# Supporting Information

## Enhanced hole extraction by electron-rich alloys in all-inorganic $CsPbBr_3$ perovskite solar cells

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## Experimental section

### Preparation of Pt and $Pt_3M$ Alloys

Four kind of solutions marked as A, B, C and D were obtained by dissolving $H_2PtCl_6·6H_2O$, $NiCl_2·6H_2O$, $CoCl_2·6H_2O$ and $FeCl_2·4H_2O$ in deionized water with concentration of 38.6 mM, respectively. 3 mL of A, 3 mL of A and 1 mL of B, 3 mL of A and 1 mL of C, 3 mL of A and 1 mL of D were added in four different beakers respectively, then 8 mL of hydrazine hydrate and 35 mL of deionized water were added. The solution was subsequently transferred to a poly(tetrafluoroethylene) (Teflon)-lined autoclave and heated at 120 °C for 12 h. After cooling to room temperature naturally, the product was centrifuged, washed, vacuum dried and waiting to be used.

### Preparation of Modified Carbon Paste

Pt or $Pt_3M$ alloys incorporated carbon pastes with dosages of 2.5 wt% were synthesized by blending Pt or $Pt_3M$ alloys into the commercial carbon paste and ball-milling for 12 h.

### Preparation of $TiO_2$ Colloid

$TiO_2$ colloid was prepared by the sol-hydrothermal method. First, 10 mL of titanium tetrabutanolate was added to 100 mL of deionized water under continuous stirring to obtain the white powder. Then, 0.8 mL of nitric acid and 10 mL of acetic acid were dripped into the prepared powder under stirring. Subsequently, 160 mL of deionized water was added and continued to be stirred for 25 min. When the reactants became translucent, the hydrothermal reaction was carried out at 200 C for 12 h. Then, 0.4 g of commercial P25 was added into the product, sonicated for 30 min, and then reacted at 200 $^{o}$C for12 h. Finally, 1 mL of OP emulsifier and 0.8 g of polyethylene glycol (PEG 20000) were introduced into the aforementioned colloid, and then concentrated to 40 mL at 80 $^oC$ to obtain the $TiO_2$ colloid.

### Fabrication of $CsPbBr_3$ PSC

All the fabrication processes of $CsPbBr_3$ PSC was performed in air atmosphere instead of under humidity-controlled conditions. The fluorine-doped tin oxide (FTO) substrates were patterned by etching with Zn powder and diluted HCl and then cleaned by sequential ultrasonic treatment in acetone, alcohol and deionized water each for 30 min. The FTO substrates were cleaned by plasma for 5 min before use. The mixed ethanolic solution of 0.5 M diethanolamine and 0.5 M titanium isopropoxide were deposited on the FTO substrate (30 s, 7000 rpm), followed by annealing in air at 500 $^oC$ for 2 h to obtain the compact $TiO_2$ ($c$-$TiO_2$) layer. The mesoscopic $TiO_2$ ($m$-$TiO_2$) layer was deposited by spin-coating the $TiO_2$ colloid (30 s, 2000 rpm) and annealing it in air at 500 $^oC$ for 30 min. Subsequently, the $TiO_2$ film was immersed in 0.04 M $TiCl_4$ aqueous solution at 70 $^oC$ for 30 min and rinsed with deionized water and ethanol, and then annealed at 450 $^oC$ for 30 min to obtain the $TiO_2$ photoanode. The perovskite film was fabricated by the multi-step solution-processed technique. Firstly, 1 M $PbBr_2$ in DMF solution was spin-coated on FTO/$c$-$TiO_2$/$m$-$TiO_2$ and annealed at 80 $^oC$ for 30 min. Then, CsBr in methanol solution was spin-coated on the $PbBr_2$ film (30 s, 2000 rpm) and annealed at 250 $^oC$ for 5 min. This step was repeated 7 times to prepare the uniform and dense $CsPbBr_3$ perovskite film. Finally, the conductive carbon slurry was coated on the $CsPbBr_3$ film by the scraper technique and then heated at 90 $^oC$ for 10 min.

