論文内容の要旨 (代替)

申請者氏名　　ADITYA WAHYU ANUGRAH

Graduate School of Science and Technology Doctoral Thesis Abstract

Laboratory name
(Supervisor)Information Device Science Laboratory
(Prof. Yukiharu Uraoka)
Student ID2221414Submission date 2025/06/24
(yyyy/mm/dd)
Name
(surname)(given name)ADITYA WAHYU ANUGRAH
TitleStudies on eco-friendly bismuth-based material for lead-free perovskite solar cell applications
(鉛フリーペロブスカイト太陽電池応用に向けた環境配慮型ビスマス系材料に関する研究)

Chapter 1 Introduction

Photovoltaic (PV) technology is projected to become the leading global renewable energy source. The future solar cells must be safe, stable, and environmentally friendly. Bismuth (Bi)-based materials offer a promising alternative to toxic lead-based perovskites, due to their low-toxic, low-cost, stable, and environmentally friendly properties. However, their power conversion efficiency remains limited due to challenges such as poor film morphology, low carrier lifetime, and especially a large bandgap, which limits their effectiveness as light-absorbing layers. Various doping strategies have been explored to narrow the bandgap, such as doping $Cs_2AgBiBr_6$ with Sb^{3+} and Ru^{3+} to reduce the bandgap to approximately 1.86 eV and 1.80 eV, respectively, and Sulfur doping in $MA_3Bi_2I_9$ to achieve a bandgap of 1.67 eV. However, the achieved bandgaps are still higher than the ideal range of 1.1-1.4 eV required for optimal solar cell efficiency, and the impact of bandgap narrowing on overall device performance is still not fully understood.

Chapter 2 Effect of post-annealing treatment on the performance of bismuth tri-iodide solar cell

Figure 1. Morphology and performance of the device

Chapter 2 presents post-annealing treatments at various temperatures from 120°C to 180°C were performed to optimize the morphology and optical properties of thin films. The treatment induced change in lattice parameter which observed from X-ray diffraction (XRD) measurement. The best grain size was shown by post-annealing at 140°C. Photovoltaic performance evaluated through current density-voltage (*J-V*) measurements showed an improvement of conversion efficiency (PCE) from 0.074% to 0.139% with post-annealing treatment. An improvement of film quality was also evaluated by impedance spectroscopy analysis, which showed that post-annealing treatment reduced internal resistance suggesting enhanced conductivity at the interfaces between the charge transport layer and metal electrodes.

Chapter 3 Bandgap reduction of bismuth tri-iodide via cesium tin iodide addition

Figure 2. Images of sample and schematic of bandgap narrowing

Chapter 3 discusses the effect of adding a mixture of CsI and SnI_2 (CTI: cesium tin iodide) on bismuth tri-iodide (BiI_3), using different volumes (10, 25, and 50 μl) of CTI with 150 μl of BiI_3. The successful incorporation of CTI into BiI_3 was confirmed by X-ray Photoelectron Spectroscopy (XPS) analysis. The addition of 25 μl of CTI showed the best morphology with a grain size value of 173 nm. In addition, by adding 25 μl of CTI, the bandgap value was successfully reduced from 1.75 eV to 1.41 eV, which is in the ideal range of 1.1-1.4 eV needed for optimal solar cell efficiency. This bandgap narrowing is attributed to the interaction of Sn 5s orbitals with Bi 6p and I 5p orbitals, which lowers the valence band maximum position. Furthermore, adding 25 μl of CTI to BiI_3 significantly improved the power conversion efficiency to 0.113%, which is higher than the 0.031% efficiency of pure BiI_3.

Chapter 4 Optimizing $Cs_2AgBiBr_6$ perovskite solar cell via p-i-n and n-i-p device structure and substrate heating treatment

Figure 3. Device structure and performance of the device

In Chapter 4, the double perovskite $Cs_2AgBiBr_6$ was successfully fabricated using a low-temperature annealing process at 140°C. The phase purity of the films was confirmed by XRD, and scanning electron microscopy (SEM) analysis showed a grain size of 151 nm. Photoluminescence (PL) measurements indicated that the carrier lifetime of the double perovskite $Cs_2AgBiBr_6$ film was approximately 6 ns. To further improve the film quality, substrate heating treatment was applied, which increased the grain size to 246 nm and enhanced the carrier lifetime to approximately 10 ns. Finally, the PSC devices were fabricated in both p-i-n and n-i-p architectures, achieving power conversion efficiencies of 0.053% and 0.460%, respectively.

Chapter 5 Conclusion

Overall, this thesis proposed a combined effort to improve both the structural, optical, and electrical properties of bismuth-based solar cells, supporting their potential as a safe and sustainable alternative for photovoltaic applications. While the results show the promise of bandgap engineering in enhancing device efficiency, the overall performance is still limited. To further advance this research, future studies include advanced computational modelling such as density functional theory (DFT), selection of hole transport layer and electron transport layer, and deeper investigations into carrier lifetime are necessary to provide more comprehensive insights and complement this research.

