Supplementary Materials for

Interfaces and Oxygen Vacancies-Enriched Catalysts Derived from Cu-Mn-Al Hydrotalcite towards High-Efficient Water-Gas Shift Reaction

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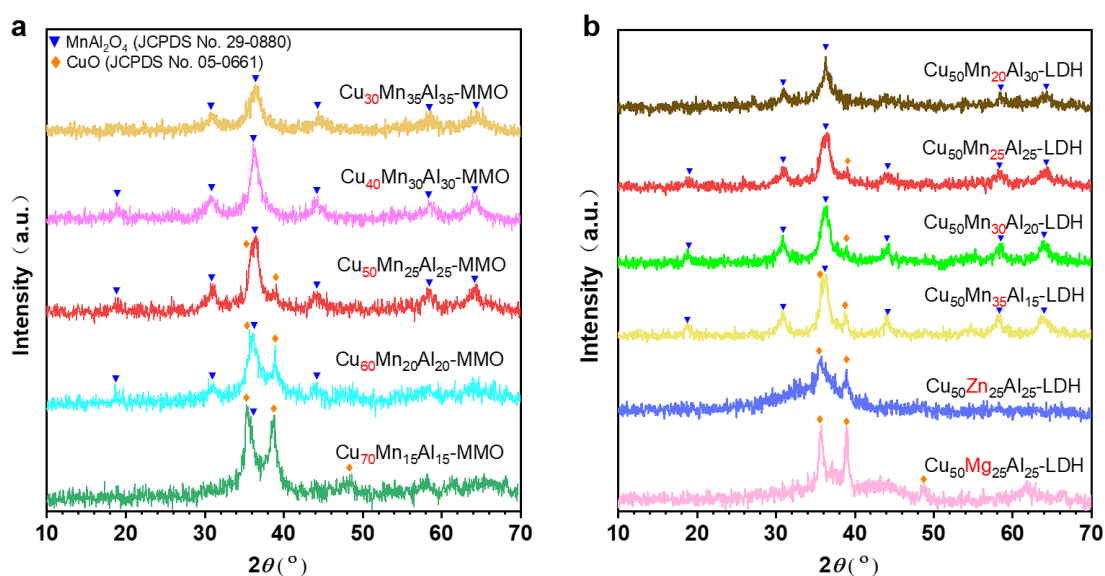* Correspondence: zhuoouhao@126.com (O.Z.)

