Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting information

Modification of $BiVO_4/WO_3$ composite photoelectrodes with Al_2O_3 via chemical vapor deposition for highly efficient oxidative H_2O_2 production from H_2O

List of authors

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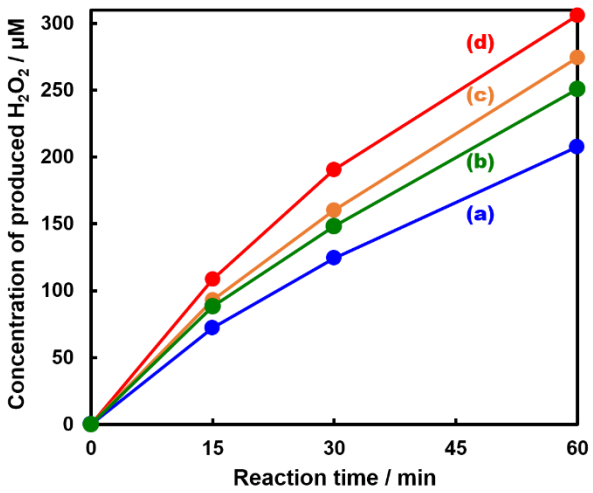

Figure S1. Concentration of H_2O_2 produced on an Al_2O_3(CVD5)/$BiVO_4$/WO_3 photoelectrode (6 cm^2). Using a two-electrode photoelectrochemical cell with an Al_2O_3(CVD5)/$BiVO_4$/WO_3 as an anode and a Pt mesh as a cathode, the concentration of electrolyte solutions were varied: (a) 0.1 M, (b) 0.5 M, (c) 1.0 M and (d) 2.0 M $KHCO_3$ aqueous solution under CO_2 bubbling and simulated solar irradiation by controlling the stable current of 1.0 mA.

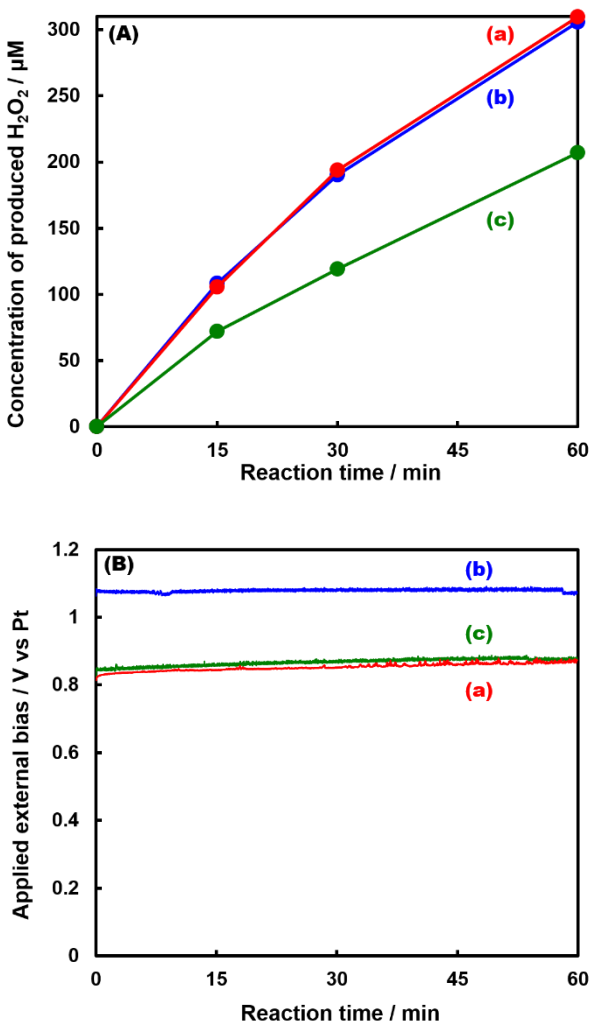

Figure S2. Time course of (A) concentration of H_2O_2 produced in the photoelectrochemical reaction using various photoelectrodes and (B) Applied external bias under stable current of 1 mA. Using a two-electrode photoelectrochemical cell with a photoanode (6 cm^2): (a) Al_2O_3(CVD5)/$BiVO_4$/WO_3; (b) Al_2O_3(MOD)/$BiVO_4$/WO_3; and (c) $BiVO_4$/WO_3. A Pt electrode was used as a cathode with 2.0 M $KHCO_3$ aqueous solution under CO_2 bubbling and simulated solar irradiation by controlling the stable current of 1.0 mA.