### Tests and Characterizations

The $J$-$V$ curves of solar cells were recorded on a CHI660E electrochemical workstation with scanning speed of 0.1 V $s^{-1}$ under irradiation of simulated solar light intensity controlled at AM 1.5 (100 mW $cm^{-2}$). The long-term stability of all-inorganic PSC was performed in air atmosphere in a temperature and humidity chamber (LHS-100CHG, keelrein Instrument Co., Ltd). The temperature and humidity were controlled at 25 $^{o}$C and 80%RH, respectively. The PCE values were obtained by measuring the *J-V* curves of test device at an interval of one day. The surface morphologies of the prepared films were characterized by a field emission scanning electron microscopy (FESEM, SU8010, Hitachi). The steady-state photoluminescence was obtained at room temperature by FLS920 all functional fluorescence spectrometer. The Time-resolved PL measurement was carried out using time-resolved fluorescence (Horiba Jobin Yvon, FL). Ultraviolet photoemission spectroscopy measurements (UPS) were characterized on a Kratos AXIS ULTRA system with ahelium discharge lamp, a concentric hemispherical analyzer for photo-excited electron detection. The incident-photo-tocurrent conversion efficiency (IPCE) was characterized by a power source (Newport 300W xenon lamp, 66920) with amonochromator (Newport Cornerstone 260) in the 300-850 nm wavelength range at room temperature.

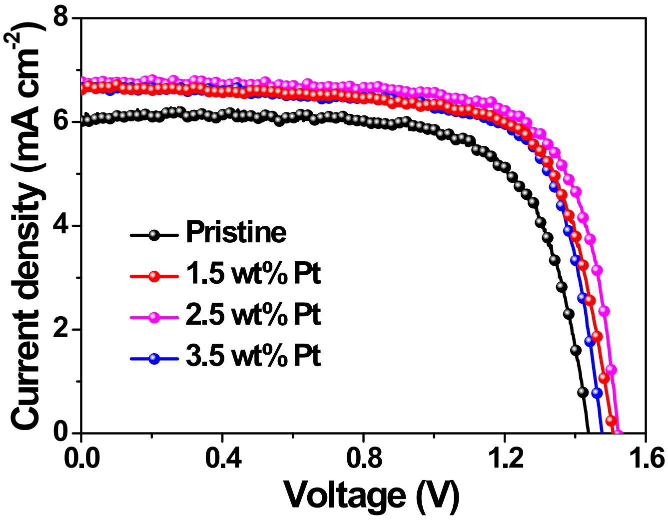


**Figure S1.** *J-V* curves of $CsPbBr_3$ PSC based on carbon electrode modified with Pt.

**Table S1** Photovoltaic data for all-inorganic $CsPbBr_3$ PSC based on Pt tairlored carbon electrode.

| Carbon electrode | $J_{sc}$ (mA cm$^{-2}$) | $V_{oc}$ (V) | *FF* (%) | PCE (%) |
|---|---|---|---|---|
| pristine | 6.01 | 1.435 | 72.70 | 6.27 |
| 1.5 wt% Pt | 6.68 | 1.478 | 72.93 | 7.20 |
| 2.5 wt% Pt | 6.77 | 1.520 | 74.05 | 7.62 |
| 3.5 wt% Pt | 6.72 | 1.508 | 73.22 | 7.42 |

**Table S2** Photovoltaic data for various all-inorganic $CsPbBr_3$ PSCs.

| Carbon electrodes | $J_{sc}$ (mA cm$^{-2}$) | $V_{oc}$ (V) | $FF$ (%) | PCE (%) |
|---|---|---|---|---|
| pristine | 6.01 | 1.435 | 72.70 | 6.27 |
| 2.5 wt% Pt | 6.77 | 1.520 | 74.05 | 7.62 |
| 2.5 wt% $Pt_3Fe$ | 6.99 | 1.541 | 76.68 | 8.26 |
| 2.5 wt% $Pt_3Ni$ | 7.08 | 1.562 | 78.22 | 8.65 |
| 2.5 wt% $Pt_3Co$ | 7.24 | 1.574 | 79.68 | 9.08 |