Figure S1. XRD patterns of the mixed metal oxides (MMO) derived from layered double hydroxides (LDH) samples.
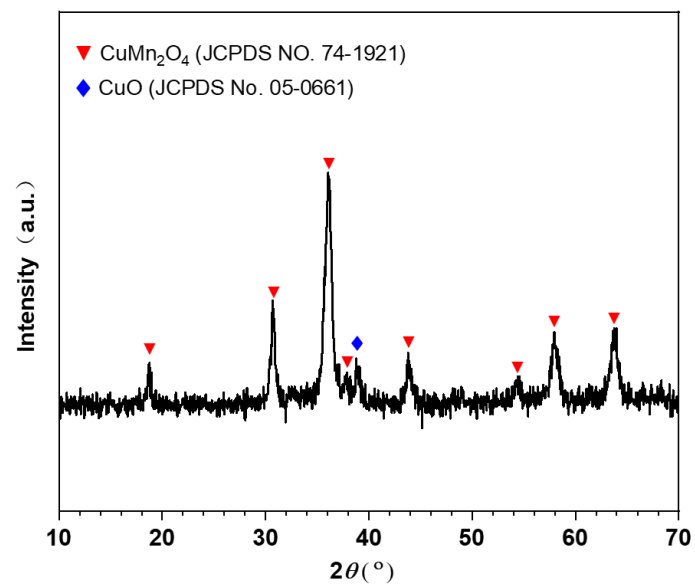
Figure S2. XRD pattern of the CuMn sample (calcined at 500°C for 3 h).
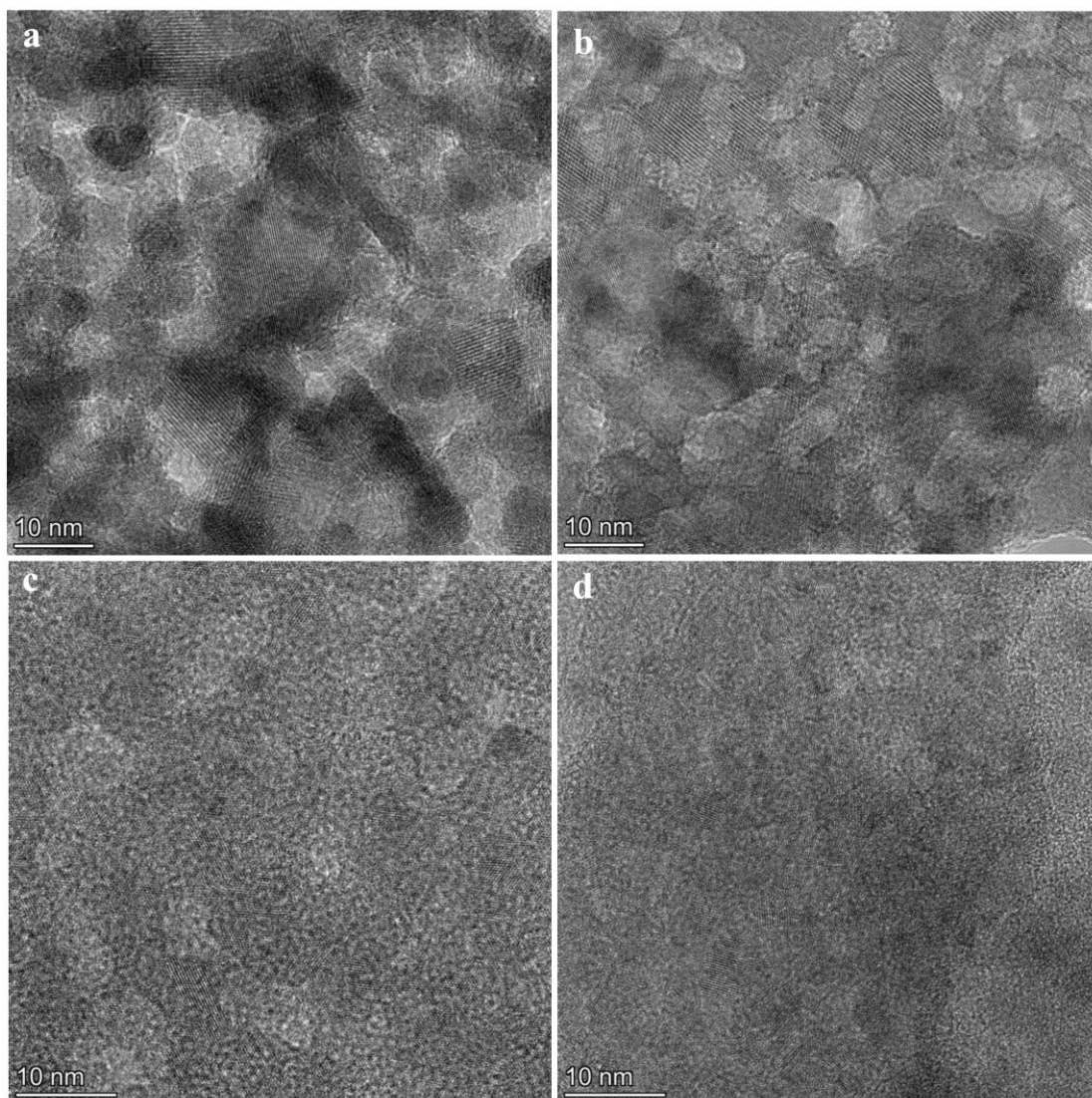
