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Figure S1. A cross-sectional SEM image of a 1%Al-LTC_{0.9}A_{0.1}/Au photoelectrode fabricated by the particle transfer method. The dashed curve indicating the border of the Al-LTC_{0.9}A_{0.1} particle layer and the underlying Au layer is included as a guide for the eye.



Figure S2. Lattice spacing of the (714) planes, d_{714} , of 1%Al-LTC_{1-x}A_x, estimated by XRD.



Figure S3. Indirect bandgap energies for 1% Al-LTC_{1-*x*}A_{*x*}. The quadratic fit, represented by the dashed curve, is a guide for the eye.



Figure S4. Indirect bandgap energies for 2% Al-LTC_{1-*x*}A_{*x*}. The quadratic fit, represented by the dashed curve, is a guide for the eye.



Figure S5. Top-view SEM images of 1%Al-LTC_{0.9}A_{0.1} photocathodes (a) before and (b) after a PEC measurement at +0.7 V vs. RHE conducted over 90 min under simulated sunlight illumination in an aqueous solution of Na₂SO₄ (0.5 M, pH 10). The photocathodes were modified with Pt by photodeposition.