Supporting Information

Morphological analysis of the prepared Ni-Fe LDH, α-MnO₂ and air electrode

Figure S1, panels (a)–(c) respectively show SEM images of Ni-Fe CO₃²⁻ LDH, α-MnO₂ and the prepared reversible air electrode surface. Then, Figure S1, panels (d)–(f) respectively show EDX maps for carbon, nickel, and manganese in the reversible air electrode. The EDX maps revealed the uniform distribution of carbon (in vulcan carbon), nickel (in Ni-Fe CO₃²⁻ LDH), and manganese (in α-MnO₂) throughout the reversible air electrode, indicating formation of favorable triple phase boundary regions.

![Figure S1 FE-SEM images of (a) Ni-Fe CO₃²⁻ LDH, (b) α-MnO₂, and (c) the reversible air electrode surface and EDX maps of (d) carbon, (e) nickel, and (f) manganese for the reversible air electrode.](image_url)
Electrocatalytic activity of the prepared Ni-Fe LDH

The electrocatalytic activity of Ni-Fe LDH in the air electrodes was evaluated using a half cell. A catalyst ink of the air electrode was prepared by mixing Ni-Fe LDH, vulcan carbon, and PTFE as binder with a weight ratio of 1 : 1 : 0.6. The ORR and OER activities of the prepared electrodes were evaluated in a three-electrode cell. The steady-state polarization curves of the prepared reversible air electrodes are shown in Fig. S2. These results show that Ni-Fe LDH has catalytic activity for ORR and OER.

Figure S2 Steady-state polarization curves for oxygen reduction reaction activity and oxygen evolution reaction of the reversible air electrodes.
Tafel plots of the prepared air electrode with or without Ni-Fe LDH

Figure S3 shows the Tafel plots of the air electrodes with or without Ni-Fe LDH. The Tafel slope of the polarization curve was decreased by the addition of Ni-Fe CO$_3$ LDH to the catalyst layer of the reversible air electrode using α-MnO$_2$ as catalyst, indicating that Ni-Fe CO$_3$ LDH formed more favorable 5 triple-phase boundary regions and facilitated the electrode reactions.

Figure S3 Tafel plots of the reversible air electrodes for oxygen reduction reaction activity and oxygen evolution reaction.