

# Supporting Information

## Promoted Electroreduction of CO<sub>2</sub> with Oxygen Vacancies on Plasma-Activated SnO<sub>x</sub>/Carbon Foam Monolithic Electrode

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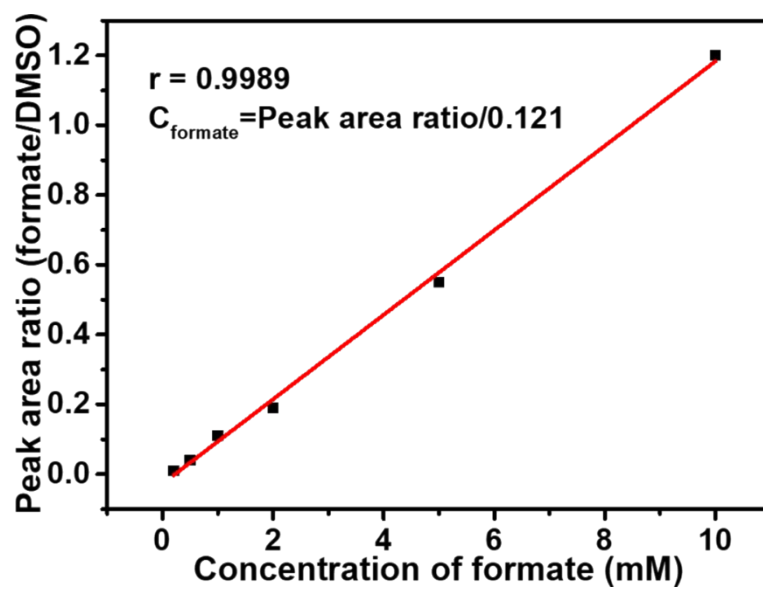
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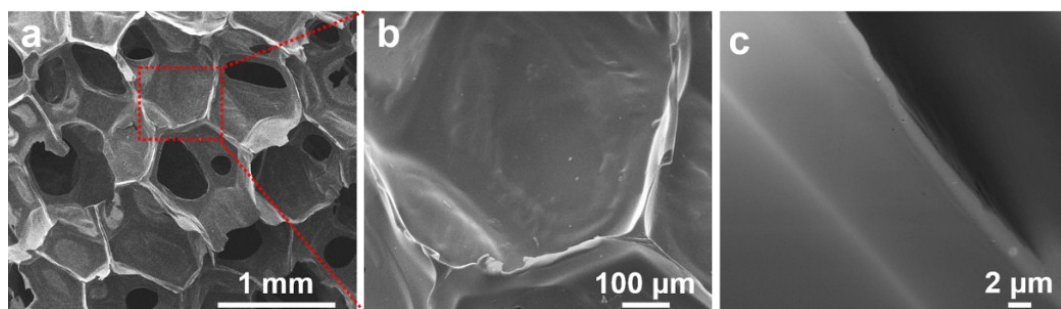
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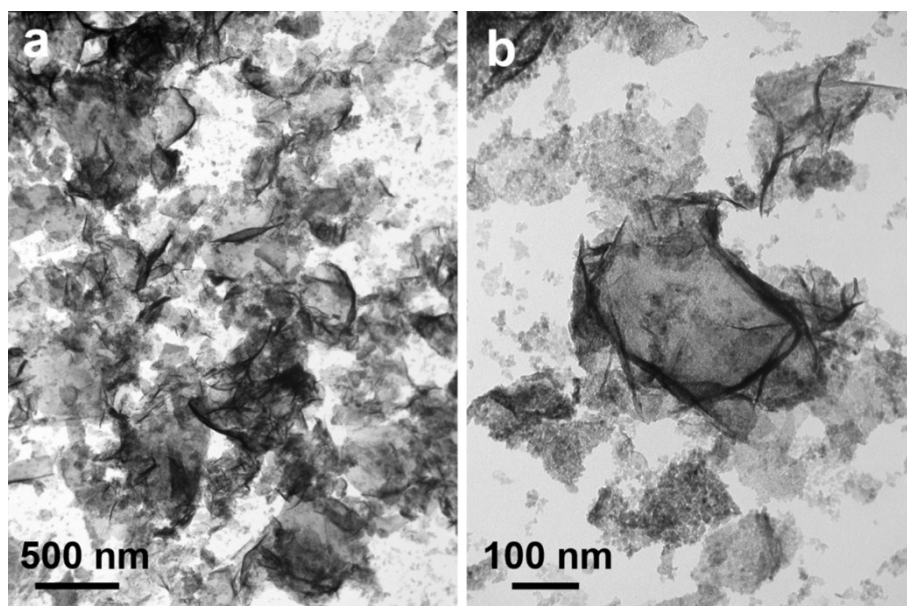
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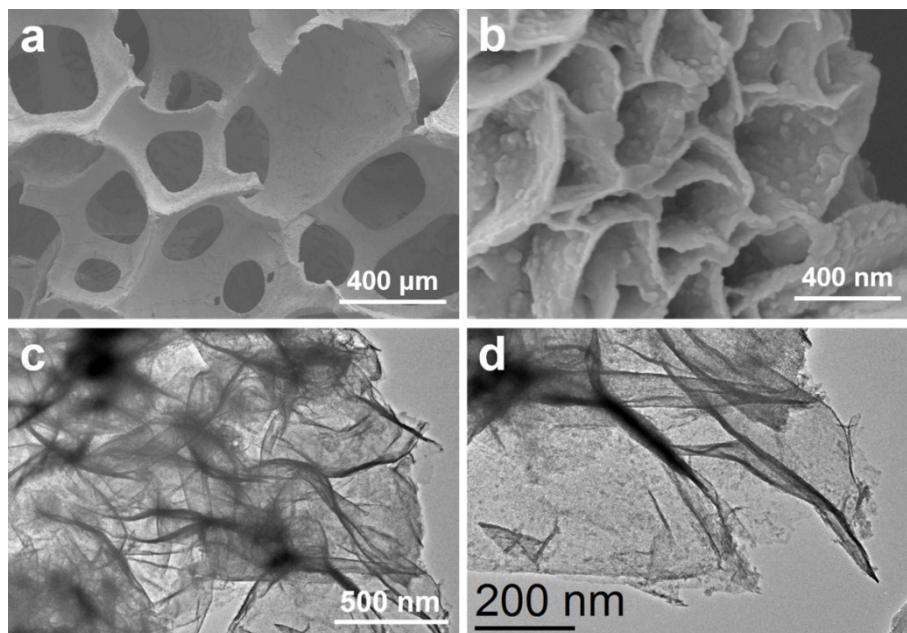
**Figure S1.** Linear relationship between the formate concentration and relative peak area ratio (formate/DMSO).



