

High-frequency ultrasonic pyrolysis of 200 nm ultrafine Fe-doped NiO hollow spheres for efficient oxygen evolution catalysis

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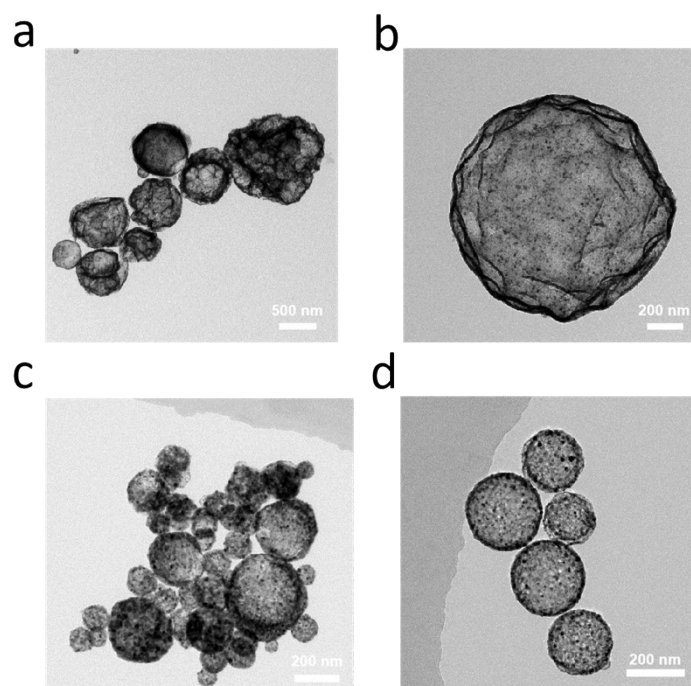
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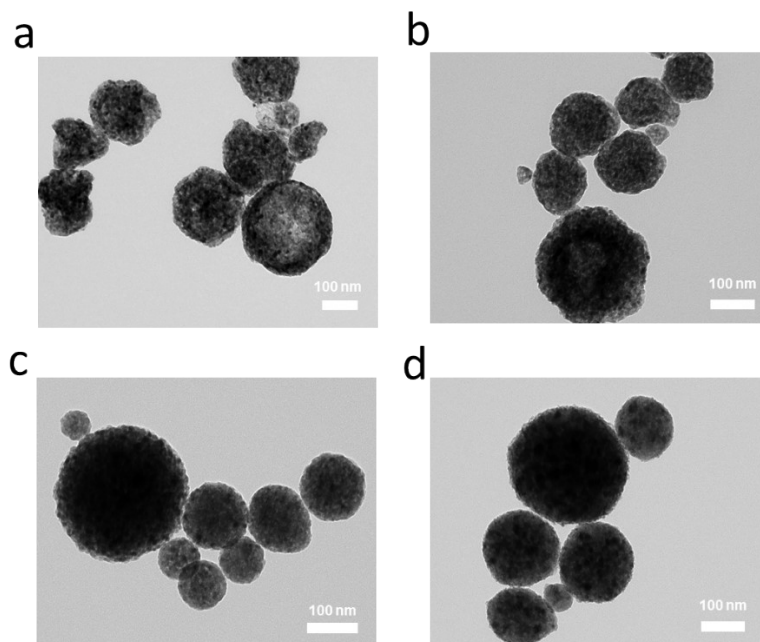
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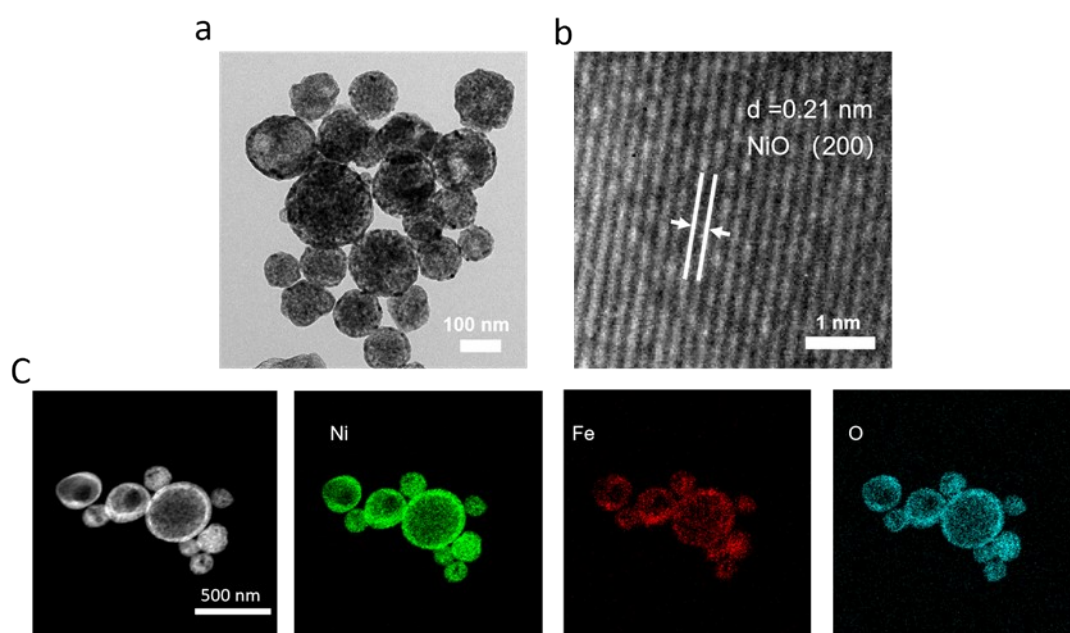
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Supplementary Figure 1. (a, b) TEM images of NiO hollow nanospheres at 1.7 MHz frequency. (c, d) TEM images of NiO hollow nanospheres at 3.0 MHz frequency.



