

1     **Urea-Nonstoichiometric Co-modulated LaMnO<sub>3</sub> for Ultra-High**  
2           **Gaseous Hg<sup>0</sup> Uptake Across a Broad Temperature Range**

3     Runlong Hao<sup>a,b,1</sup>, Zhen Qian<sup>a,b,1</sup>, Xiaomeng Zuo<sup>a</sup>, Peng Qin<sup>a</sup>, Xiaojie Yang<sup>a</sup>, Zhao Ma<sup>a,b</sup>, Bo Yuan<sup>\*,a,b</sup>

4     <sup>a</sup> *Hebei Key Lab of Power Plant Flue Gas Multi-Pollutants Control, Department of Environmental Science*  
5     *and Engineering, North China Electric Power University, Baoding, 071003, PR China.*

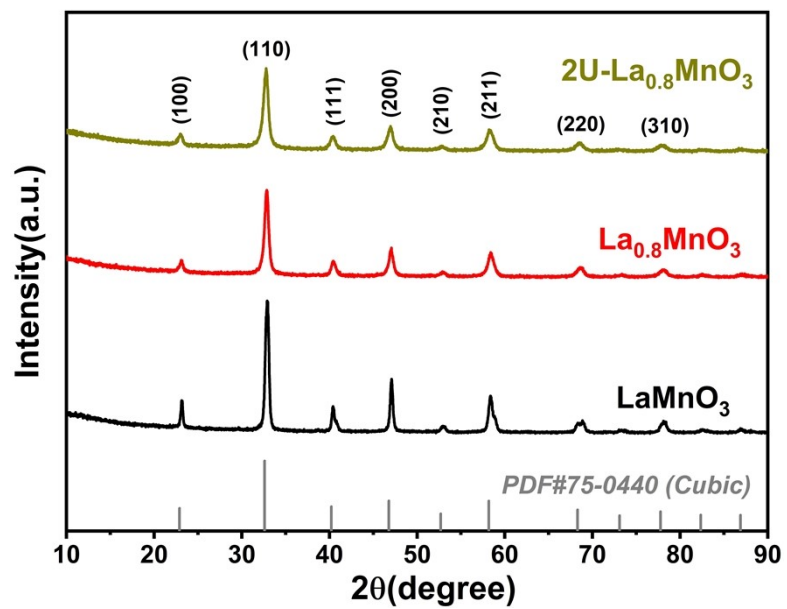
6     <sup>b</sup> *MOE Key Laboratory of Resources and Environmental Systems Optimization, College of Environmental*  
7     *Science and Engineering, North China Electric Power University, Beijing, 102206, PR China.*

8     \* *Corresponding authors E-mail: [hnyuanbo0407@163.com](mailto:hnyuanbo0407@163.com).*

9     <sup>1</sup>*Co-first author.*

## 10 **Text S1. Reagents and Materials**

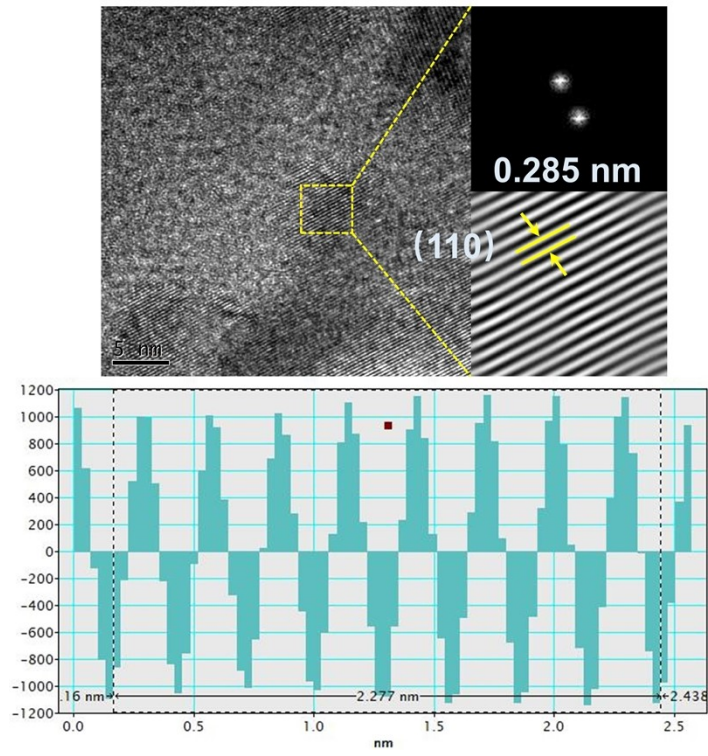
11 All reagents are of analytical grade, purchased from Macklin and Aladdin Reagent Malls (Shanghai,  
12 China). Lanthanum nitrate hexahydrate ( $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ , 99%), manganese nitrate solution ( $\text{Mn}(\text{NO}_3)_2$ , 50  
13 wt.%) citric acid ( $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ , 99.5%) and urea ( $\text{CH}_4\text{N}_2\text{O}$ , 99%) are used for the perovskite synthesis. 0.5  
14 mol/L stannous chloride ( $\text{SnCl}_2$ , 99%) was utilized to confirm whether  $\text{Hg}^{2+}$  was existed in tail gas. 4 wt%  
15 potassium permanganate ( $\text{KMnO}_4$ , 99%) and 10% v/v sulfuric acid ( $\text{H}_2\text{SO}_4$ , 95-98%) were used to capture  
16 the  $\text{Hg}^0$  in tail gas.