Figure S3. HRTEM images of the MMO samples. (a, b) $Cu_{50}Mn_{25}Al_{25}$-MMO, (c) $Cu_{50}Zn_{25}Al_{25}$-MMO, (d) $Cu_{50}Mg_{25}Al_{25}$-MMO.
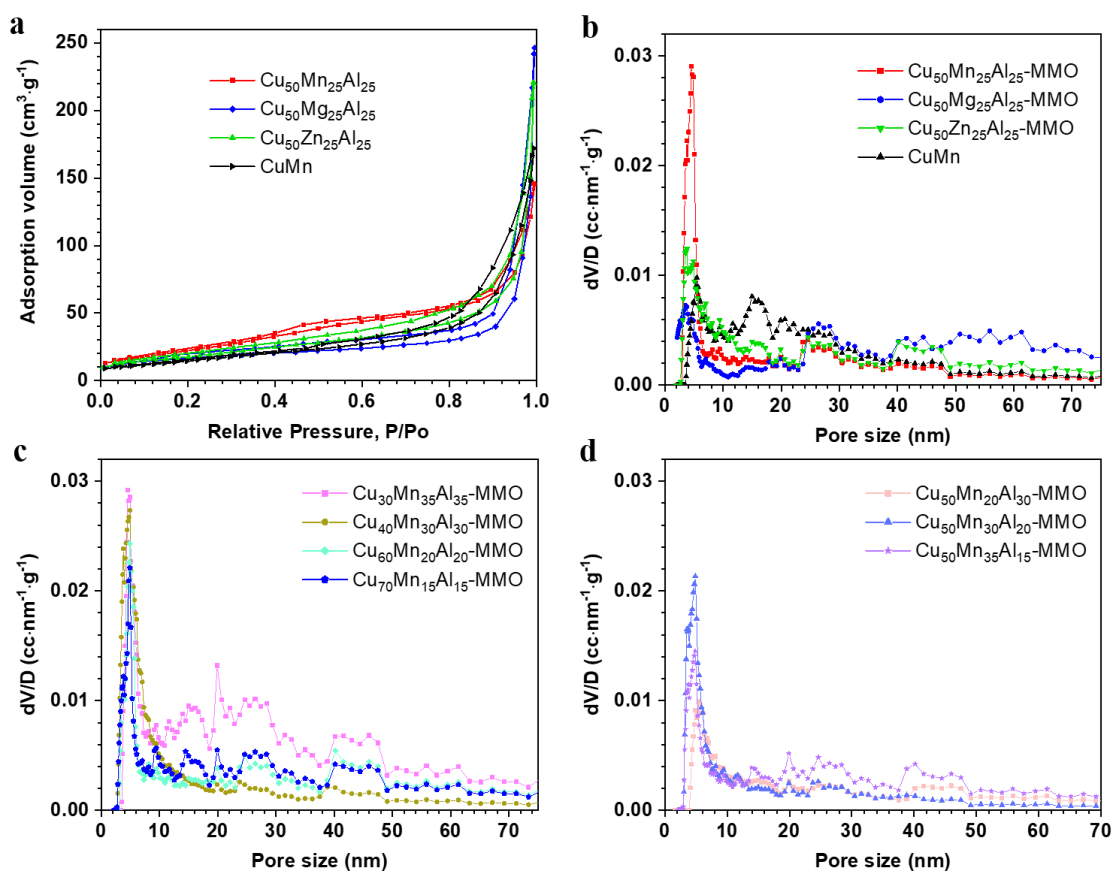
Figure S4. N_2 adsorption/desorption isotherms (a) and pore size distributions (b-d) of the samples.
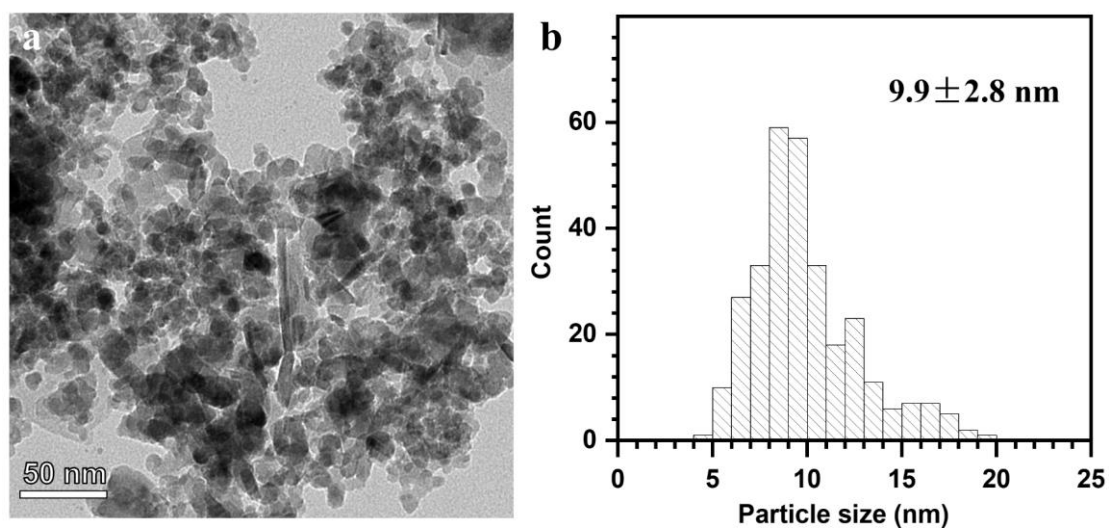

