Electronic Supplementary Information

Antisolvent Diffusion-induced Growth, Equilibrium Behaviours in Aqueous Solution and Optical Properties of $CH_3NH_3PbI_3$ Single Crystals for Photovoltaic Applications

Huawei Zhou[a], Zhonghao Nie[a], Jie Yin[b], Yuanwei Sun[a], Hongyan Zhuo[a], Da-qi Wang[a], Dacheng Li[a] ,Jianmin Dou [a], Xianxi Zhang [a*], Tingli Ma[c]

[a]*Shandong Provincial Key Laboratory of Chemical Energy Storage and Novel Cell Technology, School of Chemistry and Chemical Engineering, Liaocheng University, Liaocheng 252059, China.*
[b]*College of Materials Science and Engineering, Liaocheng University, Liaocheng 252059, Shandong, China*
[c]*Graduate School of Life Science and Systems Engineering, Kyushu Institute of Technology, 2-4 Hibikino, Wakamatsu, Kitakyushu, Fukuoka 808-0196, Japan*
** Corresponding authors: zhangxianxi@lcu.edu.cn;*

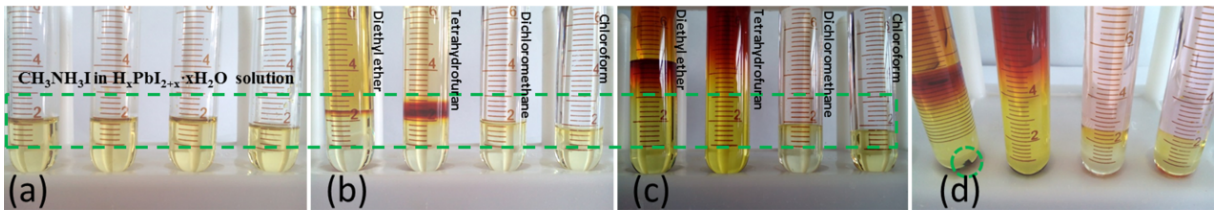

Figure S1. Photograph: (a) 40 mg CH_3NH_3I in 1.82 ml (0.1371 mol L^{-1}) $H_xPbI_{2+x}\cdot xH_2O$ solution for single crystal growth, (b) four solvent (diethyl ether, tetrahydrofuran, dichloromethane, chloroform) was inflowed into perovskite precursors, (c) four solvent was inflowed into perovskite precursors after 2 days, (d) different angle view of (c).

Single Crystal XRD data of $CH_3NH_3PbI_3$

TITL 150421b in I4/mcm

CELL 0.71073 8.8719 8.8719 12.6770 90.000 90.000 90.000

ZERR 1 0.0015 0.0015 0.0040 0.000 0.000 0.000

LATT 2

SYMM - Y , X , Z

SYMM - X , - Y , Z

SYMM Y , - X , Z

SYMM X , - Y , 0.50000 - Z

SYMM Y , X , 0.50000 - Z

SYMM - X , Y , 0.50000 - Z

SYMM - Y , - X , 0.50000 - Z

SFAC C H N O Cl I Pb

UNIT 20 40 6 5 1 1 1

OMIT -1.00 50.04

L.S. 8

EXTI 0.00891

ACTA

BOND $H

FMAP 2

PLAN 20

WGHT 0.200000 0.000000

FVAR 0.171190

TEMP 25

Pb1 7 0.000000 0.000000 0.000000 10.125000 0.023370 0.023370 =
0.000010 0.000000 0.000000 0.000000

I1 6 0.214714 0.285286 0.000000 10.250000 0.099700 0.099700 =
0.047430 0.000000 0.000000 -0.073430

```
I2        6     0.000000    0.000000    0.250000 10.125000    0.116520    0.116520 =
                0.000010    0.000000    0.000000    0.000000
Q1        1     0.084900 -0.088500    0.046800 11.000000    0.050000          6.38
Q2        1     0.342600    0.157400    0.051400 10.500000    0.050000          5.81
Q3        1     0.000000    0.000000    0.102000 10.250000    0.050000          5.51
Q4        1     0.129500    0.370500    0.051500 10.500000    0.050000          5.16
Q5        1     0.310600    0.352000    0.040100 11.000000    0.050000          4.32
Q6        1     0.000000    0.500000    0.750000 10.125000    0.050000          4.27
Q7        1     0.306900    0.448100    0.000000 10.500000    0.050000          3.10
Q8        1     0.250000    0.250000    0.250000 10.250000    0.050000          2.53
Q9        1     0.197600    0.000000    0.250000 10.500000    0.050000          2.20
Q10       1     0.116600    0.190000    0.249400 11.000000    0.050000          2.11
Q11       1     0.096000    0.059700    0.297600 11.000000    0.050000          2.10
Q12       1     0.131100    0.521900    0.000000 10.500000    0.050000          1.94
Q13       1    -0.046400    0.335700    0.000000 10.500000    0.050000          1.90
Q14       1     0.213000    0.287000    0.109900 10.500000    0.050000          1.82
Q15       1     0.333300    0.166700    0.199600 10.500000    0.050000          1.69
Q16       1    -0.028900    0.269700    0.120600 11.000000    0.050000          1.68
Q17       1     0.410700    0.089300    0.199300 10.500000    0.050000          1.57
Q18       1     0.085300    0.090900    0.131100 11.000000    0.050000          1.51
Q19       1     0.179200    0.004200    0.107300 11.000000    0.050000          1.37
Q20       1     0.000000    0.500000    0.000000 10.125000    0.050000          1.36
HKLF 4 1 -1.0000     0.0000     0.0000     0.0000 -1.0000     0.0000 -1.0000 -1.0000
2.0000
END
```

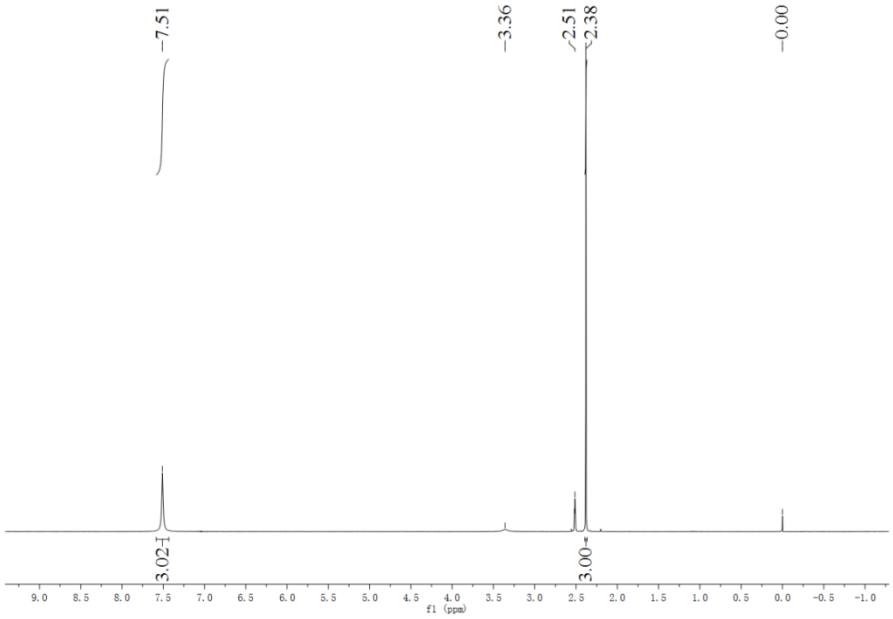

Figure S2. NMR of CH_3NH_3I