

## Supplementary Information

Simple one-step synthesis of a two-dimensional perovskite consisting of perfluoroalkyl-based ammonium spacers using acetone as the solvent

Naoyuki Nishimura <sup>a\*</sup>, Masahiro Tojo <sup>b</sup>, Yuko Takeoka <sup>c\*</sup>

<sup>a</sup> Marketing & Innovation, Asahi Kasei Corporation, 2-1 Samejima, Fuji, Shizuoka, 416-8501, Japan.

Email: nishimura.nj@om.asahi-kasei.co.jp

<sup>b</sup> Chemistry & Chemical Process Laboratory, Asahi Kasei Corporation, 2767-11 Niihama, Shionasu, Kojima, Kurashiki, Okayama, 711-8510, Japan

<sup>c</sup> Faculty of Science and Engineering, Sophia University, 7-1 Kioi-cho, Chiyoda-ku, Tokyo, 102-8554, Japan. E-mail: y-tabuch@sophia.ac.jp

## Experimental

### 1. Synthesis of $C_3F_7CH_2NH_3Br$

$C_3F_7CH_2NH_3Br$ , the precursor to the perovskite  $(C_3F_7CH_2NH_3)_2PbBr_4$ , was synthesized via a reaction between 1*H*,1*H*-perfluorobutylamine ( $C_3F_7CH_2NH_2$ ) and hydrogen bromide (HBr). In this process, aqueous HBr (48 wt%, Wako Chemical) was gradually added to an aqueous solution of  $C_3F_7CH_2NH_2$  (>95%, TCI) cooled with ice over a time span of 1 h under a nitrogen atmosphere, followed by continuous stirring of the cooled solution for a further two hours. The water was subsequently evaporated under vacuum and the resulting powder was recrystallized from methanol and then washed with diethyl ether. Elemental analysis of the synthesized  $C_3F_7CH_2NH_3Br$  was carried out (Table S1) and the results were close to the expected theoretical values for  $C_3F_7CH_2NH_3Br$ .

Table S1. Results from elemental analysis of  $C_3F_7CH_2NH_3Br$ .

	C (%)	H (%)	N (%)
Theoretical	17.2	1.8	5.0
Experimental	17.2	1.5	5.0

## 2. *Synthesis of perovskite (C<sub>3</sub>F<sub>7</sub>CH<sub>2</sub>NH<sub>3</sub>)<sub>2</sub>PbBr<sub>4</sub> films*

Samples of the 2D perovskite (C<sub>3</sub>F<sub>7</sub>CH<sub>2</sub>NH<sub>3</sub>)<sub>2</sub>PbBr<sub>4</sub> were synthesized via the simple spin-coating on substrates with precursor solutions containing C<sub>3</sub>F<sub>7</sub>CH<sub>2</sub>NH<sub>3</sub>Br and PbBr<sub>2</sub>. These perovskite precursor solutions were prepared by dissolving C<sub>3</sub>F<sub>7</sub>CH<sub>2</sub>NH<sub>3</sub>Br at 0.266 M and PbBr<sub>2</sub> at 0.133 M in acetone or DMF. In the case of the acetone-based synthesis process, the precursor solution was cast onto a glass substrate at room temperature with spin-coating at 1000 rpm for 30 s, followed by drying at room temperature. In trials using DMF as the solvent, the solution was heated at 353 K, spin-coated onto the substrate at 2000 rpm for 30 s, and subsequently heated at 313 K or 393 K for 5 min.

## 3. *Synthesis of a perovskite (C<sub>4</sub>H<sub>9</sub>NH<sub>3</sub>)<sub>2</sub>PbBr<sub>4</sub> film as a reference sample*