Figure S5. TEM image and particle size distribution for the CuMn sample.

Table S1. Comparison of the activities of the representative catalytic systems with non-noble metal for the WGS reaction

Catalysts	T (K)	Gas feed ($CO : H_2 : CO_2$)	CO/H_2O ratio	WHSV ($mL \cdot g_{cat}^{-1} \cdot h^{-1}$)	CO conversion (%)	Reaction rate [a] ($\mu mol_{CO}^{-1} \cdot g_{cat}^{-1} \cdot s^{-1}$)	Ref.
$Cu_{50}Mn_{25}Al_{25}$-MMO	473	14.9%:27.2%:7.3%	1 : 4	24000	29.2	12.9	This work
	573	14.9%:27.2%:7.3%	1 : 4	24000	96.1	42.6	
$Cu_{50}Mn_{30}Al_{20}$-MMO	473	14.9%:27.2%:7.3%	1 : 4	24000	30.3	13.4	
	573	14.9%:27.2%:7.3%	1 : 4	24000	95.8	42.5	
Commercial $Cu/ZnO/Al_2O_3$	550	5% : 0 : 0	1 : 2	36000	50	12	Zhang, Z., et al. *Nature Communications*, 2017, 8, 488.
$20Cu5Fe/SiO_2$	603	2.5%: 0 : 0	1 : 1	-	-	~3.6	Zhu M., et al. *Angew. Chem. Int. Ed.*, 2019, 58, 9083.
Cu-Mg_HAl_H	523	19%: 0 : 0	1 : 3	5520	97.3	12.7	Lee C. H., et al. *Renewable and Sustainable Energy Reviews*, 2021, 145, 111064.
$Cu_{0.3}Fe_{0.7}O_x$	473	2.2%: 0 : 0	1 : 7	42000	~73	~8.3	Yan H., et al. *Applied Catalysis B: Environmental*, 2018, 226, 182.
MgCuCe catalyst	573	6.3%:37.5%:6.3%	1 : 6	120000	~66	83	Jin S., et al. *Applied Catalysis B: Environmental*, 2021, 284, 119701.
Inverse CeO_2/Cu catalyst	573	2% : 0 : 0	1 : 5	-	-	47.3	Yan, H., et al. *Nature Communications*, 2019, 10, 3470.

