

## Electronic Supplementary Information

### The role of Ni substitution in manganite perovskite Li-O<sub>2</sub> battery

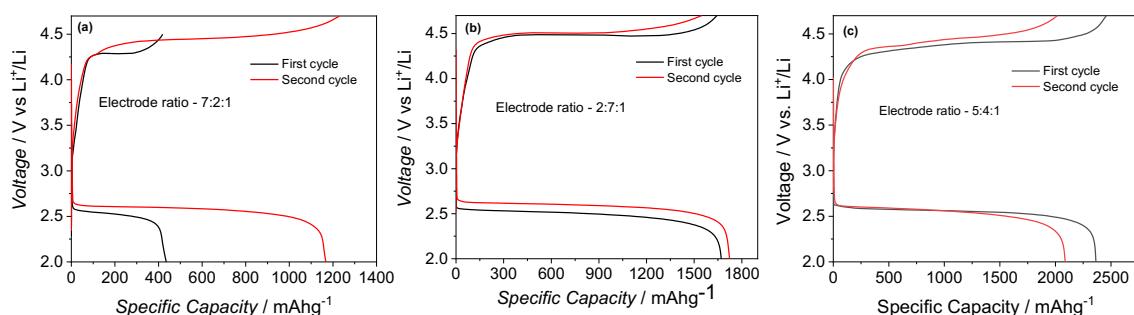
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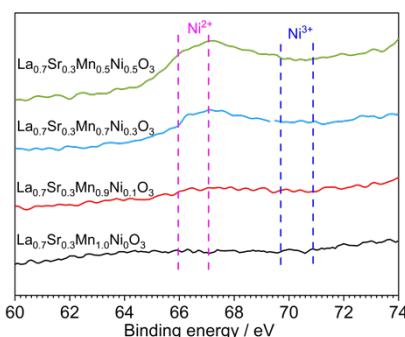
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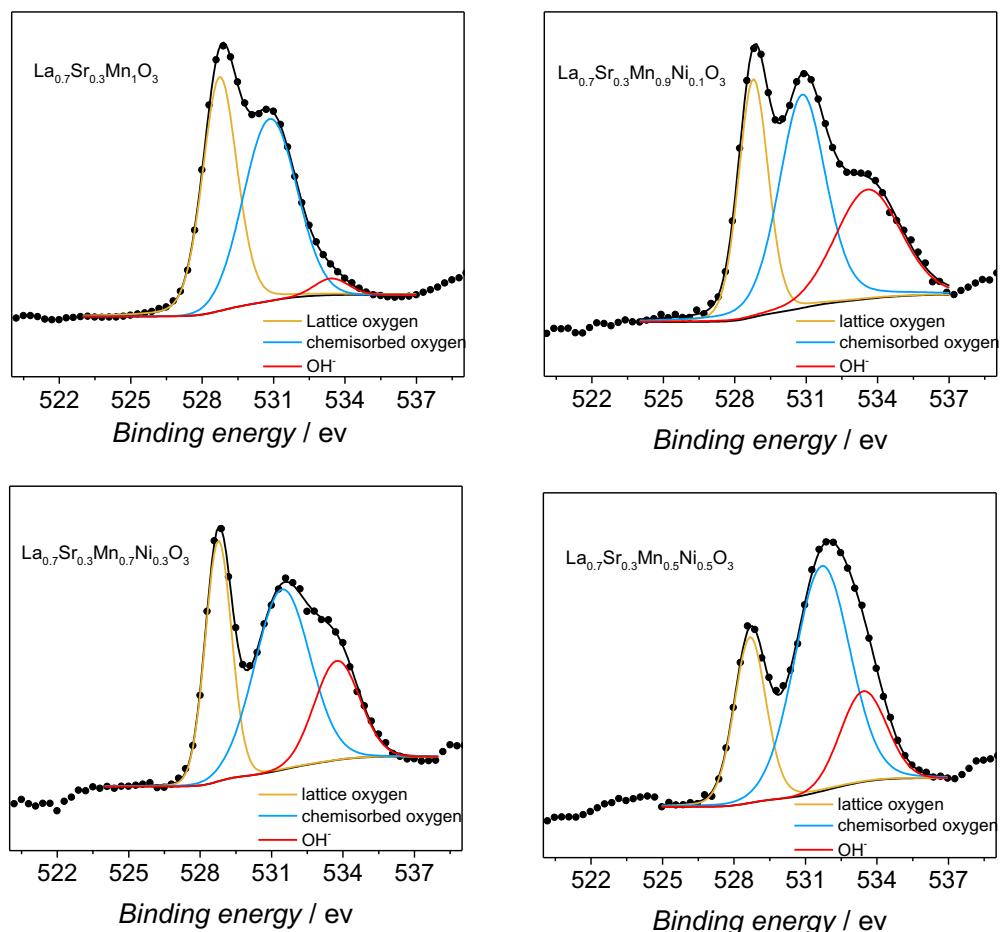
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**Figure S1.** Discharge-charge cycles for Li-O<sub>2</sub> battery composed of La<sub>0.7</sub>Sr<sub>0.3</sub>Mn<sub>0.5</sub>Ni<sub>0.5</sub> catalyst at different ratio of active material, acetylene black, and binding material, i.e. (a) 7:2:1, (b) 2:7:1, and (c) 5:4:1