17

18

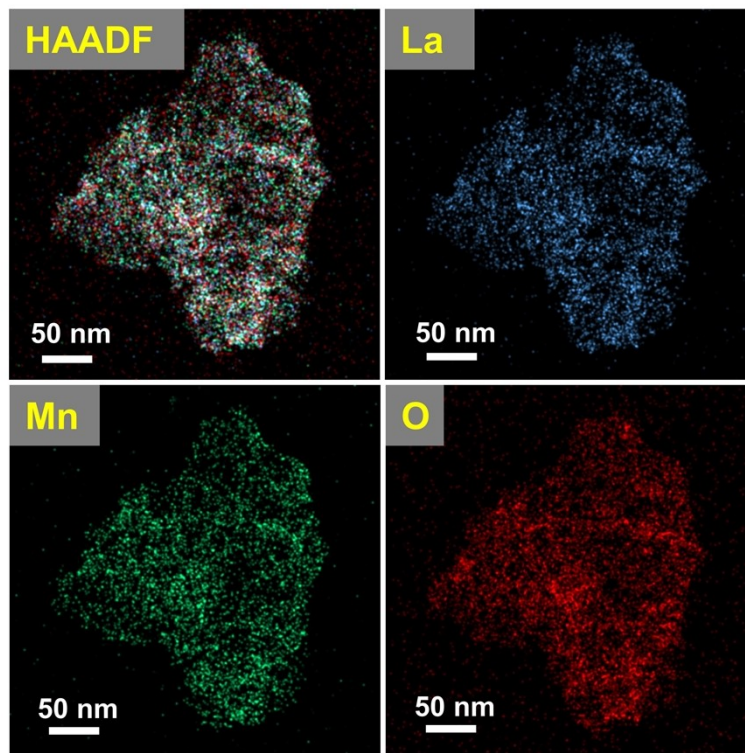
**Fig. S1.** XRD patterns of  $LaMnO_3$ ,  $La_{0.8}MnO_3$  and  $2U-La_{0.8}MnO_3$ .



19

20

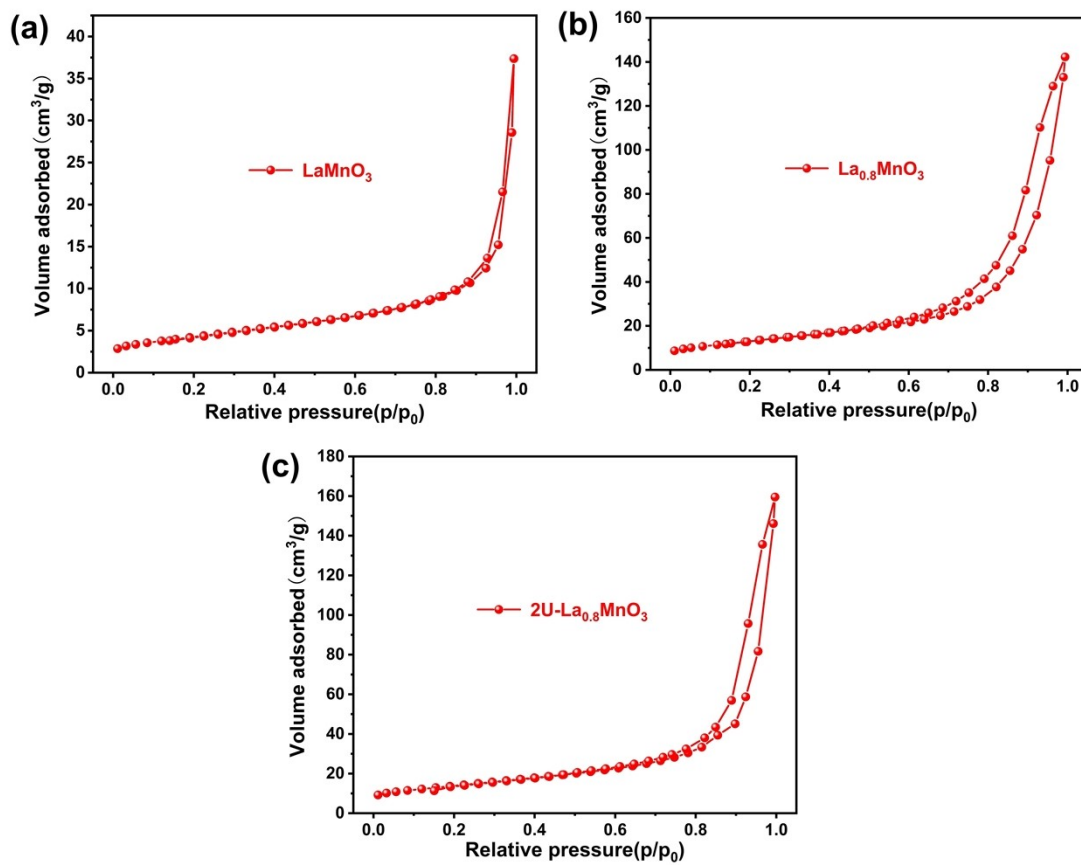
**Fig. S2.** HRTEM images of  $2\text{U-La}_{0.8}\text{MnO}_3$ .