T is the reaction temperature. [a] Micromoles of CO per grams of catalyst per second.
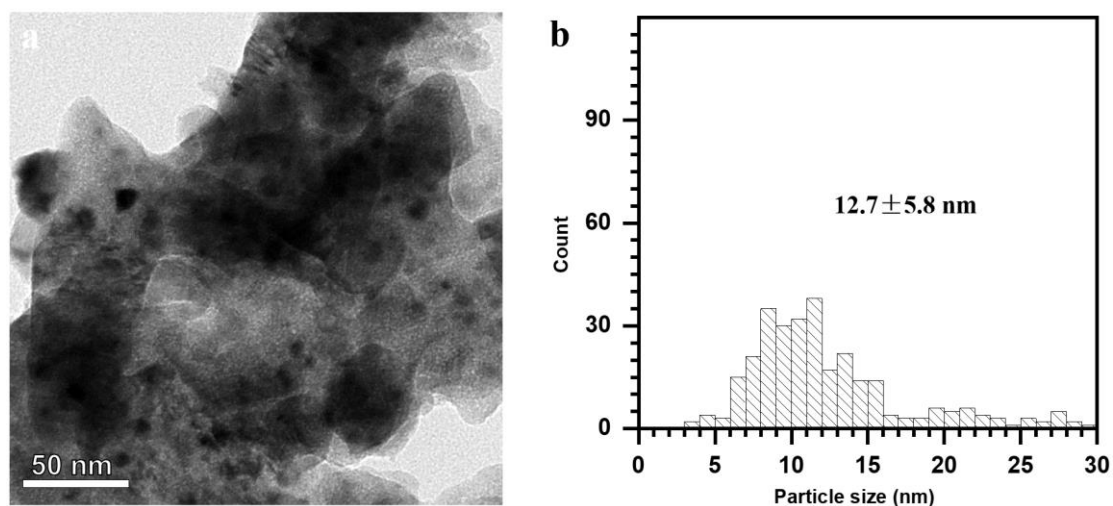
Figure S6. TEM image and particle size distribution for the CuMn catalyst after 5 h on stream in WGS reaction.
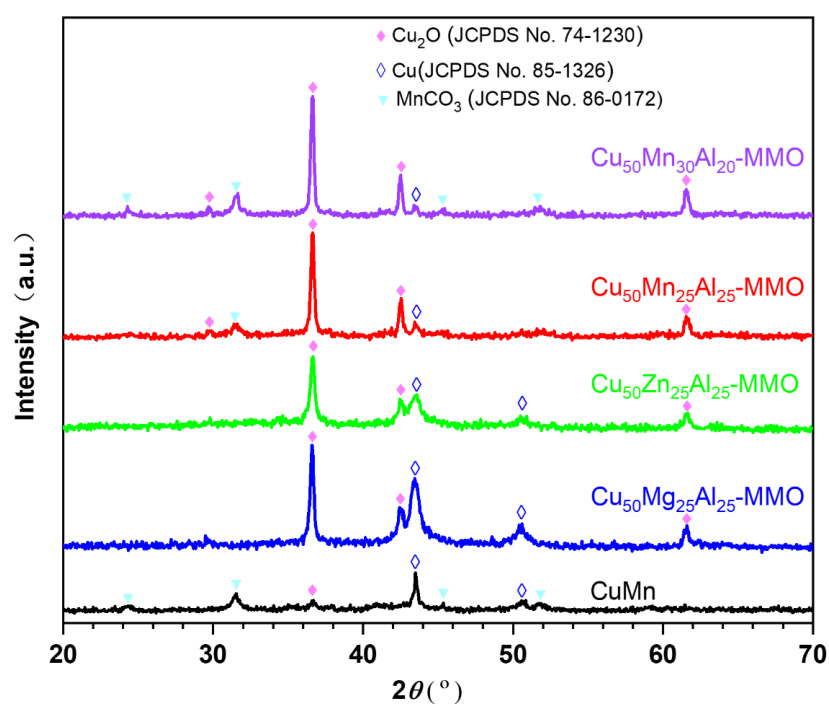

Figure S7. XRD pattern of the spent catalysts in WGS reaction for 5 h.

Table S2. The melting point, Hüttig temperature and Tamman temperature of manganese oxides [a].

Oxides	Melting point [b] (T_{melt})	Hüttig temperature ($T_{Hüt}$)	Tamman temperature (T_{Tam})
MnO	2218 K (1945 ºC)	665 K (392 ºC)	1109 K (836 ºC)
Mn_3O_4	1840 K (1567 ºC)	552 K (279 ºC)	920 K (647 ºC)
M_2O_3	1213 K (940 ºC)	364 K (91 ºC)	607 K (334 ºC)
MnO_2	808 K (535 ºC)	242 K (-31 ºC)	404 K (131 ºC)
$MnCO_3$	- (decompose)	-	-
Cu	1356 K (1083 ºC)	407 K (134 ºC)	678 K (405 ºC)
Cu_2O	1508 K (1235 ºC)	452 K (179 ºC)	754 K (481 ºC)
CuO	1719 K (1446 °C)	516 K (243 ºC)	860 K (587 ºC)

[a] The Hüttig temperature and Tamman temperature are calculated from the corresponding melting point using the following equations: $T_{Hüt} = 0.3T_{melt}$, $T_{Tam} = 0.5T_{melt}$. The temperature used in the calculation process is absolute temperature. [b] The melting point is obtained from the following literatures: Navasery M., et al. *Mordern Physics Letters B*, 2012, 26(6), 1150039; Moiseev G. K., et al. *Inorganic Materials*, 2006, 42, 632; Kosenko A. V., et al. *Journal of Phase Equilibria*, 2001, 22, 12.