A sample of (C<sub>4</sub>H<sub>9</sub>NH<sub>3</sub>)<sub>2</sub>PbBr<sub>4</sub>, a conventional 2D perovskite, was synthesized as a reference for comparison with the (C<sub>3</sub>F<sub>7</sub>CH<sub>2</sub>NH<sub>3</sub>)<sub>2</sub>PbBr<sub>4</sub>. The precursor C<sub>4</sub>H<sub>9</sub>NH<sub>3</sub>Br was synthesized in the same manner as the C<sub>3</sub>F<sub>7</sub>CH<sub>2</sub>NH<sub>3</sub>Br and the elemental analysis results for this material are provided in Table S2. A solution of this precursor in DMF at a concentration of 0.266 M together with PbBr<sub>2</sub> at 0.133 M was heated to 353 K, spin-coated at 2000 rpm for 30 s at room temperature, then heated at 393 K for 5 min. Figure S1 presents the XRD pattern for the as-prepared sample, which confirms the formation of single-phase (C<sub>4</sub>H<sub>9</sub>NH<sub>3</sub>)<sub>2</sub>PbBr<sub>4</sub> with a layered structure. The estimated *d* value of 13.7 Å for this material is similar to a previously reported value (*d* = 14.0 Å).<sup>1</sup>

Table S2. Results from the elemental analysis of  $C_4H_9NH_3Br$ .

	C (%)	H (%)	N (%)
Theoretical	31.2	7.9	9.1
Experimental	31.3	7.6	9.1

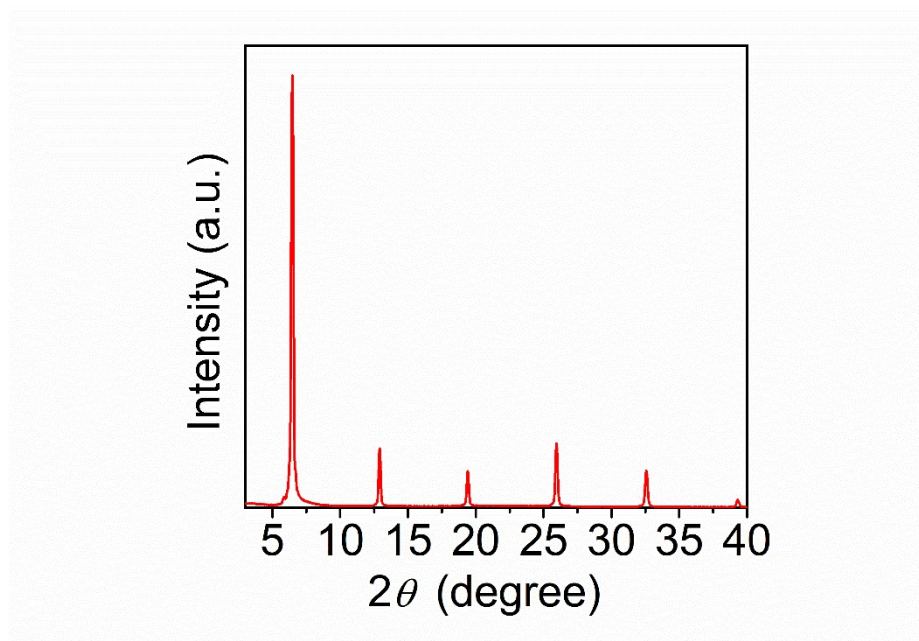


Figure S1. The XRD pattern of the synthesized  $(C_4H_9NH_3)_2PbBr_4$  film ( $d = 13.7 \text{ \AA}$ ).

#### 4. Characterization

The  $^1\text{H}$  NMR spectra of the precursors were acquired using a 300 MHz instrument (Jeol) and elemental analyses were carried out with a PE2400-II instrument (Perkin Elmer Inc.) at 1148 K. XRD patterns of the as-prepared samples were obtained with an X-ray diffractometer (Rint2000, Rigaku) in conjunction with a Ni-filtered Cu  $K\alpha$  target. The optical absorption spectra of the spin-coated films were obtained with a UV-vis-NIR spectrophotometer (UV-3100PC, SHIMADZU) at room temperature. Photoluminescence analyses were performed using an F-4500 fluorescence spectrometer (Hitachi) equipped with a Xe lamp and a monochromatic filter. The XRD, the absorption spectra, and the photoluminescence measurement were employed in air.

#### Reference

1. Y. Takeoka, K. Asai, M. Rikukawa, K. Sanui, *Bull. Chem. Soc. Jpn.*, 2006, **79**, 1607-1613.