

具有优异甲醇耐受性的 Rh 掺杂 PdCu 有序金属间化合物纳米粒子增强氧还原电催化

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Rh-Doped PdCu Ordered Intermetallics for Enhanced Oxygen Reduction Electrocatalysis with Superior Methanol Tolerance

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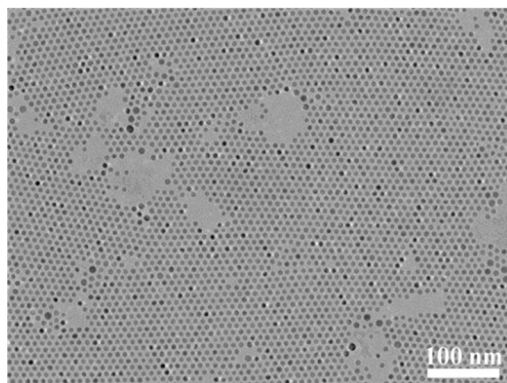


Fig. S1 The low-magnification TEM image of Rh-PdCu NPs.

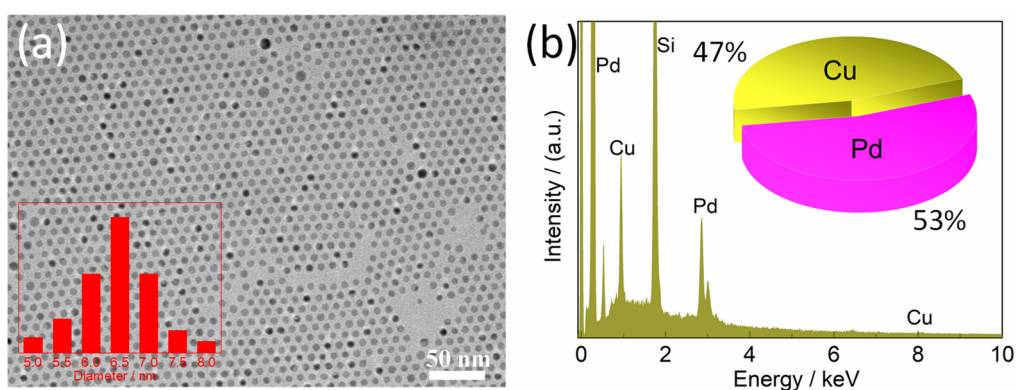


Fig. S2 (a) Representative TEM image (*inset* is the corresponding size distribution) and (b) SEM-EDS spectra of disordered PdCu NPs.

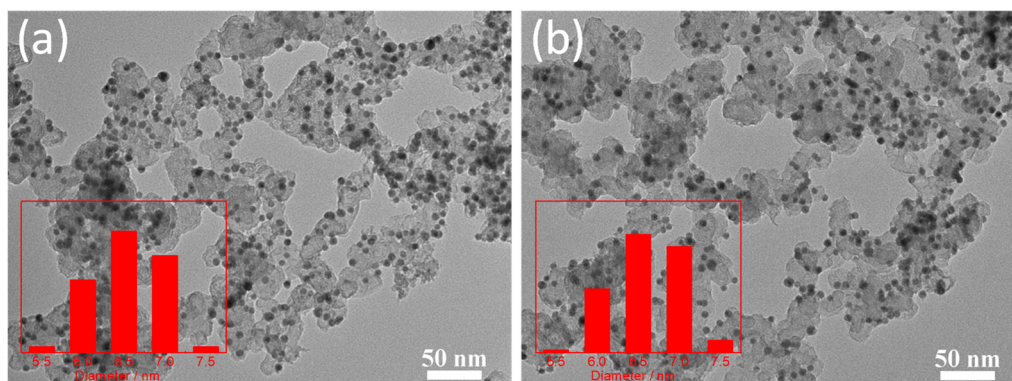


Fig. S3 Representative TEM images of (a) PdCu/C and (b) Rh-PdCu/C (*insets* are the corresponding size distributions).