21

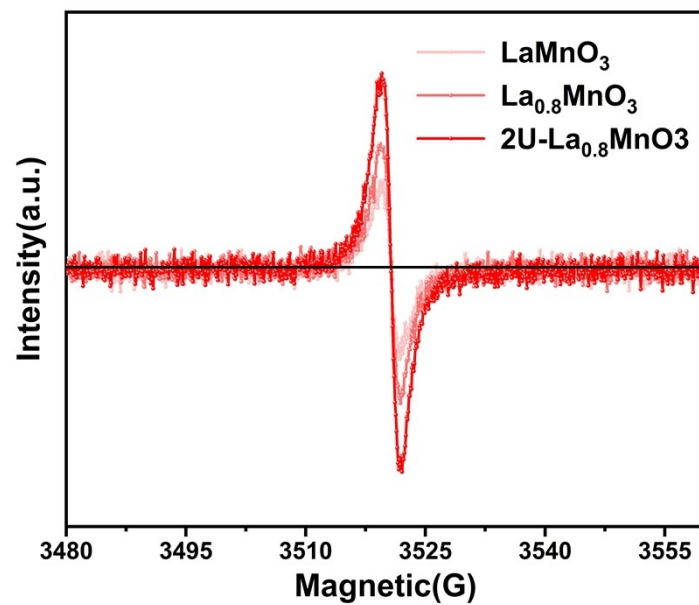
22

**Fig. S3.** EDX element mapping images of 2U-La<sub>0.8</sub>MnO<sub>3</sub>.



23

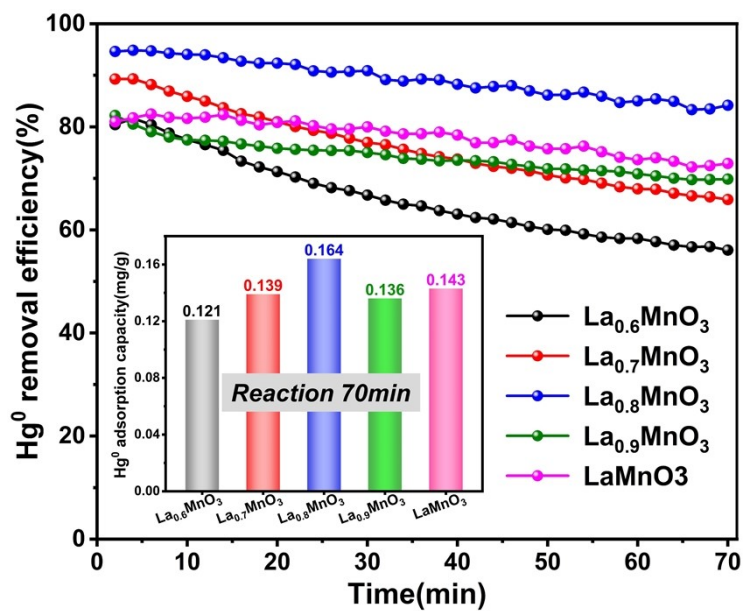
24 **Fig. S4.** The nitrogen adsorption-desorption isotherms of LaMnO<sub>3</sub>, La<sub>0.8</sub>MnO<sub>3</sub> and 2U-La<sub>0.8</sub>MnO<sub>3</sub>.