**Figure S2.** The Ni 3p core-level spectra for La<sub>0.7</sub>Sr<sub>0.3</sub>Mn<sub>1-x</sub>Ni<sub>x</sub>O<sub>3</sub> with x = 0, 0.1, 0.3, 0.5.



**Figure S3.** The O 1s core-level spectra for  $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ni}_x\text{O}_3$  with  $x = 0, 0.1, 0.3, 0.5$ . The fitted O 1s spectra consist of three major peaks at approximately 529, 531, and 533 eV. These bands can be associated with lattice oxygen ( $\text{O}^{2-}$ ), chemisorbed oxygen ( $\alpha$ ), and adsorbed water species ( $\text{OH}^-$ ).

**Table S1.** Molar ratios of  $\text{Mn}^{3+}/\text{Mn}^{4+}$  on  $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{1-x}\text{Ni}_x\text{O}_3$  ( $x = 0, 0.1, 0.3, 0.5$ ), determined from XPS fitting analysis.

Catalyst	$\text{Mn}^{3+}/\text{Mn}^{4+}$	Average oxidation state of Mn ( $\alpha$ )
$\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_1\text{Ni}_0\text{O}_3$	69/31	3.3
$\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.9}\text{Ni}_{0.1}\text{O}_3$	81/19	3.1
$\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.7}\text{Ni}_{0.3}\text{O}_3$	78/22	3.2
$\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.5}\text{Ni}_{0.5}\text{O}_3$	53/47	3.5

**Table S2.** Comparison of specific capacity values for LSMN catalyst in aprotic Li-O<sub>2</sub> battery.

Catalyst	Specific Capacity	Reference
La <sub>0.7</sub> Sr <sub>0.3</sub> MnO <sub>3</sub>	2513 mAhg <sub>electrode<sup>-1</sup></sub> @ 100 mAg <sup>-1</sup>	Current work
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.9</sub> Ni <sub>0.1</sub> O <sub>3</sub>	3556 mAhg <sub>electrode<sup>-1</sup></sub> @ 100 mAg <sup>-1</sup>	Current work
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.7</sub> Ni <sub>0.3</sub> O <sub>3</sub>	2499 mAhg <sub>electrode<sup>-1</sup></sub> @ 100 mAg <sup>-1</sup>	Current work
La <sub>0.7</sub> Sr <sub>0.3</sub> Mn <sub>0.5</sub> Ni <sub>0.5</sub> O <sub>3</sub>	2346 mAhg <sub>electrode<sup>-1</sup></sub> @ 100 mAg <sup>-1</sup>	Current work
La <sub>0.8</sub> Sr <sub>0.2</sub> Mn <sub>0.6</sub> Ni <sub>0.4</sub> O <sub>3</sub>	5364 mAhg <sub>carbon<sup>-1</sup></sub> @ 50 mAg <sup>-1</sup>	[1]
La <sub>0.8</sub> Sr <sub>0.2</sub> MnO <sub>3</sub>	4408 mAhg <sub>carbon<sup>-1</sup></sub> @ 50 mAg <sup>-1</sup>	[1]

- [1] Z. Wang *et al.*, “Nickel-Doped La<sub>0.8</sub>Sr<sub>0.2</sub>Mn<sub>1-x</sub>Ni<sub>x</sub>O<sub>3</sub> Nanoparticles Containing Abundant Oxygen Vacancies as an Optimized Bifunctional Catalyst for Oxygen Cathode in Rechargeable Lithium-Air Batteries,” *ACS Appl Mater Interfaces*, vol. 8, no. 10, pp. 6520–6528, Mar. 2016, doi: 10.1021/acsami.6b00296.