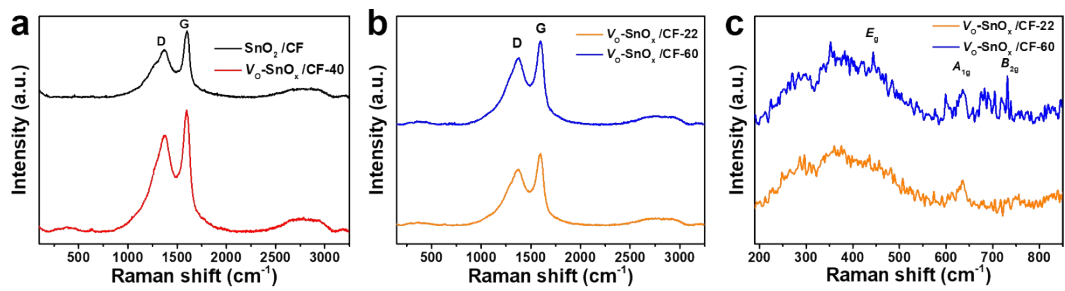
**Figure S2.** (a) Low-magnification and (b, c) high-magnification FE-SEM images of pristine carbon foam.



**Figure S3.** TEM images of pristine SnO<sub>2</sub> nanosheets.



**Figure S4.** (a) Low-magnification and (b) high-magnification FE-SEM images of  $V_{O}\text{-SnO}_x/\text{CF}$  sample. (c, d) TEM images of  $V_{O}\text{-SnO}_x/\text{CF}$  sample.



**Fig**

**ure S5.** (a) Raman spectra of pristine SnO<sub>2</sub>/CF and V<sub>0</sub>-SnO<sub>x</sub>/CF-40. (b) Raman spectra of V<sub>0</sub>-SnO<sub>x</sub>/CF-22 and V<sub>0</sub>-SnO<sub>x</sub>/CF-60. (c) High-magnification Raman spectra of V<sub>0</sub>-SnO<sub>x</sub>/CF-22 and V<sub>0</sub>-SnO<sub>x</sub>/CF-60.

