高効率プラナー構造のペロブスカイト太陽電池 CH₃NH₃PbI_{3-x}Cl_xにおける塩素イオン の決定

High Efficiency Planar CH₃NH₃PbI_{3-x}Cl_x based solar cells with determined amount of chloride ion

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In recent years, organometal halide perovskites (CH₃NH₃PbI₃) have become a new attractive photovoltaic material due to their remarkable proprieties and impressive power conversion efficiencies. In a very short time, perovskite-based solar cells as it has shown a continuous rise in cell power conversion efficiency, from less than 4% in 2009¹ to over 20% in 2015² for both meso- and planar architectures. The principal perovskite layer in meso and planar architecture is usually composed of either pure iodide (CH₃NH₃PbI₃) or mixed halide (CH₃NH₃PbI_{3-x}Cl_x) perovskite. Chemical formula of CH₃NH₃PbI_{3-x}Cl_x is being typically used for showing that a small amount of chloride salts remains in the device. According to several group chloride ions help in form high quality perovskite film, enhance the coverage, morphology, stability of the final perovskite film and CH₃NH₃PbI_{3-x}Cl_x has been often used to fabricate planar architecture devices with high power conversion efficiency (PCE). In such heterojunction structures the all interfacial connections play an important role in improving the cell performance. We have investigated alternative methods of interface engineering of compact TiO₂ layer, by using TiCl₄ and UV(O₃) treatment. TiCl₄ and UV(O₃) treatment of the compact TiO₂ layer imparted a positive effect on the cell performance, efficiency improved from 13% to over 17% (Fig.1) by the treatment. Additionally we will discuss about the amount of chloride content in the final product perovskite film. In order to determine the molar ratio of Cl and I in the final perovskite product and to know exact formula of CH₃NH₃PbI_{3-x}Cl_x, we employed the ion chromatography method. This method is believed to be a more direct and appropriate method, as compared to indirect estimation from optical properties, XRD, SEM-EDS and XPS analysis.

For analysis we prepared three different perovskite films, then dissolved in deionized water and sulfuric acid and analyzed using ion chromatography method. Considering that all the chloride ions are part of the crystal, the formula can be $CH_3NH_3PbI_{2.94}Cl_{0.06}$. Chloride anions may help in forming high quality iodide perovskite by assisting growth of the final product. In the optimal conditions, we succeeded in fabricating pure iodide perovskite solar cells that exhibited PCE as high as 17.3%.

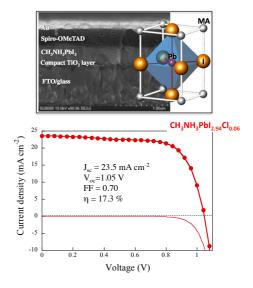


Figure 1. J-V curves of the best performing planar heterojunction perovskite solar cell. 3,4

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