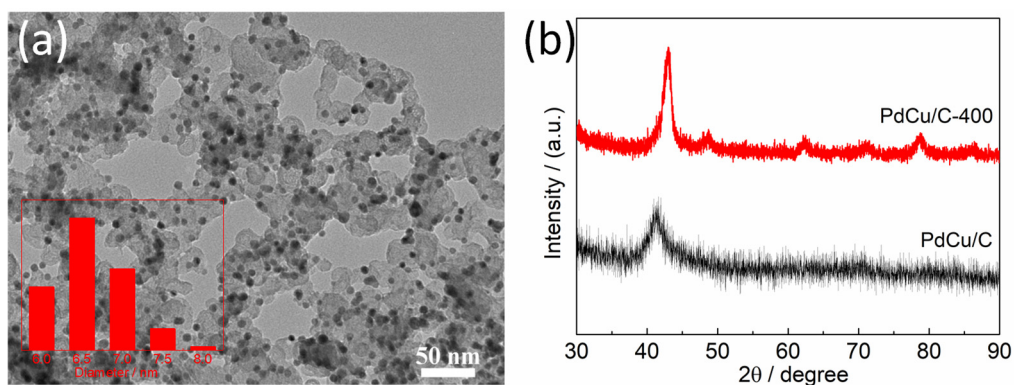


Fig. S4 (a) Representative TEM image of PdCu/C-400 (*inset* is the corresponding size distribution). (b) PXRD patterns of PdCu/C with different annealing temperature.

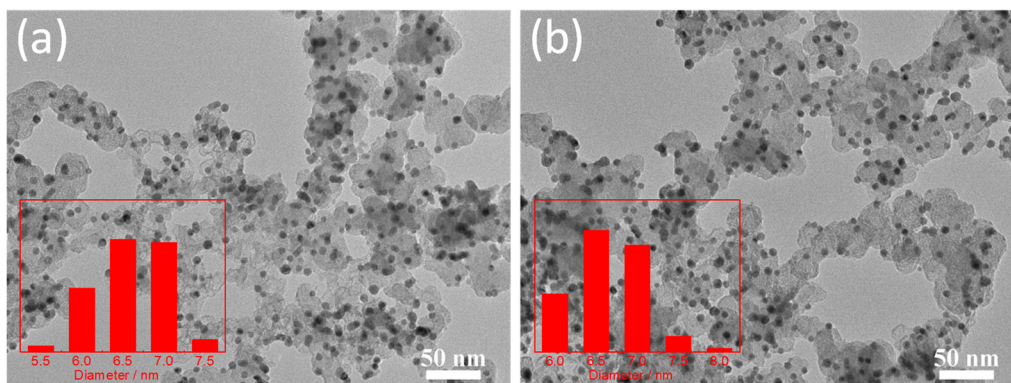


Fig. S5 Representative TEM images of (a) Rh-PdCu/C-300 and (b) Rh-PdCu/C-400 (insets are the corresponding size distributions).

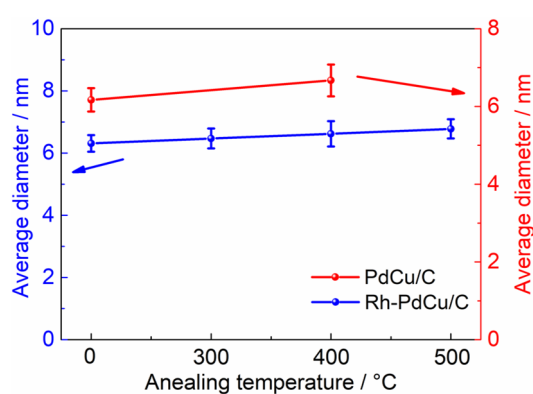


Fig. S6 The change of average diameter of PdCu/C and Rh-PdCu/C with different annealing temperature.

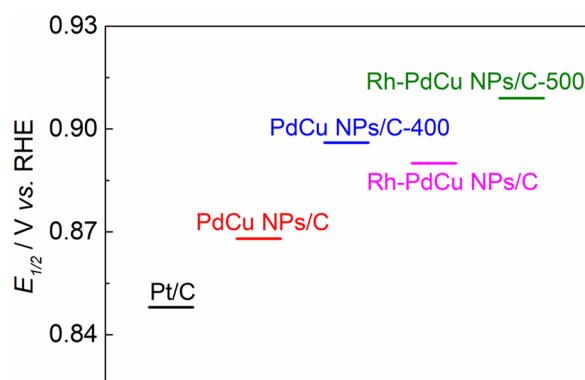


Fig. S7 The half-wave potential ($E_{1/2}$) values of different catalysts.