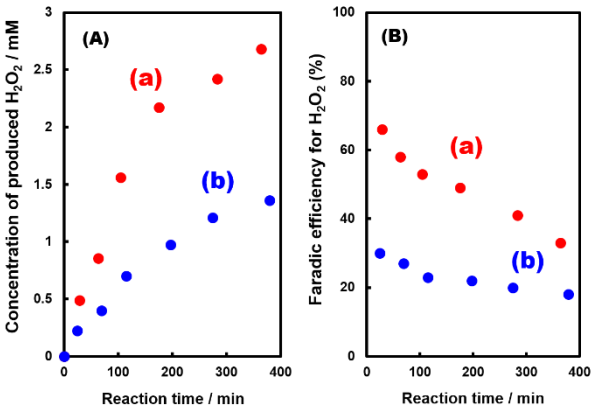

Figure S3. Time course of (A) concentration of produced H_2O_2 and (B) FE(H_2O_2) in the prolonged photoelectrochemical reaction using the photoelectrode under an applied external bias of 1.0 V vs Pt. Using a two-electrode photoelectrochemical cell with a photoanode (6 cm^2): (a) Al_2O_3(CVD5)/$BiVO_4$/WO_3 and (b) $BiVO_4$/WO_3. A Pt mesh electrode was used with 2.0 M $KHCO_3$ aqueous solution under CO_2 bubbling and simulated solar irradiation.

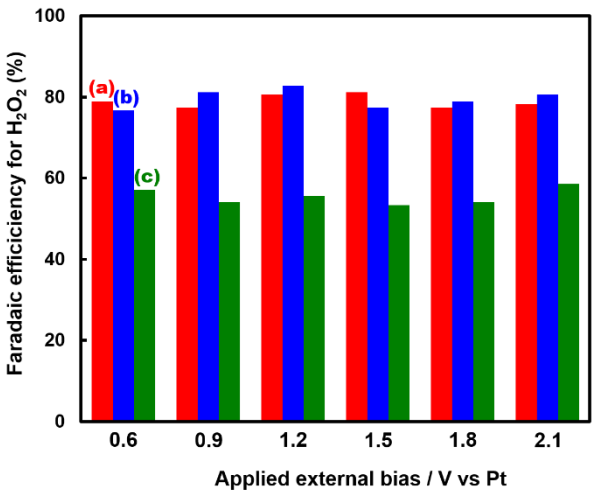

Figure S4. The faradaic efficiency (FE) of H_2O_2 production in the photoelectrochemical reaction after passing 0.9 C using photoelectrode (6 cm^2) (a) Al_2O_3(CVD5)/$BiVO_4$/WO_3, (b) Al_2O_3(MOD)/$BiVO_4$/WO_3, and (c) $BiVO_4$/WO_3 with an applied external bias of 0.6 V – 2.1 V vs a Pt counter electrode in a two-electrode photoelectrochemical cell containing a 2.0 M $KHCO_3$ aqueous solution under CO_2 bubbling and simulated solar irradiation.

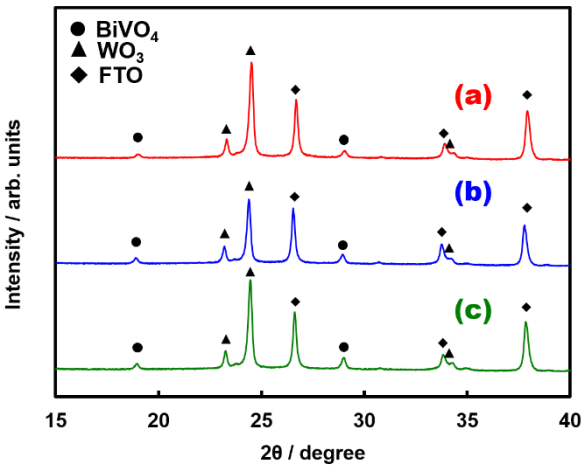

Figure S5. XRD patterns of (a) Al_2O_3(CVD5)/$BiVO_4$/WO_3, (b) Al_2O_3(MOD)/$BiVO_4$/WO_3, and (c) $BiVO_4$/WO_3.

