 Thesis Title	Dominant effect of perovskite grain size on solar cell performance and improved stability by all-inorganic light absorber

要旨（英文800語程度）
Thesis Summary (approx.800 English Words)

In this thesis, the predominant effect of perovskite grain size on solar cell performance is clarified, and an air-stable all-inorganic material is proposed.

Chapter 1 introduces information on the lead-halide perovskite solar cells (PSCs), which have attracted exciting attention due to their outstanding high power-conversion efficiency. The grain size is the dominant effect on any type of semiconductor solar cells, such as Si and CIGS. In contrast, it is an unclear effect on PSCs. In PSCs with inorganic structure (Transparent conductive oxide (TCO)/ Inorganic electron transport layer (ETL) /Perovskite /Hole transport layer (HTL)/Counter electrode), the grain size has been controlled by changing one or more parameters in the literatures; (1) the additive ratio in perovskite layer, (2) the doping percentage, (3) the perovskite formation temperature, and (4) ratio of raw materials. In these reports, the grain size effect could not be separated from the incidental effects (e.g., grain quality, grain boundary, and perovskite thickness were changed). Therefore, this problem inspired the objectives to clarify the direct effect of perovskite grain size and solar cell performance. The stability of PSC is also a critical issue that constrains PSCs from a commercial scale. Thus, the new material is proposed to be an all-inorganic air-stable perovskite material to improve PSCs stability.

Chapter 2 describes the optimization of the low-cost and straightforward spray pyrolysis to deposit a flat compact TiO_2 (c-TiO_2) as ETL with controlling the surface roughness (Rms). The effect of different solvents (ethanol, 2-propanol, 1-butanol, and 2-butanol), spray temperature (Ts), and annealing temperature (Ta) induced different Rms of c-TiO_2. According to heterogeneous nucleation theory, the surface roughness of c-TiO_2 can be one of the parameters that change the surface energy in the heterogeneous nucleation process. The largest grain size was achieved by using 1-butanol as a solvent. Therefore,

the optimization of the spray pyrolysis technique is one route to lower the surface roughness of c-TiO_2, enlarge the perovskite grain size, and prevent the imperfection of c-TiO_2 surface.

Chapter 3 describes clearly the dominant effect of grain size of methylammonium lead iodide ($MAPbI_3$) perovskite controlled by Rms of c-TiO_2 on the performance of perovskite solar cells. The direct dominant effect of $MAPbI_3$ grain size on an inorganic PSC performance was observed by controlling Rms of the c-TiO_2 as an ETL. For PSC with an active area of 0.54cm^2, perovskite grain sizes changed from 150 nm to 350 nm, the current density increased from 8 to 17 mA/cm^2, and the photoelectric conversion efficiency increased from 6.1% to 11.4%, respectively. Therefore, to increase perovskite grain size is an effective way for a high-efficiency, large-area, and wafer-size PSCs in commercial modules. This work also found that the average $MAPbI_3$ grain size can be controlled only by Rms of c-TiO_2 layer without additional factors. The stability of $MAPbI_3$ based solar cells measured under ambient operation also discussed by using solar cells with different $MAPbI_3$ grain sizes (100, 250, and 399nm). The results showed slower degradation in a sample with large grain sizes. However, its normalized PCE drastically decreased from 100% to 50% in 24 hours.

Chapter 4 describes research on an all-inorganic light absorber for perovskite solar cells. A tolerance factor was calculated as an indicator to predict the stability of a perovskite structure from desired inorganic ions. Density function theory (DFT) was adopted to estimate the electronic band structure, bandgap, and density of state (DOS). DFT with the generalized gradient approximation (GGA) showed an indirect bandgap (1.48eV). An air-stable all-inorganic material was synthesized by solid-state reaction. UV-vis and photoluminescence (PL) spectra were measured for the absorption and excitation spectrum, respectively. From the experiment results, this material showed an appropriate bandgap (1.8eV) for solar cell application. X-ray diffraction (XRD) showed phase purity and stability of powder and thin film. However, PL measurement under low temperature and X-ray photoelectron spectroscopy (XPS) revealed the impurity state on the thin-film surface after leaving in ambient for 7 days. These findings are beneficial to the understanding of new material electrical and optical properties.

Finally, in Chapter 5, the new material was deposited through a solution process for the first time and applied in solar cell applications. The same structure with standard PSC was used to fabricate a solar cell (TCO/c-TiO_2/inorganic absorber/HTL/electrode). However, the initial highest PCE showed only 0.12%. The possible reasons for low efficiency came from a poor thin film morphology, an indirect bandgap, and poor contact between each layer in a solar cell. Although this material has the potential for a solar cell application, it needs further improvement for thin film deposition and device fabrication.