**Table S3** Comparison of photovoltaic data for state-of-the-art $CsPbBr_3$ PSCs.

| Devices | $J_{sc}$ (mA cm$^{-2}$) | $V_{oc}$ (V) | FF (%) | PCE (%) | Stability | Ref. |
|---|---|---|---|---|---|---|
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/$Pt_3Co$-carbon | 7.24 | 1.574 | 79.68 | 9.08 | 20 days (80% RH, 25 °C) | This work |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/CQDs/$CsPbBr_3$/Br-GO/carbon | 7.88 | 1.602 | 80.01 | 10.10 | 30 days (85%RH, 25 °C) | 1 |
| FTO/$SnO_2$/GQDs/$CsPbBr_3$/carbon | 7.94 | 1.585 | 82.2 | 10.34 | 10 days (80%RH, 25 °C) | 2 |
| Muscovite/ITO/Sb-$TiO_2$/$CsPbBr_3$/carbon | 4.79 | 1.530 | 77.9 | 5.71 | 135 days (70-95%RH, 23-32 °C) | 3 |
| ITO-PEN/ZnO/$CsPbBr_3$/$Cu_2O$/Spiro-OMeTAD/Au | 5.46 | 1.30 | 70.44 | 5.00 | 10 days (85 °C, $N_2$), 60 days (25 °C, 50% RH) | 4 |
| FTO/*N*-$TiO_2$-NRAs/$CsPbBr_3$/carbon | 6.55 | 1.58 | 81.96 | 8.50 | 50 days (25 °C, 80% RH) | 5 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/M$CsPbBr_3$/carbon | 7.42 | 1.584 | 82.11 | 9.65 | 30 days (85% RH, 20 °C; 0% RH, 85 °C) | 6 |
| FTO/$SnO_2$/$TiO_xCl_{4-2x}$/$Rb^+$-$CsPbBr_3$/carbon | 7.96 | 1.629 | 80.5 | 10.44 | 18 days (80% RH, 25 °C) | 7 |
| FTO/Sb-$TiO_2$/$CsPbBr_3$/carbon | 6.70 | 1.654 | 80.4 | 8.91 | 30 days (80% RH, 25 °C) | 8 |
| FTO/*L*-$TiO_2$:$MoSe_2$/$CsPbBr_3$/carbon | 7.88 | 1.615 | 78.7 | 10.02 | 10 days (80% RH, 25 °C) | 9 |
| FTO/$SnO_2$/$CsPbBr_3$/*N*-CQDs/carbon | 7.87 | 1.622 | 80.1 | 10.71 | 20 days (85% RH, room temperature) | 10 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/$CuInS_2$/ZnS QDs/LPP-carbon | 7.73 | 1.626 | 86.3 | 10.85 | 80 days (80% RH, 25 °C); 40 days (0% RH, 80 °C); 30 days (solar irradiation) | 11 |

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|---|---|---|---|---|---|---|
| FTO/*c*-$TiO_2$/$CsPbBr_3$/carbon | 6.89 | 1.49 | 79 | 8.11 | 20 days (85% RH, 20 °C); 25 days (40% RH, 70 °C) | 12 |
| FTO/$TiO_2$/$CsPb_{0.998}Co_{0.002}Br_3$/Spiro-OMeTAD/Au | 7.45 | 1.357 | 84.84 | 8.57 | 9 days (air environment) | 13 |
| FTO/$SnO_2$/$CsPbBr_3$/$CsSnBr_3$/carbon | 7.80 | 1.610 | 84.4 | 10.60 | 10 days (80% RH, 25 °C) | 14 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$Sm^{3+}$-$CsPbBr_3$/Cu(Cr, Ba)$O_2$/carbon | 7.81 | 1.615 | 85.5 | 10.79 | 60 days (80% RH, 25 °C); 40 days (80°C, 0 % RH) | 15 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$Sr^{2+}$-$CsPbBr_3$/carbon | 7.71 | 1.54 | 81.1 | 9.63 | 33 days (80% RH, 25 °C) | 16 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/GQDs/$CsPbBr_3$/MnS/carbon | 8.28 | 1.52 | 83 | 10.45 | 150 days (80% RH, 25 °C); 100 days (80% RH, 85 °C) | 17 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/Spiro-OMeTAD/Ag | 6.41 | 1.37 | 72 | 6.32 | 15 days (79% RH, 27.6 °C) | 18 |
| FTO/$TiO_2$/$CsPbBr_3$/carbon | 7.48 | 1.19 | 68.8 | 6.12 | 90 days (30% RH, 25 °C) | 19 |
| FTO/*c*-$TiO_2$/PTI-$CsPbBr_3$/spiro-OMeTAD/Ag | 9.78 | 1.498 | 74.47 | 10.91 | 41 days (45% RH); 3 days (100°C) | 20 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/GQDs/$CsPbBr_3$/P3HT/carbon | 7.02 | 1.36 | 68 | 6.49 | 40 days (ambient condition) | 21 |
| FTO/*c*-$TiO_2$/$SnO_2$/$CsPbBr_3$/CuPc/carbon | 8.24 | 1.31 | 81.4 | 8.79 | 41 days (40% RH, 25 °C); 30 days (60 °C) | 22 |
| FTO/*c*-$TiO_2$/$CsPbBr_3$/carbon | 7.37 | 1.545 | 82.2 | 9.35 | 40 days (100 °C) | 23 |
| FTO/*c*-$TiO_2$/$CsPbBr_3$/$Ti_3C_2$-MXene/carbon | 8.54 | 1.444 | 73.08 | 9.01 | 79 days (45% RH, 25 °C); 25 days (80 °C) | 24 |

