Electronic Supplementary Material (ESI) for Catalysis Science &Technology.

This journal is © The Royal Society of Chemistry 2019

Supporting Information

pH Universal Ru@N-doped Carbon Catalyst for Efficient and Fast Hydrogen Evolution

Baocheng Zheng,[a] Li Ma,[a] Bing Li, [a] Dong Chen, [a] Xueliang Li, [a] Jianbo He [a], Jianhui Xie* [a], Marc Robert*,[b] and Tai-Chu Lau*,[c]

[a]Anhui Province Key Laboratory of Advanced Catalytic Materials and Reaction Engineering, School of Chemistry and Chemical Engineering, Hefei University of Technology, Hefei 230009, People's Republic of China.

[b]Université de Paris, Laboratoire d'Electrochimie Moléculaire, CNRS, F-75006 Paris, France.

[c]Department of Chemistry, City University of Hong Kong, Tat Chee, Avenue, Kowloon Tong, Hong Kong, People's Republic of China.

*Corresponding Author's Email: *jianhuixie@hfut,edu.cn*, robert@u-paris.fr,

bhtclau@cityu.edu.hk

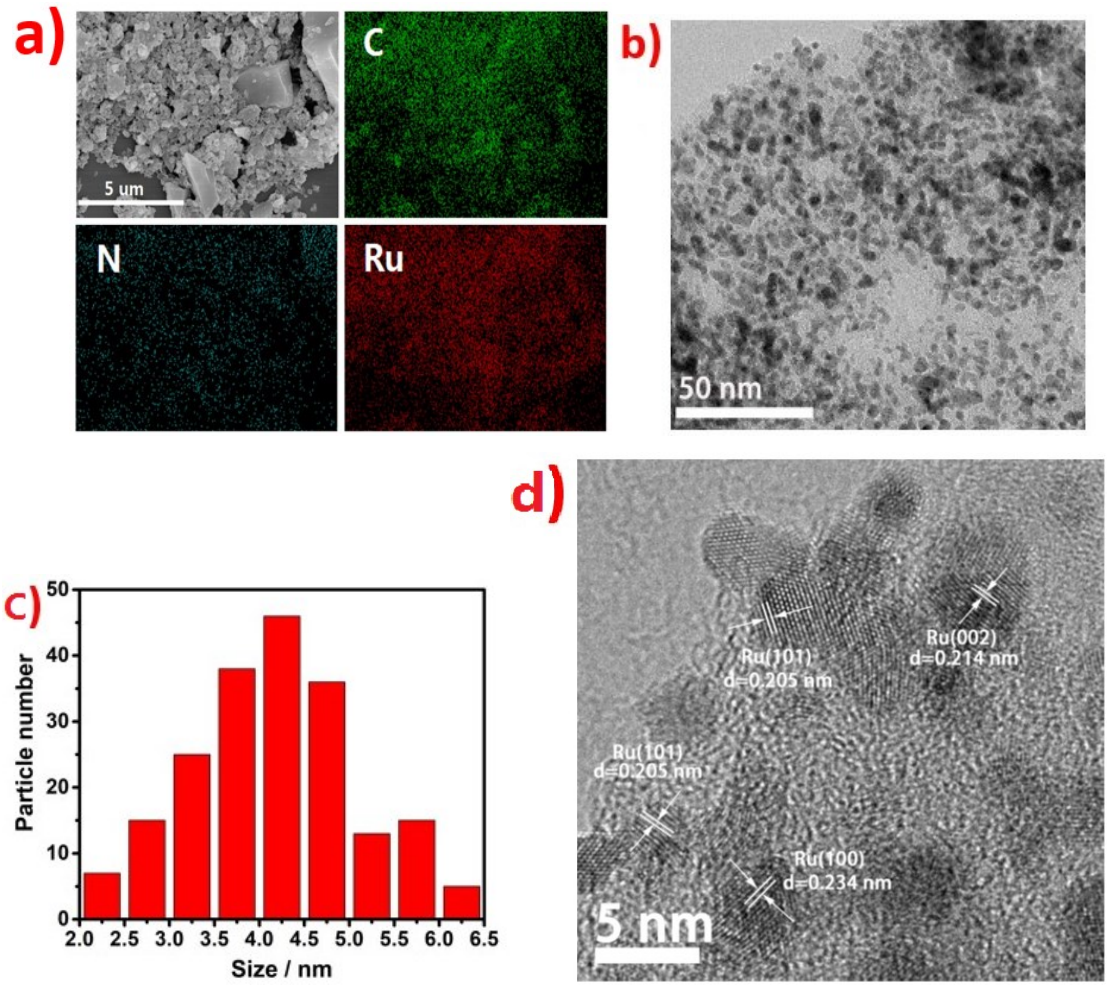


Figure S1. a) SEM and EDS-mapping images of Ru@NC(1:2). b) TEM images. c) The corresponding particle size distribution of the Ru nanoparticles. d) HRTEM image.