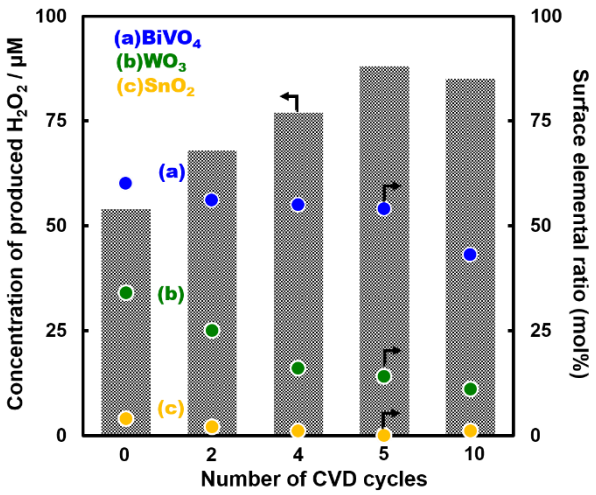

Figure S6. Concentration of H_2O_2 produced after 0.9 C of electric charge was passed (left) and surface elemental ratio calculated from XPS spectra of Al_2O_3(CVDn)/$BiVO_4$/WO_3 (n = 0 - 10) photoelectrodes (6 cm^2) (right). A 0.5 M $KHCO_3$ aqueous solution (CO_2 bubbling) was used with a Pt mesh electrode under CO_2 bubbling and simulated solar irradiation by controlling the stable current of 1.0 mA during the reaction.

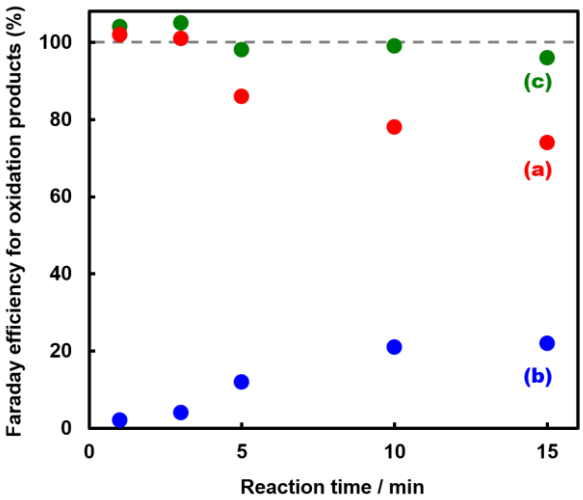

Figure S7. Time courses of FE for (a) H_2O_2, (b) O_2, and (c) H_2O_2 + O_2 in the photoelectrochemical reaction using Al_2O_3(CVD5)/$BiVO_4$/WO_3 photoelectrode (6 cm^2). A two-electrode photoelectrochemical cell with an photoanode and a Pt wire as a cathode electrode was used in a 2.0 M $KHCO_3$ aqueous solution under CO_2 bubbling and simulated solar irradiation by controlling the stable current of 1.0 mA.