25

26

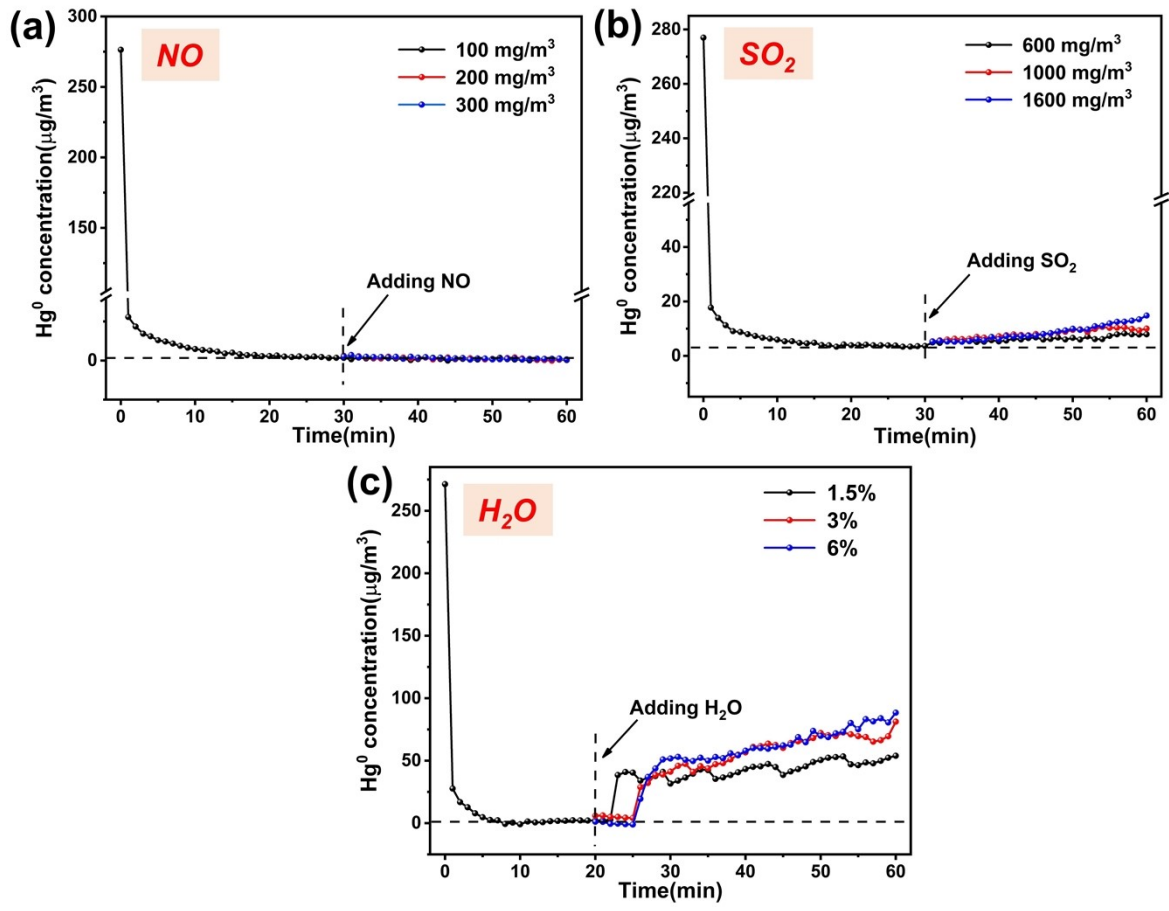
**Fig. S5.** EPR spectra of  $\text{LaMnO}_3$ ,  $\text{La}_{0.8}\text{MnO}_3$  and  $2\text{U-La}_{0.8}\text{MnO}_3$ .



27

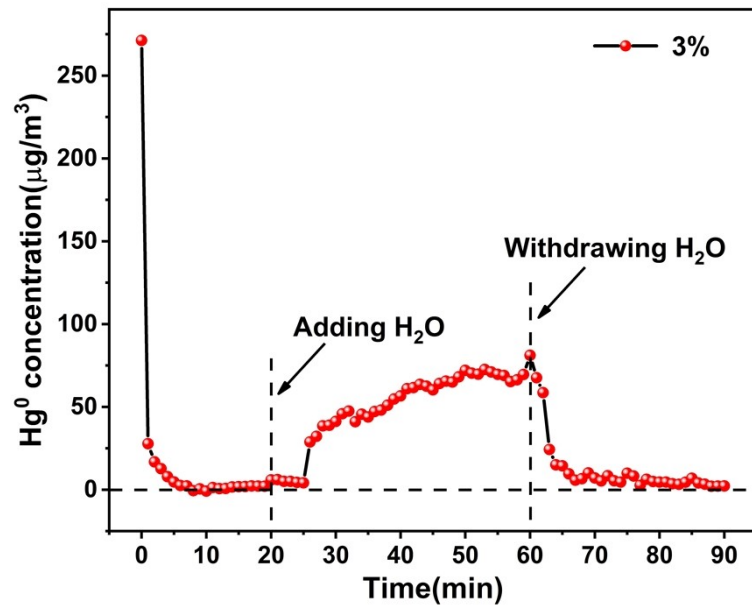
28 **Fig. S6.** The Hg<sup>0</sup> removal efficiency of La<sub>1-x</sub>MnO<sub>3</sub> (x= 0, 0.1, 0.2, 0.3, 0.4), and the inset indicate the  
 29 dynamic adsorption capacity when the Hg<sup>0</sup> breakthrough reached 5%.