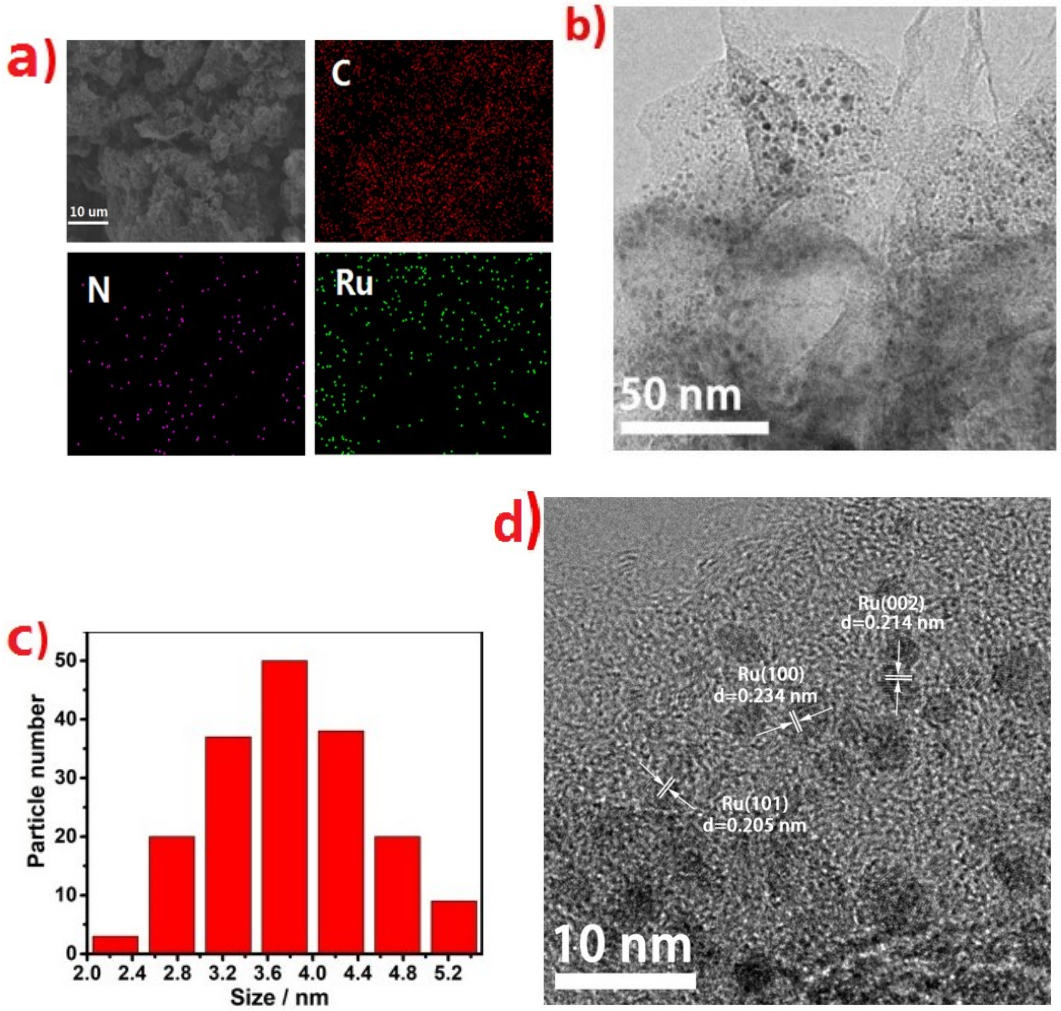


Figure S2. a) SEM and EDS-mapping images of Ru@NC(1:10). b) TEM images. c) The corresponding particle size distribution of the Ru nanoparticles. d) HRTEM image.