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論文審査結果の要旨 (代替)

申請者氏名　ADITYA WAHYU ANUGRAH

Summary of Doctoral Thesis

Title of Doctoral Thesis: Studies on eco-friendly bismuth-based material for lead-free perovskite solar cell applications (鉛フリーペロブスカイト太陽電池応用に向けた環境配慮型ビスマス系材料に関する研究)

Name: Aditya Wahyu Anugrah

Summary of Doctoral Thesis:

Solar cell photovoltaic (PV) technology is projected to become the leading global renewable energy source, with future solar cells needing to be safe, stable, and environmentally friendly. Bismuth-based materials offer a promising alternative to toxic lead-based perovskites, due to their non-toxic, low-cost, stable, and environmentally friendly properties. However, their power conversion efficiency remains limited due to challenges such as poor film morphology, low carrier lifetime, and especially a large bandgap, which limits their effectiveness as light-absorbing layers. Various doping strategies have been explored to narrow the bandgap, such as doping $Cs_2AgBiBr_6$ with Sb^{3+} and Ru^{3+} to reduce the bandgap to approximately 1.86 eV and 1.80 eV, respectively, and sulfur doping in MA_3Bi2I_9 to achieve a bandgap of 1.67 eV. However, the achieved bandgap is still higher than the ideal range of 1.1-1.4 eV required for optimal solar cell efficiency, and the impact of bandgap narrowing on overall device performance is still not fully understood.

This thesis aims to address these challenges through post-annealing treatments to improve film quality and determine the optimal annealing temperature of bismuth-based material which explained in Chapter 2. This optimized temperature annealing is then applied in Chapter 3, where further explore effective doping strategies to reduce the bandgap and enhance solar cell efficiency. To address the issue of low carrier lifetime, double perovskite bismuth-based material was employed, which offers relatively higher carrier lifetimes as discussed in Chapter 4. The double perovskite bismuth-based solar cells were fabricated using a low-temperature process, and a simple substrate heating treatment was proposed to improve its film morphology and enhance the carrier lifetime.

Chapter 2 presents post-annealing treatments at various temperatures from 25^{o}C to 180^{o}C were performed to optimize the morphology and optical properties of thin films. The treatment induced lattice structural changes, as confirmed by X-ray diffraction measurement. The best grain size was shown by post-annealing at 140^{o}C. An improvement of film quality was also evaluated by impedance spectroscopy analysis, which showed that post-annealing treatment reduced internal resistance, suggesting enhanced conductivity at the interfaces between the charge transport layer and metal electrodes. Photovoltaic performance evaluated through current density-voltage (*J–V*) measurements under standard Air Mass (AM1.5) one-sun conditions showed an improvement of power conversion efficiency from 0.074% to 0.139% with post-annealing treatment.

Chapter 3 discusses the effect of adding a mixture of CsI and SnI_2 (CTI: cesium tin iodide) on bismuth tri-iodide (BiI_3), using different volumes (10, 25, and 50 μl) of CTI with 150 μl of BiI_3. The successful incorporation of CTI into BiI_3 was confirmed by X-ray photoelectron spectroscopy analysis. The addition of 25 μl of CTI showed the best morphology with a grain size value of 173 nm. In addition, by adding 25 μl of CTI, the bandgap value was successfully reduced from 1.75 eV to 1.41 eV, which is in the ideal range of 1.1-1.4 eV needed for optimal solar cell efficiency. This bandgap narrowing is attributed to the interaction of Sn *5s* orbitals with Bi *6p* and I *5p* orbitals, which lowers the valence band maximum position. Furthermore, adding 25 μl of CTI to BiI_3 significantly improved the power conversion efficiency to 0.113%, which is higher than the 0.031% efficiency of pure BiI_3.

In Chapter 4, the double perovskite $Cs_2AgBiBr_6$ was successfully fabricated for the first time using a low-temperature annealing process at 140^{o}C. The phase purity of the films was confirmed by X-ray diffraction, and scanning electron microscopy analysis showed a grain size of 151 nm. Photoluminescence measurements indicated that the carrier lifetime of the double perovskite $Cs_2AgBiBr_6$ film was approximately 6 ns. To further improve the film quality, substrate heating treatment was applied, which increased the grain size to 264 nm and enhanced the carrier lifetime to approximately 10 ns. Finally, the solar cell devices were fabricated in both p-i-n and n-i-p architectures, achieving power conversion efficiencies of

0.053% and 0.460%, respectively.
Overall, this thesis proposed a combined effort to improve both the structural, optical, and electrical properties of bismuth-based solar cells, supporting their potential as a safe and sustainable alternative for photovoltaic applications.

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