**Fig. S7.** Influences of (a)NO, (b)SO<sub>2</sub> and (c)H<sub>2</sub>O on the Hg<sup>0</sup> removal efficiency.

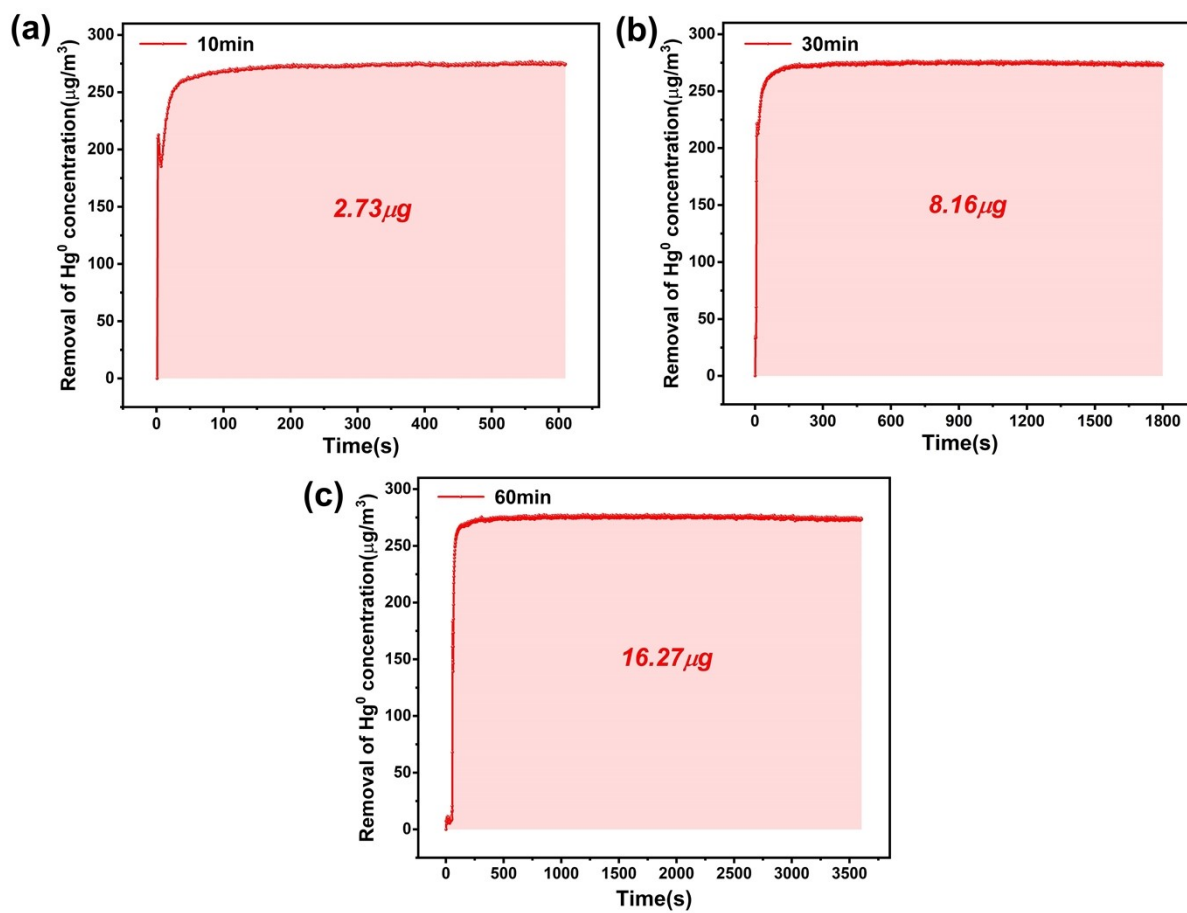
30  
31  
32



33

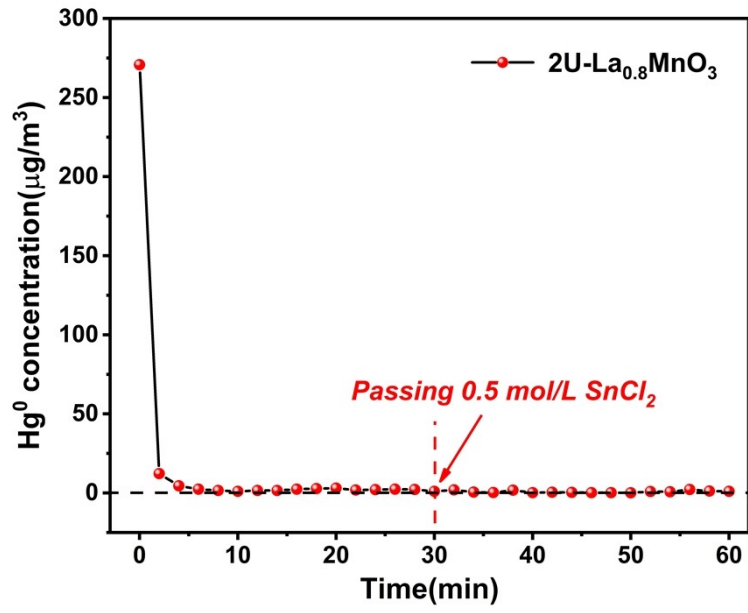
34

**Fig. S8.** The dynamic influence of H<sub>2</sub>O on the Hg<sup>0</sup> removal efficiency.



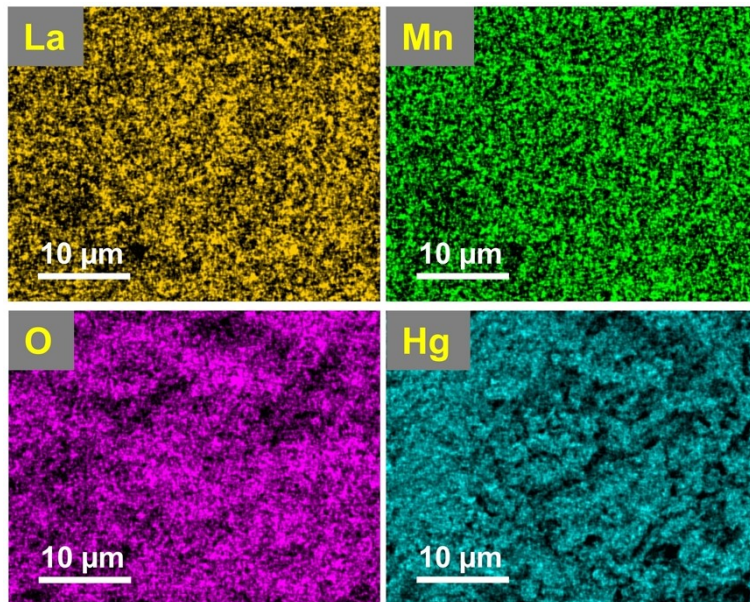