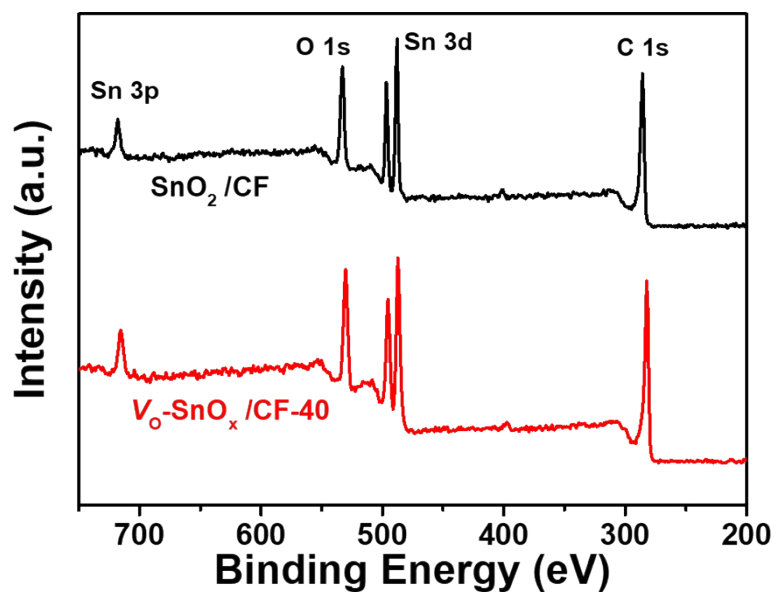
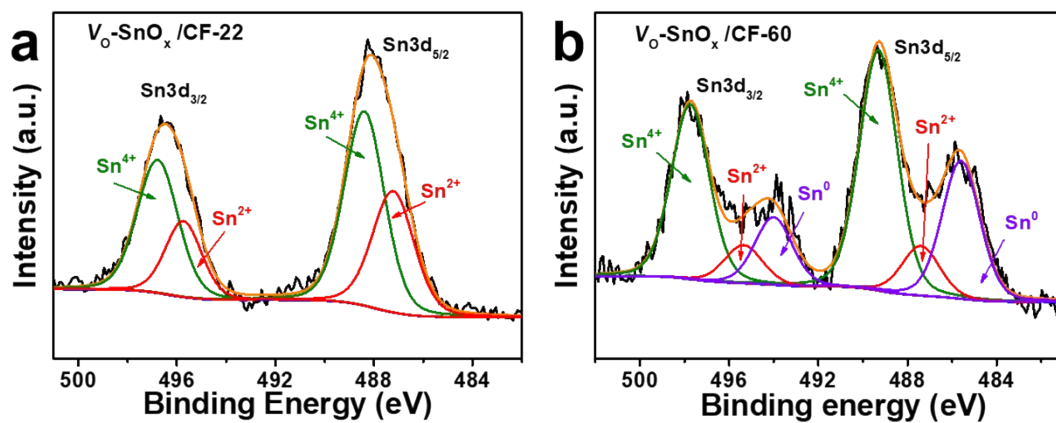
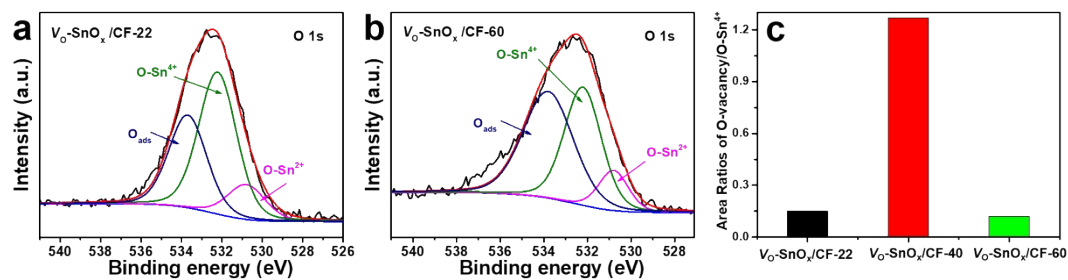


Figure S6. XPS spectra of pristine SnO<sub>2</sub>/CF and V<sub>O</sub>-SnO<sub>2</sub>/CF-40 samples.