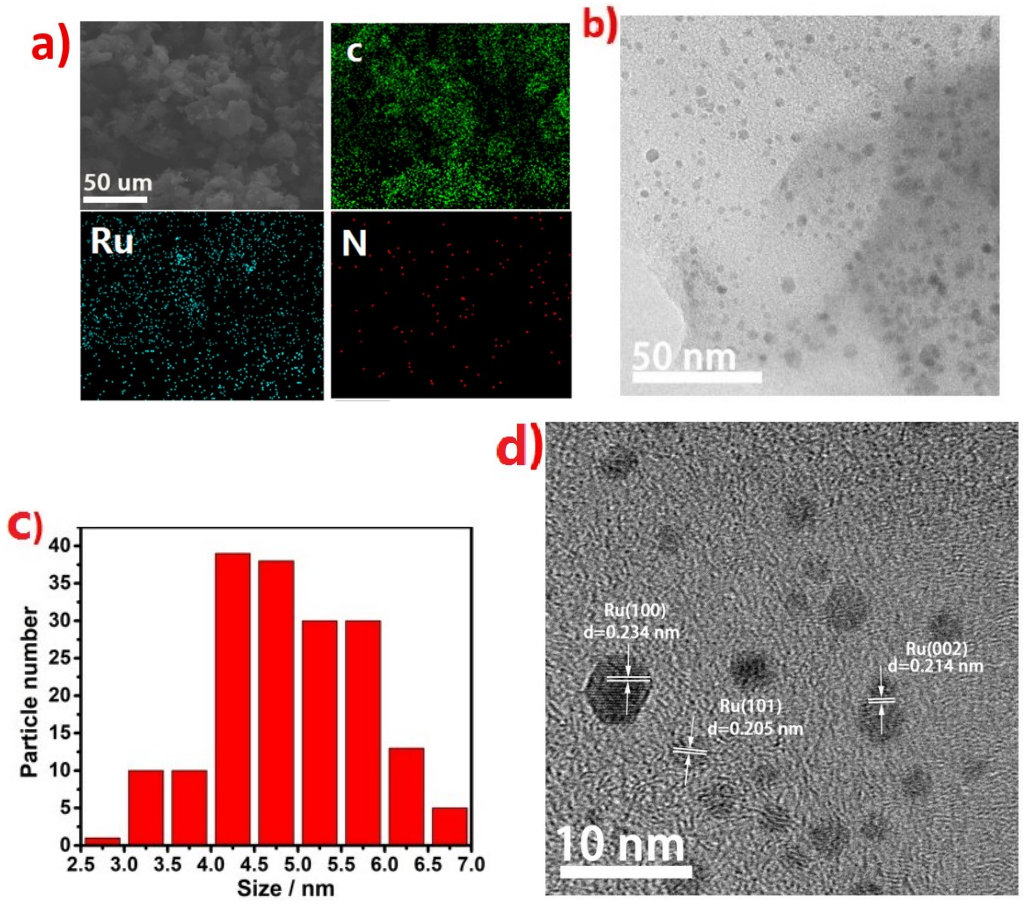


Figure S3. a) SEM and EDS-mapping images of Ru@NC(1:20). b) TEM images. c) The corresponding particle size distribution of the Ru nanoparticles. d) HRTEM image.