35

36 **Fig. S9.** Experimental calculation of  $\text{Hg}^0$  released after reaction for (a) 10, (b) 30 and (c) 60 min.



37

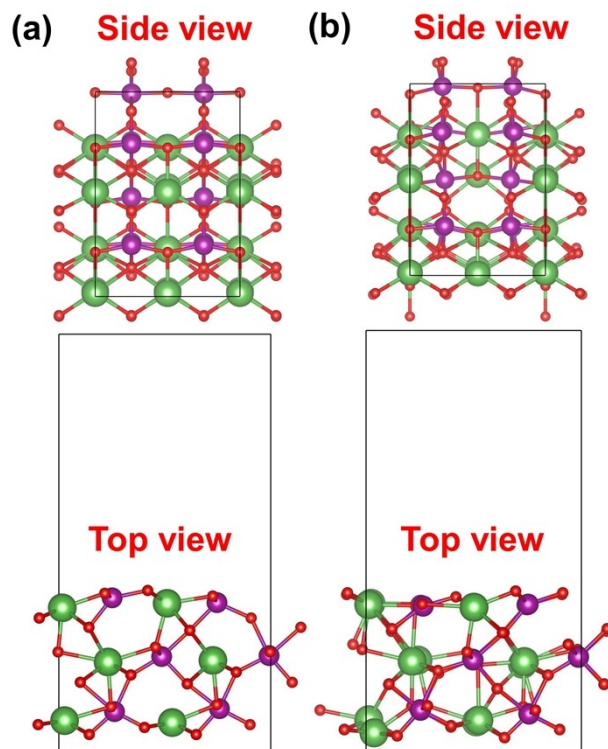
38 **Fig. S10.** The Hg<sup>0</sup> removal performance with and without SnCl<sub>2</sub> solution.