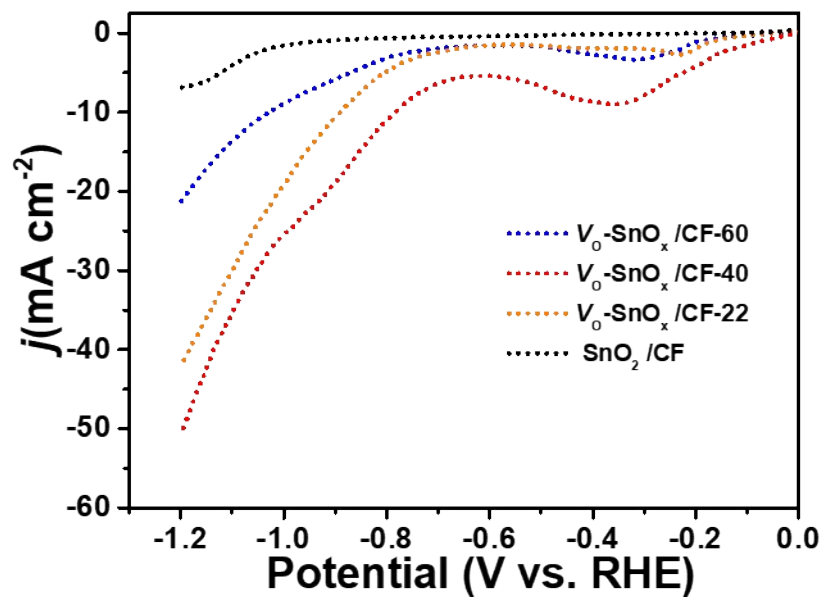


**Figure S7.** High-resolution XPS spectra of Sn 3d for  $V_O-SnO_x/CF-22$  (a) and  $V_O-SnO_x/CF-60$  (b).

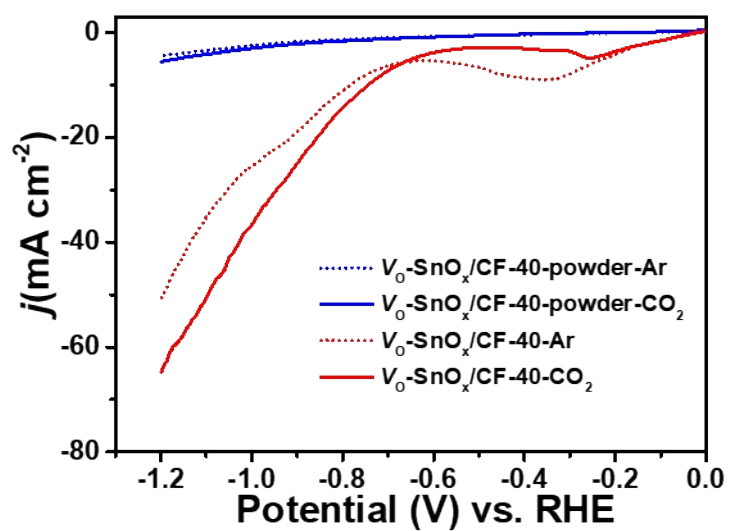




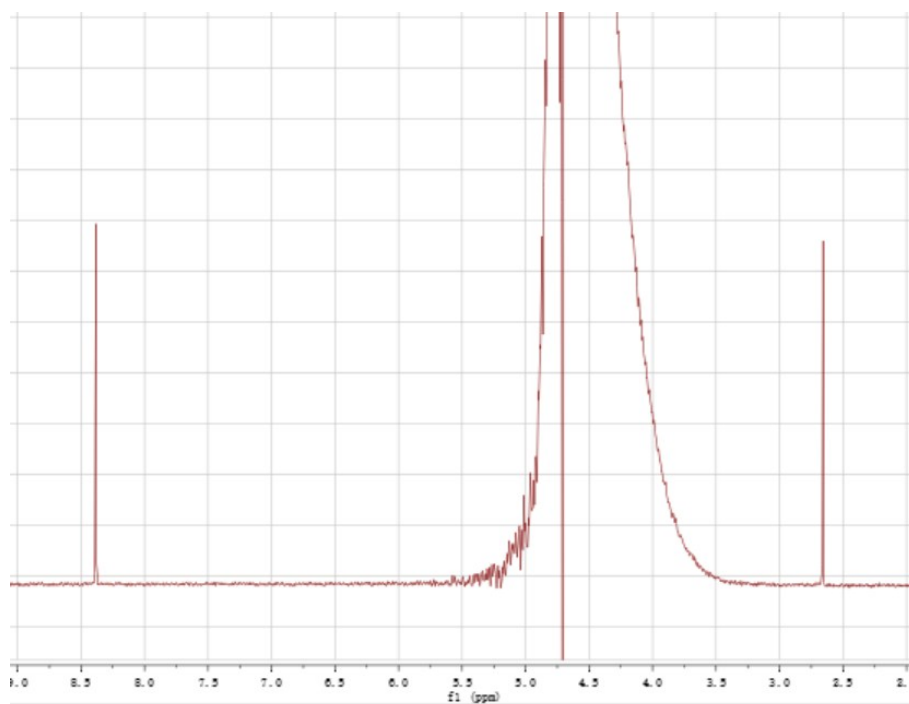
**Figure S8.** XPS spectra of O 1s for  $V_0\text{-SnO}_2/\text{CF-22}$  (a) and  $V_0\text{-SnO}_2/\text{CF-60}$  (b), the calculated ratio of integral areas for the peaks of O-vacancy and  $\text{O-Sn}^{4+}$  in O 1s XPS spectra of different  $V_0\text{-SnO}_2/\text{CFs}$  (c).



