

Pb and Pb-free Perovskite Absorbers Synthesized by Reactive Polyiodide Melt Method Applied to Photovoltaics

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Introduction

Based on the strategic advantages of “reactive polyiodide melts” method (RPM), which was jointly developed by researchers from AIST, MSU and EPFL published in Nature Nanotechnology [1], we aim at synthesizing low-toxicity Pb-free materials in order to solve the problem of toxic Pb-based Perovskite materials.

Goals

We will describe the “reactive polyiodide melt” method (RPM) and its application to Pb-based Perovskite solar cells (PVs). Then, we will demonstrate that this approach is also suitable to synthesize Pb-free materials with either Perovskite or non-Perovskite crystal structure. Our long-term goal is to fabricate low-toxicity Pb-free PVs.

Pb-based perovskite materials

Concept

1st step Pb film
2nd step MAI film
3rd step RMP method I₂ vapor (Iodization) MAPbI₃ film

Solar Cells

3rd step RMP method I₂ vapor (Iodization) MAPbI₃ film

MAI film
Pb film
substrate

MAI (~250 nm)
Pb (~60 nm)
FTCO
Glass

Iodization Reaction

MAPbI₃ (~550 nm)
FTCO
Glass

405 nm, 975 nm, 545 nm

time=1 final
Intensity (a.u.)
2theta (degrees)
Intensity (n.u.)
time=0 initial

In-situ XRD measurements during conversion of the Pb/MAI stoichiometric bilayer into MAPbI₃ perovskite film.

1) Pb and MAI stacked films in stoichiometry ratio were deposited sequentially by vacuum (we are already investigating die-coating, dip-coating, spray... on glass, PET substrate...)
2) Controlled iodization of MAI to form MAI₂ in-situ, which directly converts Pb into MAPbI₃ → fast and complete reaction, which lead to uniform, large grains, pin-holes free films

Pb-based MAPbI₃ and (Cs,FA,MA)PbI₃ perovskite PVs

efficiency 1.7%

efficiency 16%

MAPbI₃ based solar cells

Microporous structure
Planar structure

Deposition of Pb/Cs/FA/MAI layers
Conversion in I₂ vapor
XPS analysis suggests Cs_{0.12}MA_{0.88}FA_{0.83}PbI_{2.99}

efficiency 16%

Planar structure

Hysteresis ratio H(hv) = (j_{sc} - j_{sc}^0) / j_{sc}

Pb-free perovskite and non-perovskite materials

Towards the discovery and the synthesis of low-toxicity Pb-free materials

- Synthesis of low-toxicity Pb-free materials
 $X + MAI + I_2 \rightarrow MAXI_3$ or non-Perovskite
where X is an element of the periodic table or alloys
- Selection criteria:
Direct optical band gap ~1.5eV
High electron and hole mobility ~10 cm²(V.s)
Low electron and hole effective mass
Stable 3-dimensional Crystal structure
- Source of information:
Material databases
Theoretical calculations

Extending our concept to various metals

$Bi/3MAI + 3I_2 \rightarrow MA_3BiI_9$ {0D}

XRD
RPM process
Solvent process

Conclusions

We have demonstrated that “reactive polyiodide melt” method (RPM) is suitable:

- To fabricate of Pb-based Perovskite solar cells with efficiency as high as 17%
- To synthesize Bi-based materials and to explore a wide range of Pb-free materials

Our long-term goal is to discover, synthesize and fabricate low-toxicity Pb-free PVs.

References and Acknowledgements

References:
[1] “Strategic advantages of reactive polyiodide melts for scalable perovskite photovoltaics” I. Turkevych, S. Kazaoui, N. A. Belich, A. Y. Grishko, S. A. Fateev, A. A. Petrov, T. Urano, S. Aramaki, S. Kosar, M. Kondo, E. A. Goodilin, M. Grätzel, A. B. Tarasov Nature Nanotechnology 14, 57-63 (2019).

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