# Electronic Supplementary Information 

# Enhanced Yield-Mobility Products in Hybrid Halide Ruddlesden-Popper Compounds with Aromatic Ammonium Spacers 

Naveen R. Venkatesan, ${ }^{\text {ab }}$ Ali Mahdi, ${ }^{\text {bc }}$ Brian Barraza, ${ }^{c}$ Guang Wu, ${ }^{\text {c }}$ Michael L. Chabinyc, ${ }^{* a b}$ and Ram Seshadri ${ }^{*}{ }^{*} c$

${ }^{\text {a }}$ Materials Department, University of California, Santa Barbara, CA 93106, United States
b Materials Research Laboratory, University of California, Santa Barbara, CA 93106, United States
c Department of Chemistry and Biochemistry, University of California, Santa Barbara, CA 93106, United States
*Corresponding Authors: mchabinyc@engineering.ucsb.edu
seshadri@mrl.ucsb.edu

## S1. Single-crystal X-ray diffraction patterns

Single-crystal X-ray diffraction data was collected using a Bruker KAPPA APEX II diffractometer equipped with an APEX II CCD detector utilizing a TRIUMPH monochromator and a Mo-K $\alpha$ Xray source ( $\lambda=0.71703 \AA$ ). Diffraction images are shown below in Figures S1 and S2.


Figure S1. Sample X-ray diffraction image of (BPEA) $)_{2} \mathrm{PbI}_{4}$ obtained from grown single-crystals.


Figure S2. Sample X-ray diffraction image of $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} l_{7}$ obtained from grown singlecrystals.

## S2. Observed bond angles and distances in (BPEA) ${ }_{2} \mathrm{Pbl}_{4}$ and (BPEA) $)_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{I}_{7}$

Structural details of $(\mathrm{BPEA})_{2} \mathrm{Pbl}_{4}$ and $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{I}_{7}$ are tabulated below. Bond distances for the four equatorial and two axial iodides, along with all $\mathrm{I}-\mathrm{Pb}-\mathrm{I}$ and $\mathrm{Pb}-\mathrm{I}-\mathrm{Pb}$ (if applicable) bond angles are presented. For comparison, the bond lengths and angles from $(\mathrm{BA})_{2} \mathrm{Pbl}_{4}$ and $(\mathrm{BA})_{2}(\mathrm{MA}) \mathrm{Pb}_{2} 1_{7}$ are listed in Table S2. ${ }^{1}$

Table S1. Structural details of $(\mathrm{BPEA})_{2} \mathrm{Pbl}_{4}$ and $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{l}_{7}$
$(\mathrm{BPEA})_{2} \mathrm{PbI}_{4} \quad(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{I}_{7}$

## Bond Distances

| $\mathrm{Pb}-\mathrm{I}_{\text {axial (1) }}(\mathrm{A})$ | 3.2073(14) | 3.131(3) |
| :---: | :---: | :---: |
| $\mathrm{Pb}-\mathrm{Iaxial}_{\text {(2) }}(\mathrm{A})$ | 3.2073(14) | 3.2617(12) |
| $\mathrm{Pb}-\mathrm{I}_{\text {axial }}(3)(\mathrm{A})$ | N/A | 3.2617(12) |
| $\mathrm{Pb}-\mathrm{I}_{\text {axial ( }}(4)$ ( A$)$ | N/A | 3.131(3) |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (1) }}(\mathrm{A})$ | 3.108(5) | 3.141(4) |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (2) }}(\mathrm{A})$ | $3.174(5)$ | 3.179(3) |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (3) }}(\mathrm{A})$ | 3.222(5) | $3.191(4)$ |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (4) }}(\mathrm{A})$ | 3.154(5) | $3.157(4)$ |
|  | Bond Angles |  |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{laxial}_{\text {(1) }}\left({ }^{\circ}\right)$ | 176.4(3) | 176.38(9) |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{laxial}^{(2)}\left({ }^{\circ}\right)$ | N/A | 176.38(9) |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{l}_{\text {equatorial (1) }}\left({ }^{\circ}{ }^{\text {a }}\right.$ | 179.25(14) | 176.31(8) |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{l}_{\text {equatorial (2) }}\left({ }^{\circ}\right.$ ) | 178.18(16) | 176.28(9) |
| $\mathrm{Pb}-\mathrm{I}-\mathrm{Pb}_{\text {axial }}\left({ }^{\circ}\right.$ ) | N/A | 179.2(3) |
| $\mathrm{Pb}-\mathrm{I}-\mathrm{Pb}_{\text {equatorial }}\left({ }^{\circ}\right.$ ) | 152.49(15) | 154.52(11) |