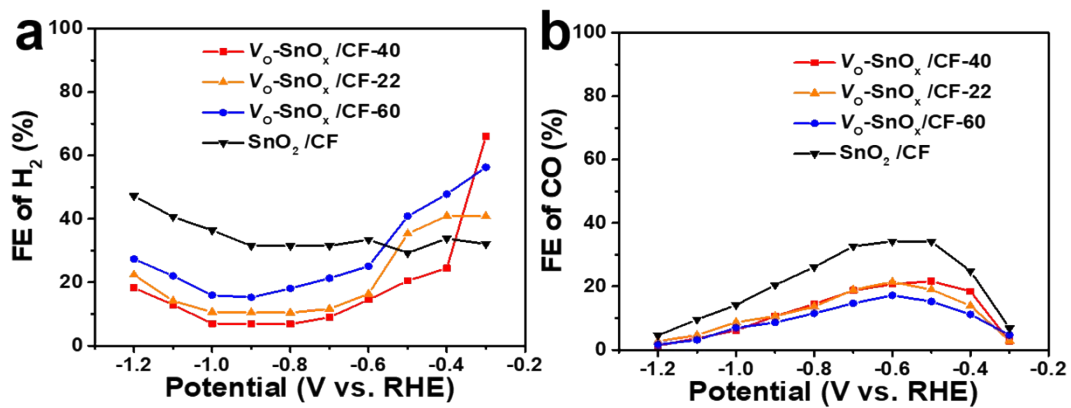
**Figure S9.** Linear sweep voltammetry curves of the prepared electrodes in Ar-saturated 0.1 M KHCO<sub>3</sub> aqueous solution at a scan rate of 20 mV s<sup>-1</sup>.



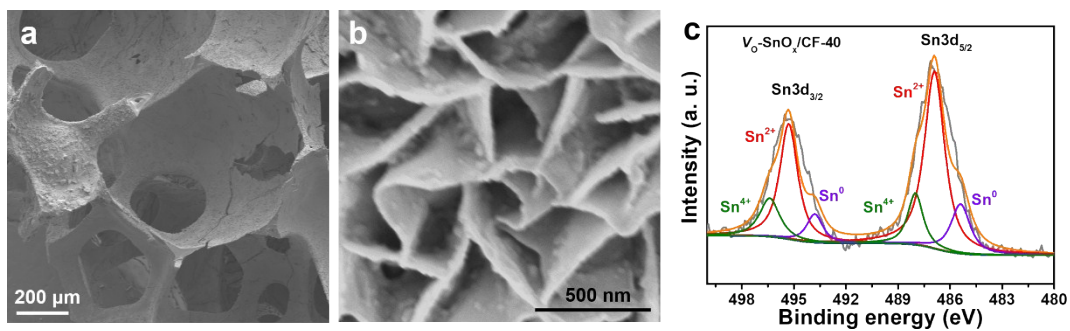
**Figure S10.** Linear sweep voltammetry curves of the  $V_0$ -SnO<sub>x</sub>/CF-40 powder and the  $V_0$ -SnO<sub>x</sub>/CF-40 monolithic electrode in Ar- and CO<sub>2</sub>-saturated 0.1 M KHCO<sub>3</sub> aqueous solution at a scan rate of 20 mV s<sup>-1</sup>.



