## Refining Mn-Ni Synergy for the Design of Efficient Catalysts in

## **Electrochemical Ethanol Oxidation**

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Figure S1. Electrochemical deposition current density over various Mn/NF electrodes.



Figure S2. Comparison of Nyquist plots before and after stability test over Mn/NF-0 (a), Mn/NF-1.6 (b), Mn/NF-3.6 (c), Mn/NF-6.1 (d), and Mn/NF-8.6 (e) electrodes. In an electrolyte with 1M KOH and 1 M ethanol, at 50 mV s<sup>-1</sup>.



Figure S3. Comparison of Tafel plots before and after stability test over Mn/NF-0 (a), Mn/NF-1.6 (b), Mn/NF-3.6 (c), Mn/NF-6.1 (d), and Mn/NF-8.6 (e) electrodes. In an electrolyte with 1M KOH and 1 M ethanol, at 10 mV s<sup>-1</sup>. The potentials are IR compensated.



Figure S4. Double-layer capacitance measurements for determining ECSA for various Mn/NF electrodes in 1mol  $L^{-1}$  KOH with 1mol  $L^{-1}$  EtOH.



Figure S5. Electrochemical deposition current density over Mn/NF-6.1 and Mn/carbonpaper. As the deposition current density over carbon paper is much lower, a longerdepositiontimewasapplied.



Figure S6. CV curves for ethanol oxidation over Mn/NF-6.1 and Mn/carbon paper. In an electrolyte with 1M KOH and 1 M ethanol, at 50 mV s<sup>-1</sup>. The potentials are IR compensated.



Figure S7. ECSA-based CV curves obtained at the Mn/Ni electrode after stability testing.

	$\mathrm{BE^{a}}\ \mathrm{for}\ \mathrm{Mn^{3+}}$	BE for Mn <sup>4+</sup>	$Mn^{3+}/Mn^{4+}$	M-OH/M-O ratio
	(eV)	(eV)	ratio	
NF/Mn-1.6	642.27	644.71	1.97	0.66
NF/Mn-3.6	642.33	644.75	1.85	0.81
NF/Mn-6.1	642.31	644.64	1.77	1.18
NF/Mn-8.6	642.33	644.65	1.72	1.30

Table S1. Binding energies for  $Mn^{3+}$  and  $Mn^{4+}$ , as well as the  $Mn^{3+}/Mn^{4+}$  and  $(O_{ad}+O-H_{ad})/O_L$  ratio.

<sup>a</sup> Binding energy.