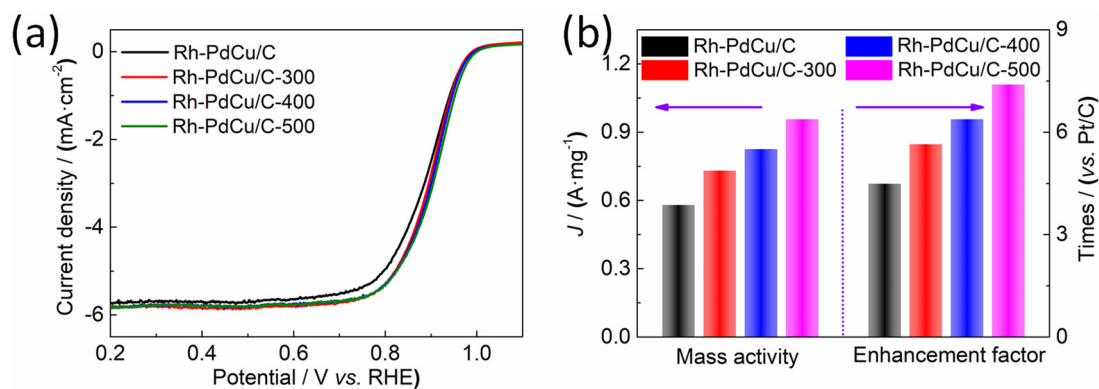


Fig. S8 (a) ORR polarization curves and (b) the mass activities and enhancement factors (vs the commercial Pt/C) at 0.9 V (vs RHE) of Rh-PdCu/C with different annealing temperature.

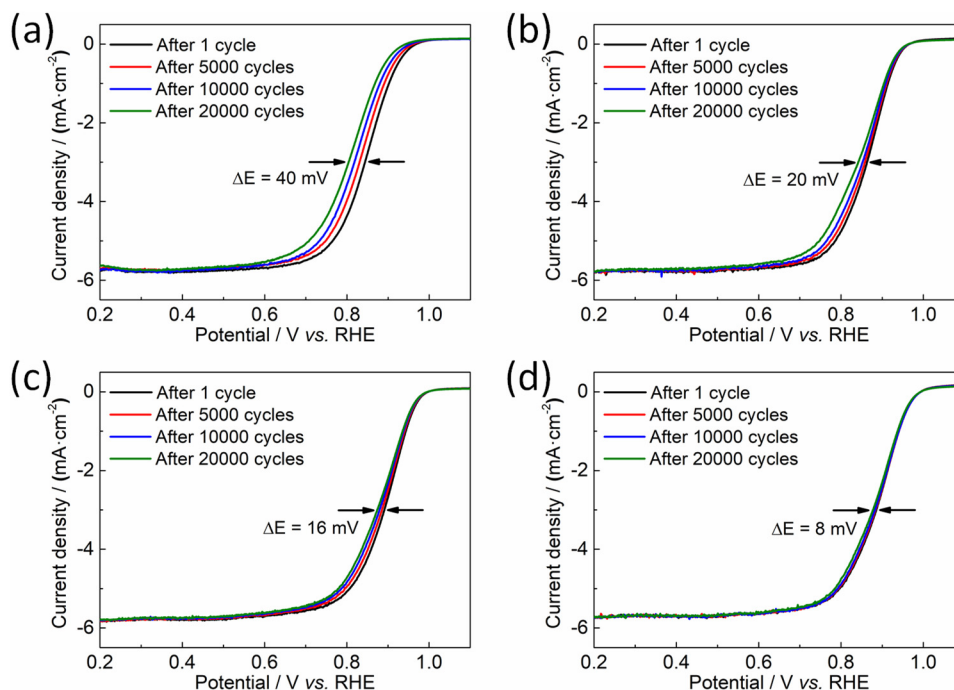


Fig. S9 ORR polarization curves of (a) commercial Pt/C, (b) disordered PdCu/C, (c) ordered PdCu/C and (d) disordered Rh-PdCu/C before and after different potential cycles.

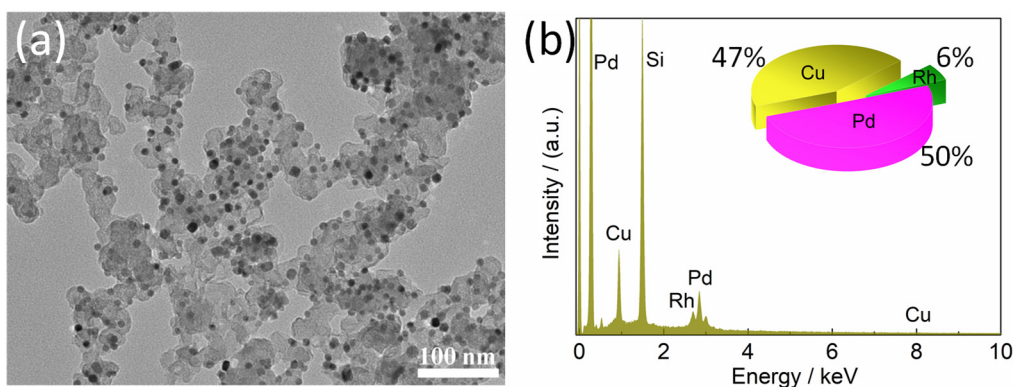


Fig. S10 (a) Representative TEM image and (d) SEM-EDS spectra of Rh-PdCu/C-500 after ADTs.