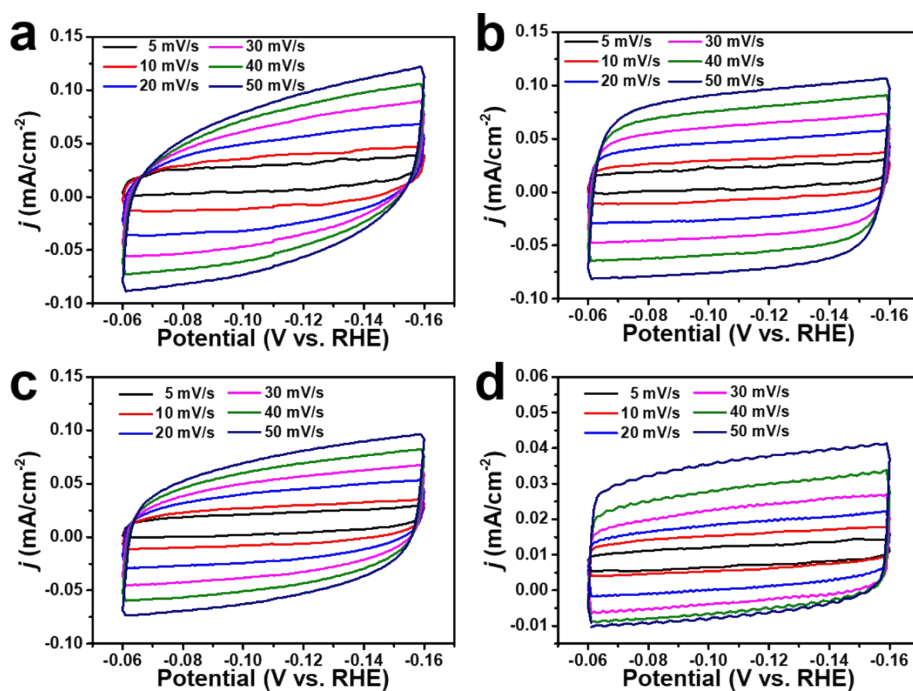
**Figure S11.**  $^1\text{H}$ -NMR spectrum of 0.1 M  $\text{KHCO}_3$  electrolyte after 2 h electrolysis at -1.0 V (vs. RHE) of the  $\text{VO-SnO}_x/\text{CF-40}$ .



**Figure S12.** FE of H<sub>2</sub> (a) and CO (b) for various electrodes at different electrolytic potentials.



**Figure S13.** Physical characterization of  $V_O$ - $\text{SnO}_x$ /CF-40 after 8 h  $\text{CO}_2$  reduction. (a) low-resolution FESEM image, (b) high-resolution FESEM image, (c) XPS spectrum of Sn 3d.



**Figure S14.** Cyclic voltammograms at the range of -0.06 to -0.16 V vs. RHE with different scan rates (5, 10, 20, 30, 40, and 50  $\text{mV s}^{-1}$ ) for  $V_{\text{O}}\text{-SnO}_x/\text{CF-22}$  (a),  $V_{\text{O}}\text{-SnO}_x/\text{CF-40}$  (b),  $V_{\text{O}}\text{-SnO}_x/\text{CF-60}$  (c), and pristine  $\text{SnO}_2/\text{CF}$  (d).

### Supplementary Note 1:

#### Determination of electrochemically active surface area (ECSA).

The ECSAs of the electrodes were determined by the measurement of the double-layer capacitance ( $C_{\text{dl}}$ ). To measure the  $C_{\text{dl}}$ , Cyclic voltammograms were measured in a non-faradaic region (potential range between -0.06 and -0.16 V vs. RHE) of the voltammogram at varied scan rates (5, 10, 20, 30, 40, and 50  $\text{mV s}^{-1}$ ). The capacitive currents of  $\Delta j (j_{\text{a}} - j_{\text{c}}) @ -0.11 \text{ V}/2$  (where  $j_{\text{a}}$  and  $j_{\text{c}}$  were the anodic and cathodic current densities, respectively) are plotted as a function of the CV scan rates. These data were fitted to a line. The slope of the line is the double layer capacitance ( $C_{\text{dl}}$ ), which is proportional to the ECSA of electrodes.