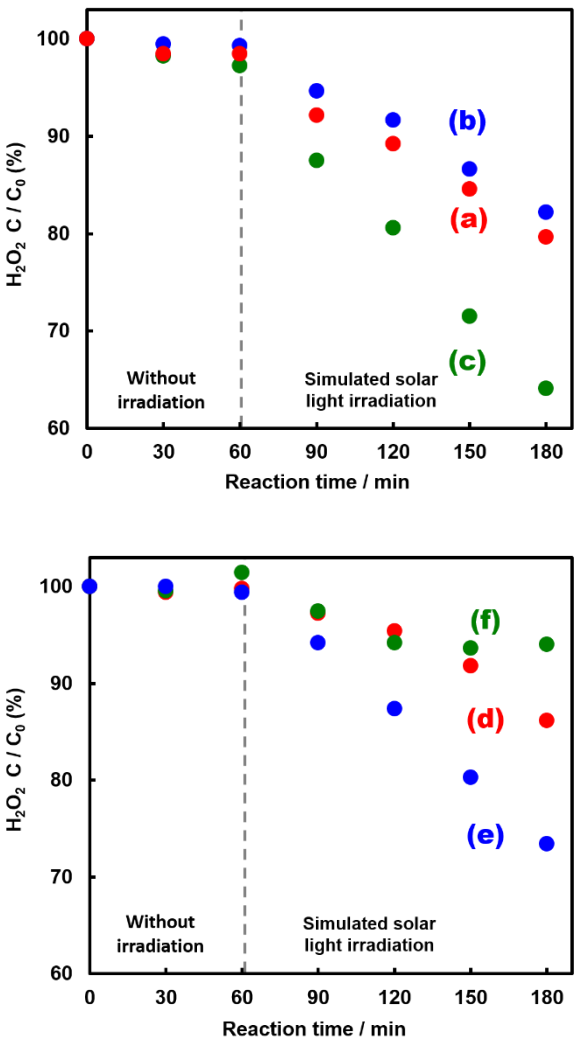

Figure S8. Photodecomposition rate (C/C_0) of H_2O_2 on each electrode (6 cm^2): (a) Al_2O_3(CVD5)/$BiVO_4$/WO_3, (b) Al_2O_3(MOD)/$BiVO_4$/WO_3, (c) $BiVO_4$/WO_3, (d) $BiVO_4$, (e) WO_3 and (f) FTO without external bias. Electrolyte solution: 0.5 M $KHCO_3$ aqueous solution with 550 μM H_2O_2 under CO_2 bubbling and simulated solar irradiation.

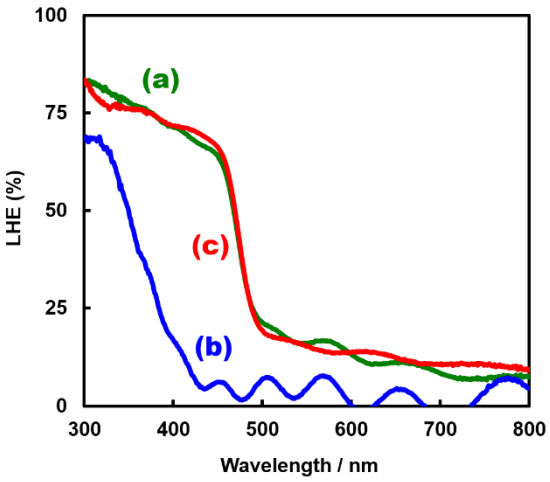

Figure S9. Light harvesting efficiency (LHE) of (a) $BiVO_4/WO_3$, (b) $BiVO_4$, and (c) WO_3 photoelectrode by irradiation from the front side.

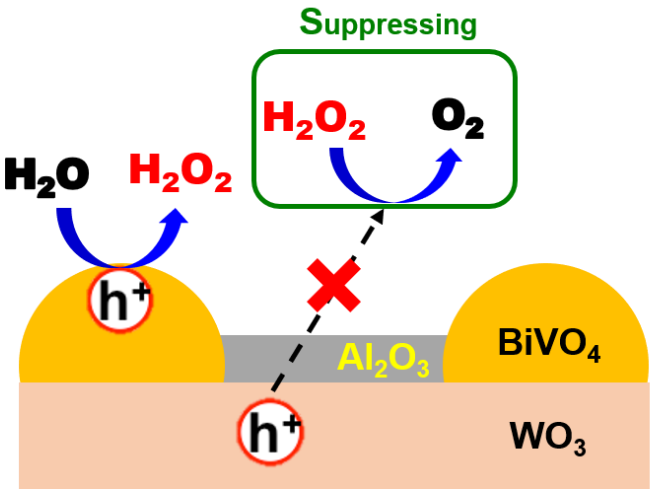

Figure S10 Speculated scheme for the suppression of H_2O_2 decomposition on Al_2O_3(CVD)/$BiVO_4$/WO_3 photoelectrode.

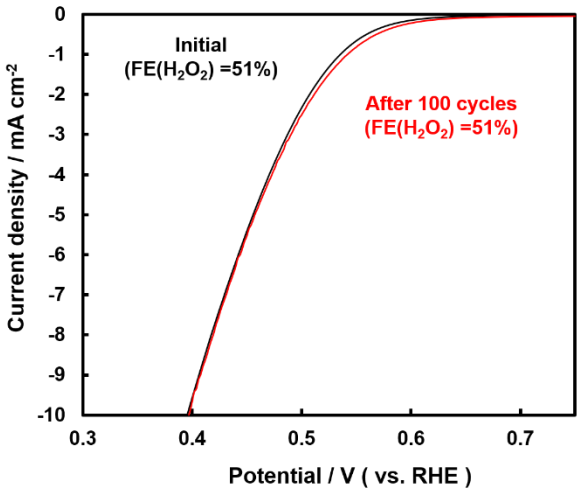

Figure S11 The comparison of I-V curve and faradaic efficiency (FE) for H_2O_2 production on WSoy/GnP-CP cathode. The cathode was utilized before / after 100 cycles of CV measurement operated under the same condition of Figure 7.