Supporting Information

Morphological analysis of the prepared Ni-Fe LDH, $\alpha\text{-MnO}_2$ and air electrode

Figure S1, panels (a)–(c) respectively show SEM images of Ni-Fe CO_3^{2-} 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 5 maps revealed the uniform distribution of carbon (in vulcan carbon), nickel (in Ni-Fe CO_3^{2-} 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 CO_3^{2-} 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.

30

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 5 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^{2-} LDH to the catalyst layer of the reversibleair electrode using α -MnO₂ as catalyst, indicating that Ni-Fe CO_3^{2-} 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.