39

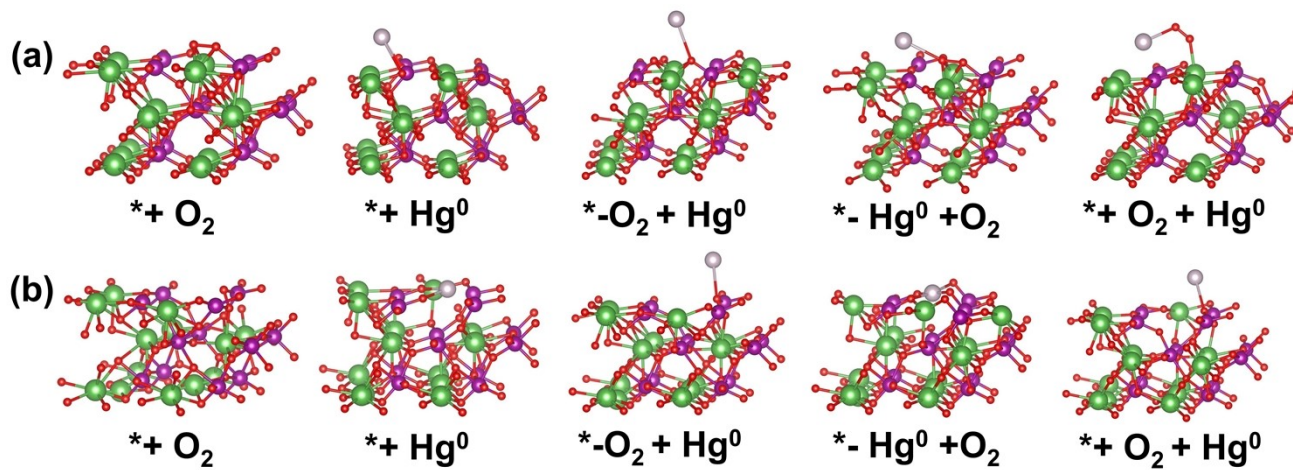
40

**Fig. S11.** EDX element mapping images of spent 2U-La<sub>0.8</sub>MnO<sub>3</sub>.



41

42 **Fig. S12.** Calculated side and top view configurations of (a)  $\text{LaMnO}_3$  and (b)  $2\text{U-La}_{0.8}\text{MnO}_3$ .



43

44 **Fig. S13.** The configurations of O<sub>2</sub> and Hg<sup>0</sup> on (a) LaMnO<sub>3</sub> and (b) 2U-La<sub>0.8</sub>MnO<sub>3</sub> (110) surface (\*).

45 **Table S1.** The comparison of the Hg<sup>0</sup> adsorption capacity of 2U-La<sub>0.8</sub>MnO<sub>3</sub> with other oxides

Oxides	Temperature	Removal efficiency	Hg <sup>0</sup> Adsorption capacity (mg/g)	Breakthrough ratio/adsorption time	References
2U-La <sub>0.8</sub> MnO <sub>3</sub>	40 ~ 250	100%	23.86	100%	<b>This work</b>
Mn/γ-Fe <sub>2</sub> O <sub>3</sub>	200	-	3.54	55%	1
(Fe <sub>2</sub> Ti) <sub>0.8</sub> O <sub>4</sub>	250	-	3.94	23%	2
α-MnO <sub>2</sub>	150	92%	6.94	10 h	3
LaMnO <sub>3</sub>	150	-	7.65	100%	4
CeO <sub>2</sub> /TiO <sub>2</sub>	200 ~ 250	> 90%	0.012	4 h	5
Fe <sub>3</sub> O <sub>4-x</sub> Se <sub>y</sub>	100	100%	8.80	100%	6
MoS <sub>3</sub> /TiO <sub>2</sub>	100	-	14.90	75%	7
La <sub>0.8</sub> Ce <sub>0.2</sub> MnO <sub>3</sub>	50 ~ 200	> 80%	5.83	30 h	8
Fe <sub>2</sub> O <sub>3</sub> /TiO <sub>2</sub>	50 ~ 150	> 95%	2.69	10 h	9
CeO <sub>2</sub> -CrO <sub>x</sub>	50 ~ 100	100%	0.168	6.7 h	10
α-Fe <sub>2</sub> O <sub>3</sub> /SnO <sub>2</sub>	400	99%	4.84	2 h	11
LaFeO <sub>3</sub>	40 ~ 160	> 80%	2.397	1 h	12
Ce-Pd/γ-Al <sub>2</sub> O <sub>3</sub>	250	> 98%	0.038	4.2 h	13