Supplementary Figure 2. TEM images of $\text{Ni}_{0.8}\text{Fe}_{0.2}\text{O}$, $\text{Ni}_{0.7}\text{Fe}_{0.3}\text{O}$, $\text{Ni}_{0.6}\text{Fe}_{0.4}\text{O}$ and $\text{Ni}_{0.5}\text{Fe}_{0.5}\text{O}$.



Supplementary Figure 3. (a) TEM image, (b) HRTEM image and (c) EDS mapping of $\text{Ni}_{0.9}\text{Fe}_{0.1}\text{O}$ hollow nanospheres after OER.

Supplementary Table 1. Comparisons of the η_{10} of $\text{Ni}_{0.9}\text{Fe}_{0.1}\text{O}$ hollow nanospheres with representative electrocatalysts reported previously.

Catalyst	Electrode substrate	Overpotential (mV)	Electrolyte	Reference
$\text{Ni}_{0.9}\text{Fe}_{0.1}\text{O}$ hollow nanospheres	Glassy carbon	288	1 M KOH	This work
Hollow cobalt nickel oxides microspheres	Glassy carbon	310	1 M KOH	1
Porous NiO hollow spheres	Glassy carbon	323	1 M KOH	2
Porous NiO nano flowers	Glassy carbon	346	1 M KOH	2
3D porous carbon@Ni/NiO	Glassy carbon	353	1 M KOH	3
Ni-Co mixed oxide nanocages	Glassy carbon	380	1 M KOH	4
Hollow NiCo_2O_4 arrays	Glassy carbon	340	1 M KOH	5
$\text{CoNiO}_2/\text{SNC}$	Glassy carbon	280	1 M KOH	6
CoNiO_x/NC	Glassy carbon	335	1 M KOH	6
Multi-shelled $\text{Co}_{0.5}\text{Ni}_{0.5}$ oxide/phosphide	Glassy carbon	268	1 M KOH	7
Porous nanoscale NiO/NiCo_2O_4	Glassy carbon	264	1 M KOH	8
U-$\text{NiO}/\text{NiCo}_2\text{O}_4$	Glassy carbon	387	0.1M KOH	9
C-$\text{NiO}/\text{NiCo}_2\text{O}_4$	Glassy carbon	430	0.1M KOH	9
Multilayer hollow MnCo_2O_4 microsphere	Glassy carbon	400	0.1M KOH	10
3D NiFe_2O_4 hollow spheres	Glassy carbon	370	0.1M KOH	11