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|---|---|---|---|---|---|---|
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$Sn^{2+}$-$CsPbBr_3$/carbon | 7.66 | 1.37 | 82.22 | 8.63 | 90 days (10% RH, 25 °C; air environment) | 25 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/carbon | 7.40 | 1.22 | 84.1 | 7.37 | 62 days (30-35% RH, 20-25 °C) | 26 |
| FTO/*c*-$TiO_2$/$CsPbBr_3$/$CsPbBr_3$-$CsPb_2Br_5$/$CsPbBr_3$-$Cs_4PbBr_6$/carbon | 9.24 | 1.461 | 75.39 | 10.17 | 125 days (45% RH, room temperature); 29 days (100 °C) | 27 |
| FTO/ZnO/$CsPbBr_3$-$CsPb_2Br_5$/Spiro-OMeTAD/Au | 6.17 | 1.43 | 77.2 | 6.81 | 100 days (45% RH, 25 °C) | 28 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/Spiro-OMeTAD/Au | 6.52 | 1.34 | 69.0 | 6.05 | 100 days (40~70% RH, room temperature) | 29 |
| ITO/ZnO/$CsPbBr_3$/Spiro-OMeTAD/Au | 6.15 | 1.38 | 70.51 | 5.98 | 5 days (under UV in $N_2$) | 30 |
| FTO/*c*-$TiO_2$/$CsPbBr_3$/carbon | 6.46 | 1.34 | 68.04 | 5.86 | 10 days 50~60% RH, 25 °C) | 31 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/GQDs/$CsPbBr_3$/carbon | 8.12 | 1.458 | 82.1 | 9.72 | 130 days (90% RH, 25 °C); 40 days (0% RH, 80 °C) | 32 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$Sm^{3+}$-$CsPbBr_3$/carbon | 7.48 | 1.594 | 85.1 | 10.14 | 110 days (80% RH, 25 °C) | 33 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$Rb^{+}$-$CsPbBr_3$/carbon | 7.73 | 1.552 | 82.2 | 9.86 | 29 days (80% RH, 25 °C) | 34 |
| FTO/*c*-$TiO_2$/$CsPbBr_3$/spiro-OMeTAD/Au | 6.97 | 1.27 | 78.5 | 6.95 | 60 days (30% RH, 25 °C) | 35 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/*m*-$ZrO_2$/$CsPbBr_3$/*m*-carbon | 7.75 | 1.44 | 73.52 | 8.2 | No data provided | 36 |

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|---|---|---|---|---|---|---|
| FTO/*c*-$TiO_2$/$CsPbBr_3$-$CsPb_2Br_5$/spiro-OMeTAD/Ag | 8.48 | 1.296 | 75.9 | 8.34 | 41 days (45% RH) | 37 |
| FTO/$TiO_2$/CQD-$CsPbBr_3$ IO/Spiro-OMeTAD/Au | 11.34 | 1.06 | 69.0 | 8.29 | 45 days (30% RH) | 38 |
| FTO/*c*-$TiO_2$/$CsPbBr_3$/spiro-OMeTAD/Au | 5.6 | 1.5 | 62 | 5.4 | No data provided | 39 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/PTAA/Au | 6.70 | 1.25 | 73.0 | 6.20 | 14 days (15-70% RH) | 40 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/carbon | 5.70 | 1.29 | 68.0 | 5.00 | 10 days (30-50% RH, 80 °C ) | 41 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/carbon | 7.4 | 1.24 | 73.0 | 6.7 | 90 days (90-95% RH, 25 °C) | 42 |
| FTO/*m*-$TiO_2$/$CsPbBr_3$/PTAA/Au | 6.16 | 1.27 | 73 | 5.72 | No data provided | 43 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/carbon/$CsPbBr_3$/*m*-$TiO_2$/*c*-$TiO_2$/ FTO | 7.1 | 1.388 | 77 | 7.55 | 41 days (60% RH, 25 °C) | 44 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/carbon-PANi/G | 6.87 | 1.59 | 81.21 | 8.87 | 50 days (80% RH, 25 °C) | 45 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/($MoO_2$/NC)carbon | 7.2 | 1.532 | 85.2 | 9.4 | 33 days (80% RH, 25 °C); 10 days (0% RH, 85 °C) | 46 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/($CuInS_2$/ZnS QDs)/carbon | 7.47 | 1.45 | 77.73 | 8.42 | 35 days (80% RH, 25 °C) | 47 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/PtNi-carbon | 6.78 | 1.432 | 81 | 7.86 | 20 days (80% RH, 25 °C) | 48 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/PVAc/GO/carbon | 7.41 | 1.553 | 82.80 | 9.53 | 30 days (80% RH, 25 °C), 20 days (0% RH, 85 °C), 10 days (85% RH, 85 °C), 20 days ( continuous illuminationat their MPP) | 49 |