To facilitate the comparison of ECSA data between different electrodes, the monolithic electrodes were ground to powder for ECSA testing. The powder was then dripped onto the surface of a glassy carbon electrode ( $S = 0.196 \text{ cm}^2$ ) for CV testing at varied scan rates (5, 10, 20, 30, 40, and 50  $\text{mV s}^{-1}$ ).

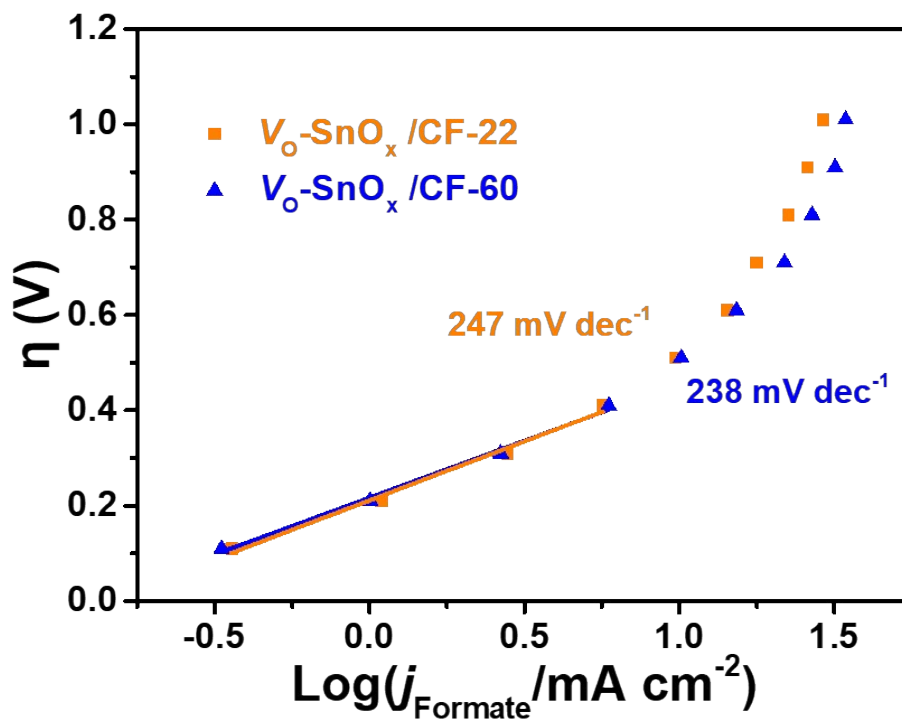
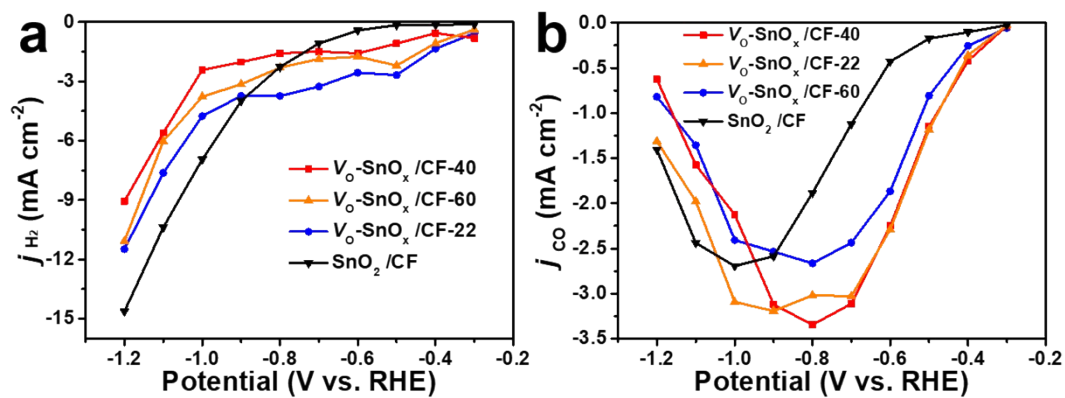


Figure S15. Tafel plots for  $V_{\text{O}}\text{-SnO}_2/\text{CF-22}$  and  $V_{\text{O}}\text{-SnO}_2/\text{CF-60}$ .





**Figure S16.** Partial current density of H<sub>2</sub> (a) and CO (b) for various electrodes at different electrolytic potentials.

### Supplementary Note 2:

The partial current densities at different potentials were calculated by multiplying the overall geometric current densities and the corresponding faradic efficiencies.