References :

- 1 J. Zhao, X. Wang, X. Wang, Y. Li, X. Yang, G. Li and F. Li, Ultrathin Porous Nanosheet-Assembled Hollow Cobalt Nickel Oxide Microspheres with Optimized Compositions for Efficient Oxygen Evolution Reaction, *Inorg. Chem. Front.*, 2018, **5**, 1886-1893.
- 2 B. P. Reddy, K. Mallikarjuna, M. Kumar, M. C. Sekhar, Y. Suh and S. Park, Highly Porous Metal Organic Framework Derived NiO Hollow Spheres and Flowers for Oxygen Evolution Reaction and Supercapacitors, *Ceram. Int.*, 2021, **47**, 3312-3321.
- 3 Y. Li, J. Huang, G. Rao, C. Wu, X. Du, Y. Sun, X. Wang and C. Yang, Enhanced Water Oxidation Activity of 3D Porous Carbon by Incorporation of Heterogeneous Ni/NiO Nanoparticles, *Appl. Surf. Sci.*, 2020, **530**, 147192.
- 4 L. Han, X. Yu and X. W. D. Lou, Formation of Prussian-Blue-Analog Nanocages Via a Direct Etching Method and their Conversion Into Ni-Co-Mixed Oxide for Enhanced Oxygen Evolution, *Adv. Mater.*, 2016, **28**, 4601-4605.
- 5 C. Guan, X. Liu, W. Ren, X. Li, C. Cheng and J. Wang, Rational Design of Metal-Organic Framework Derived Hollow NiCo₂O₄ Arrays for Flexible Supercapacitor and Electrocatalysis, *Adv. Energy Mater.*, 2017, **7**, 1602391.
- 6 Q. Zhang, W. Han, Z. Xu, Y. Li, L. Chen, Z. Bai, L. Yang and X. Wang, Hollow Waxberry-Like Cobalt-Nickel Oxide/S,N-Codoped Carbon Nanospheres as a Trifunctional electrocatalyst for OER, ORR, and HER, *RSC Adv.*, 2020, **10**, 27788-27793.
- 7 Y. Yang, H. Wan, G. Chen, N. Zhang, J. Li, W. Ma, X. Liu and R. Ma, Multi-Shelled Cobalt-Nickel Oxide/Phosphide Hollow Spheres for an Efficient Oxygen Evolution Reaction, *Dalton Trans.*, 2020, **49**, 10918-10927.
- 8 S. Sun, X. Jin, B. Cong, X. Zhou, W. Hong and G. Chen, Construction of porous nanoscale NiO/NiCo₂O₄ heterostructure for highly enhanced electrocatalytic oxygen evolution activity, *J. Catal.*, 2019, **379**, 1-9.
- 9 A. Cetin and E. N. Esenturk, Hierarchical Nanowire and Nanoplate-Assembled NiCo₂O₄-NiO Biphasic Microspheres as Effective Electrocatalysts for Oxygen

Evolution Reaction, *Mater. Today Chem.*, 2019, **14**, 100215.

- 10 K. Zeng, W. Li, Y. Zhou, Z. Sun, C. Lu, J. Yan, J. Choi and R. Yang, Multilayer Hollow MnCo_2O_4 Microsphere with Oxygen Vacancies as Efficient Electrocatalyst for Oxygen Evolution Reaction, *Chem. Eng. J. (Lausanne, Switz.)*, 2021, **421**, 127831.
- 11 A. Martínez-Lázaro, A. Rico-Zavala, F. I. Espinosa-Lagunes, J. Torres-González, L. Álvarez-Contreras, M. P. Gurrola, L. G. Arriaga, J. Ledesma-García and E. Ortiz-Ortega, Microfluidic Water Splitting Cell Using 3D NiFe_2O_4 Hollow Spheres, *J. Power Sources*, 2019, **412**, 505-513.