Table S2. Structural details of $(\mathrm{BA})_{2} \mathrm{PbI}_{4}$ and $(\mathrm{BA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} l_{7}$ taken from reference 1.
$(\mathrm{BA})_{2} \mathrm{PbI}_{4} \quad(\mathrm{BA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{I}_{7}$
Bond Distances

|  | Bond Distances |  |
| :---: | :---: | :---: |
| $\mathrm{Pb}-\mathrm{Iaxial}_{\text {(1) }}(\mathrm{A})$ | 3.201(16) | 3.08(3) |
| $\mathrm{Pb}-\mathrm{Iaxial}_{\text {(2) }}(\mathrm{A})$ | 3.213(13) | 3.28(3) |
| $\mathrm{Pb}-\mathrm{Iaxial}_{\text {( } 3 \text { ( }}(\mathrm{A})$ | N/A | 3.25(3) |
| $\mathrm{Pb}-\mathrm{Iaxial}_{\text {(4) }}(\mathrm{A})$ | N/A | 3.08(3) |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (1) }}(\mathrm{A})$ | 3.164(7) | $3.171(4)$ |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (2) }}(\mathrm{A})$ | 3.212(8) | $3.169(5)$ |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial ( } 3 \text { ( }}(\mathrm{A})$ | $3.191(6)$ | $3.169(5)$ |
| $\mathrm{Pb}-\mathrm{l}_{\text {equatorial (4) }}(\mathrm{A})$ | 3.160(7) | 3.171(4) |
|  | Bond Angles |  |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{Iaxial}_{\text {(1) }}\left({ }^{\circ}\right)$ | 178.6(3) | 177.3(3) |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{laxial}_{\text {(2) }}\left({ }^{\circ}\right)$ | N/A | 177.3(3) |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{I}_{\text {equatorial (1) }}\left({ }^{\circ}\right.$ ) | 179.2(5) | 175.5(9) |
| $\mathrm{I}-\mathrm{Pb}-\mathrm{I}_{\text {equatorial (2) }}\left({ }^{\circ}\right.$ ) | 179.1(5) | 175.5(9) |
| $\mathrm{Pb}-\mathrm{I}-\mathrm{Pb}_{\text {axial }}\left({ }^{\circ}\right.$ ) | N/A | 165.64(16) |
| $\mathrm{Pb}-\mathrm{I}-\mathrm{Pb}_{\text {equatorial }}\left({ }^{\circ}\right.$ ) | 155.1(3) | 164.2(10) |

## S3. Qualitative comparison of $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{I}_{7}$ with isotropic and textured XRD simulations

The single-crystal structures of $(\mathrm{BPEA})_{2} \mathrm{PbI}_{4}$ and $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} l_{7}$ were used to simulate powder diffraction patterns and compared with actual measurements. Figure S 3 shows the comparison of $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{l}_{7}$ to a completely isotropic, untextured simulation of powder diffraction, showing poor agreement (isotropic (BPEA) $)_{2} \mathrm{PbI}_{4}$ is presented as the simulation in Figure 2 of the main manuscript). When texturing along the layer stacking direction ([010]) is added, the agreement between simulation and experiment is clear.


Figure S3. Powder X-ray diffraction of synthesized powder of $(B P E A)_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} 1_{7}$ compared to simulated diffraction patterns of an isotropic sample and a [010]-textured sample, illustrating structural agreement with the latter.

## S4. PXRD of aged powders compared to $\mathrm{Pbl}_{2}$

The aged ( 5 weeks in ambient) powders of $(\mathrm{BPEA})_{2} \mathrm{Pbl}_{4}$ and $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} \mathrm{l}_{7}$ were compared to the expected diffraction pattern of $\mathrm{Pbl}_{2}$ to show that there was no material degradation after exposure to ambient conditions.


Figure S4. Powder X-ray diffraction patterns of $(\mathrm{BPEA})_{2} \mathrm{Pbl}_{4}$ and $(\mathrm{BPEA})_{2}\left(\mathrm{CH}_{3} \mathrm{NH}_{3}\right) \mathrm{Pb}_{2} l_{7}$ powders aged for five weeks in ambient conditions, along with simulated diffraction pattern of $\mathrm{Pbl}_{2}$. Comparison with the simulated pattern illustrates no degradation of Ruddlesden-Popper phases into lead iodide.

## References

1 C. C. Stoumpos, D. H. Cao, D. J. Clark, J. Young, J. M. Rondinelli, J. I. Jang, J. T. Hupp and M. G. Kanatzidis, Chem. Mater., 2016, 28, 2852-2867.