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|---|---|---|---|---|---|---|
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/$MoS_2$ QDs/carbon | 6.55 | 1.307 | 79.4 | 6.80 | 29 days (80% RH) | 50 |
| FTO/GQDs/$CsPbBr_3$/PQDs/carbon | 5.08 | 1.21 | 66.7 | 4.10 | 8 days (80% RH, 25 °C) | 51 |
| FTO/$TiO_2$/$CsPbBr_3$/spiro-MeOTAD/Au | 7.05 | 1.41 | 55 | 5.5 | No data provided | 52 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/spiro-MeOTAD/Ag | 8.47 | 1.02 | 71.6 | 6.21 | 12 days (40% RH), 12 days (50 °C) | 53 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/AIGS QDs/carbon | 7.43 | 1.460 | 80.31 | 8.46 | 10 days (ambient) | 54 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/CdZnSe@ZnSe QDs/carbon | 7.25 | 1.498 | 79.6 | 8.65 | 90 days (60-80%RH, 25 °C) | 55 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/P3HT:ZnPc/carbon | 7.652 | 1.578 | 83.06 | 10.03 | 30 days (70%RH, 20 °C) | 56 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/$CsPbBr_xI_{3-x}$ NCs/carbon | 8.66 | 1.490 | 73.3 | 9.45 | 38 days (80%RH, 25 °C) | 57 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPb_{0.995}Zn_{0.005}Br_3$/carbon | 7.30 | 1.560 | 80.61 | 9.18 | 31 days (80%RH, 25 °C) | 58 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_3$/$CsSnBr_2I$ QDs/carbon | 8.70 | 1.39 | 75.5 | 9.13 | 30 days (80%RH, 25 °C; 0%RH, 80 °C) | 59 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/CQDs/$CsPbBr_3$/PQDs/carbon | 7.24 | 1.45 | 75.6 | 7.93 | 58 days (80%RH, 25 °C) | 60 |
| FTO/$SnO_2$-$TiO_xCl_{4-2x}$/$WS_2$/$CsPbBr_3$/carbon | 7.95 | 1.70 | 79 | 10.65 | 120 days (80% RH, 25 °C) | 61 |
| FTO/*c*-$TiO_2$/$CsPb_{1-x}Co_xBr_3$/carbon | 7.48 | 1.38 | 84 | 8.67 | 100 days (air); 24 days (85 $^o$C) | 62 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsP_{b1-x}Eu_xBr_3$/carbon | 6.33 | 1.45 | 79.19 | 7.28 | 35 days (dark, 15-20% RH) | 63 |

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|---|---|---|---|---|---|---|
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/AC/$CsPbBr_3$/ZnPc/carbon | 7.64 | 1.606 | 82.47 | 10.12 | 30 days (80% RH, 25 $^oC$) | 64 |
| FTO/*c*-$TiO_2$/*m*-$TiO_2$/$CsPbBr_{3-x}Cl_x$/carbon | 7.47 | 1.571 | 82.93 | 9.93 | 60 days (80% RH, 25 $^oC$; 0% RH, 80 $^oC$) | 65 |
| FTO/Ga-$SnO_2$/$CsPbBr_3$/carbon | 8.42 | 1.354 | 71.32 | 8.13 | 50 days (40-25 °C, 50% RH) | 66 |

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