## 46 References

- 47 (1) Yang, S.; Guo, Y.; Yan, N.; Qu, Z.; Xie, J.; Yang, C.; Jia, J. Capture of Gaseous Elemental Mercury  
48 from Flue Gas Using a Magnetic and Sulfur Poisoning Resistant Sorbent Mn/ $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> at Lower  
49 Temperatures. *J. Hazard. Mater.* **2011**, *186*, 508-515.
- 50 (2) Yang, S.; Guo, Y.; Yan, N.; Wu, D.; He, H.; Qu, Z.; Yang, C.; Zhou, Q.; Jia, J. Nanosized Cation-  
51 deficient Fe-Ti spinel: a Novel Magnetic Sorbent for Elemental Mercury Capture from Flue Gas. *ACS Appl.*  
52 *Mater. Interfaces.* **2011**, *3*, 209-217.
- 53 (3) Xu, H.; Qu, Z.; Zhao, S.; Mei, J.; Quan, F.; Yan, N. Different Crystal-forms of One-dimensional MnO<sub>2</sub>  
54 Nanomaterials for the Catalytic Oxidation and Adsorption of Elemental Mercury. *J. Hazard. Mater.* **2015**,  
55 *299*, 86-93.
- 56 (4) Xu, H.; Qu, Z.; Zong, C.; Quan, F.; Mei, J.; Yan, N. Catalytic Oxidation and Adsorption of Hg<sup>0</sup> over  
57 Low-temperature NH<sub>3</sub>-SCR LaMnO<sub>3</sub> Perovskite Oxide from Flue Gas. *Appl. Catal., B* **2016**, *186*, 30-40.
- 58 (5) Li, H.; Wu, C.; Li, Y.; Zhang, J. CeO<sub>2</sub>-TiO<sub>2</sub> Catalysts for Catalytic Oxidation of Elemental Mercury in  
59 Low-rank Coal Combustion Flue Gas. *Environ. Sci. Technol.* **2011**, *45*, 7394-7400.
- 60 (6) Yang, Z.; Li, H.; Yang, Q.; Qu, W.; Zhao, J.; Feng, Y.; Hu, Y.; Yang, J.; Shih, K. Development of  
61 Selenized Magnetite (Fe<sub>3</sub>O<sub>4-x</sub>Se<sub>y</sub>) as an Efficient and Recyclable Trap for Elemental Mercury Sequestration  
62 from Coal Combustion Flue Gas. *Chem. Eng. J.* **2020**, *394*, 125022.
- 63 (7) Mei, J.; Wang, C.; Kong, L.; Liu, X.; Hu, Q.; Zhao, H.; Yang, S. Outstanding Performance of Recyclable  
64 Amorphous MoS<sub>3</sub> Supported on TiO<sub>2</sub> for Capturing High Concentrations of Gaseous Elemental Mercury:  
65 Mechanism, Kinetics, and Application. *Environ. Sci. Technol.* **2019**, *53*, 4480-4489.
- 66 (8) Yang, J.; Na, Y.; Hu, Y.; Zhu, P.; Meng, F.; Guo, Q.; Yang, Z.; Qu, W.; Li, H. Granulation of Mn-based  
67 Perovskite Adsorbent for Cyclic Hg<sup>0</sup> Capture from Coal Combustion Flue Gas. *Chem. Eng. J.* **2023**, *459*,  
68 141679.

- 69 (9) Xing, X.; Zhang, X.; Tang, J.; Cui, L.; Dong, Y. Removal of Gaseous Elemental Mercury from  
70 Simulated Syngas over Fe<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> Sorbents. *Fuel* **2022**, *311*, 122614.
- 71 (10) Ye, D.; Wang, X.; Wang, R.; Hu, Y.; Liu, X.; Liu, H. Relationship between Hg<sup>0</sup> Capture Performance  
72 and Physicochemical Properties of CeO<sub>2</sub>-CrO<sub>x</sub> Mixed Oxides. *J. Environ. Chem. Eng.* **2022**, *10*, 108252.
- 73 (11) Ma, Y.; Xu, T.; Li, L.; Wang, J.; Li, Y.; Zhang, H. Core-shell Nanostructure α-Fe<sub>2</sub>O<sub>3</sub>/SnO<sub>2</sub> Binary  
74 Oxides for the Catalytic Oxidation and Adsorption of Elemental Mercury from Flue Gas. *J. Environ. Chem.*  
75 *Eng.* **2021**, *9*, 105137.
- 76 (12) Pei, H.; Li, X.; Song, Y.; Zhang, M.; Wang, D.; Wu, J.; Wang, F.; Zhang, Y.; Zhao, X.; Jia, T. LaFeO<sub>3</sub>  
77 Perovskite Nanoparticles for Efficient Capture of Elemental Mercury from Coal-fired Flue Gas. *Fuel*. **2022**,  
78 *309*, 122134.
- 79 (13) Huo, Q.; Yue, C.; Wang, Y.; Han, L.; Wang, J.; Chen, S.; Bao, W.; Chang, L.; Xie, K. Effect of  
80 Impregnation Sequence of Pd/Ce/γ-Al<sub>2</sub>O<sub>3</sub> Sorbents on Hg<sup>0</sup> Removal from Coal Derived Fuel Gas.  
81 *Chemosphere*. **2020**, *249*, 126164.