Table S1 Comparison of ORR electrocatalytic activities of several electrocatalysts in this work with other reported PdCu-based electrocatalysts from recent published works.

No.	Catalysts	Electrolytes	Mass activities ($A \cdot mg^{-1}$)	References
1	Rh-PdCu/C-500	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.96 @ 0.9 \text{ V vs RHE}$	This work
2	Rh-PdCu/C	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.58 @ 0.9 \text{ V vs RHE}$	This work
3	PdCu/C-400	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.69 @ 0.9 \text{ V vs RHE}$	This work
4	PdCu/C	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.30 @ 0.9 \text{ V vs RHE}$	This work
5	PdCu nanocubes	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$\sim 1.1 @ -0.15 \text{ V vs Ag/AgCl}$	Ref. 1
6	Au/Cu ₄₀ Pd ₆₀ NPs	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.43 @ -0.1 \text{ V vs Ag/AgCl}$	Ref. 2
7	G-Cu ₃ Pd NCPs	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.43 @ -0.4 \text{ V vs SCE}$	Ref. 3
9	PdCuCo NPs/C-375°C	$0.1 \text{ mol} \cdot L^{-1} \text{ NaOH}$	$0.13 @ 0.90 \text{ V vs RHE}$	Ref. 4
10	PdCuNi-AB-t/C	$0.1 \text{ mol} \cdot L^{-1} \text{ NaOH}$	$0.45 @ 0.90 \text{ V vs RHE}$	Ref. 5
11	5.8 nm-PdCu NPs/C	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.307 @ 0.80 \text{ V vs RHE}$	Ref. 6
12	Pd ₅₉ Cu ₃₀ Co ₁₁ nanoalloys	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.38 @ 0.90 \text{ V vs RHE}$	Ref. 7
13	50 nm CuPd TPs	$0.1 \text{ mol} \cdot L^{-1} \text{ KOH}$	$0.29 @ 0.90 \text{ V vs RHE}$	Ref. 8

References

- (1) Gao, Q.; Ju, Y. M.; An, D.; Gao, M. R.; Cui, C. H.; Liu, J. W.; Cong, H. P.; Yu, S. H. *ChemSusChem* **2013**, *6*, 1878. doi: 10.1002/cssc.201300404
- (2) Guo, S.; Zhang, X.; Zhu, W.; He, K.; Su, D.; Mendoza-Garcia, A.; Ho, S. F.; Lu, G.; Sun, S. *J. Am. Chem. Soc.* **2014**, *136*, 15026.
doi: 10.1021/ja508256g
- (3) Zheng, Y.; Zhso, S.; Liu, S.; Yin, H.; Chen, Y. Y.; Bao, J.; Han, M.; Dai, Z. *ACS Appl. Mater. Interfaces* **2015**, *7*, 5347. doi: 10.1021/acsami.5b01541
- (4) Jiang, K.; Wang, P.; Guo, S.; Zhang, X.; Shen, X.; Lu, G.; Su, D.; Huang, X. *Angew. Chem. Int. Ed.* **2016**, *55*, 9030. doi: 10.1002/anie.201603022
- (5) Wang, H.; Luo, W.; Zhu, L.; Zhao, Z.; E, B.; Tu, W.; Ke, X.; Sui, M.; Chen, C.; Chen, Q.; *et al.* *Adv. Funct. Mater.* **2018**, *28*, 1707219.
doi: 10.1002/adfm.201707219
- (6) Yang, Y.; Dai, C.; Wu, D.; Liu, Z.; Cheng, D. *ChemElectroChem* **2018**, *5*, 2571. doi: 10.1002/celec.201800332
- (7) Li, C.; Yuan, Q.; Ni, B.; He, T.; Zhang, S.; Long, Y.; Gu, L.; Wang, X. *Nat. Commun.* **2018**, *9*, 3702. doi: 10.1038/s41467-018-06043-1
- (8) Zhang, L.; Chen, S.; Dai, Y.; Shen, Z.; Wei, M.; Huang, R.; Li, H.; Zheng, T.; Zhang, Y.; Zhou, S.; *et al.* *ChemCatChem* **2018**, *10*, 925.
doi: 10.1002/cctc.201701578