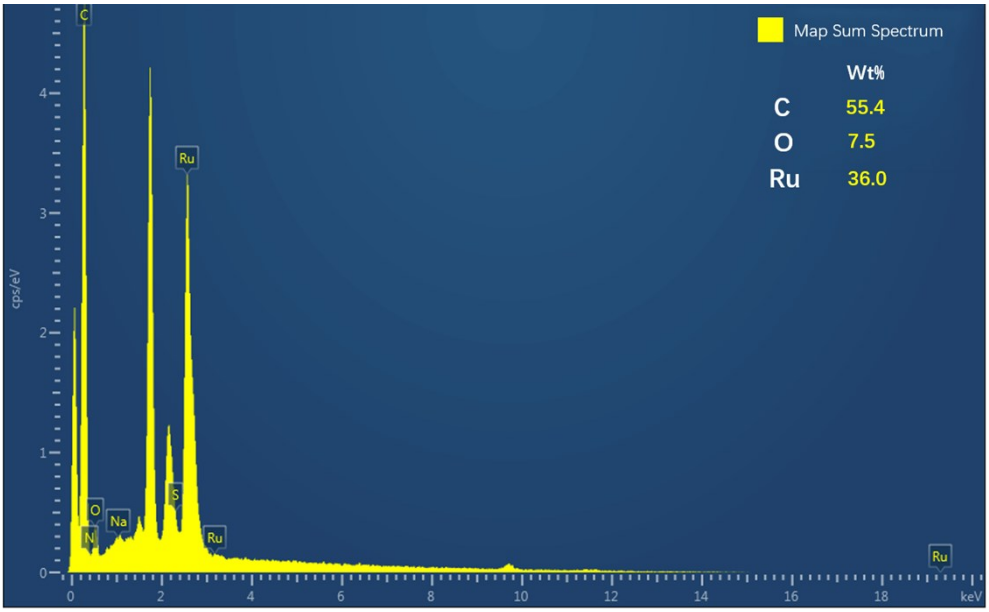


Fig. S4. EDS image of Ru@NC(1:5).

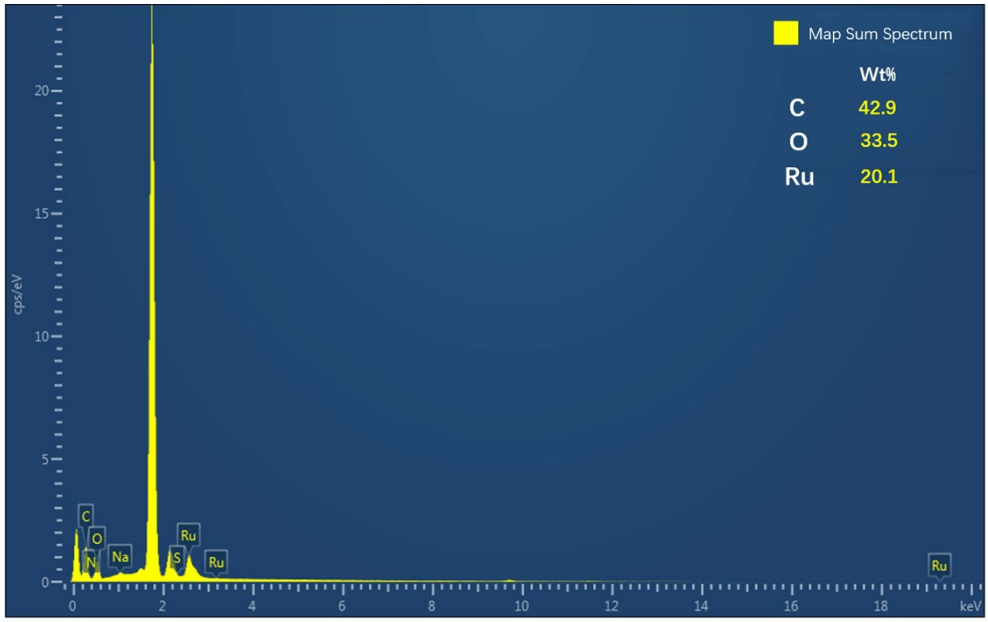


Fig. S5. EDS image of Ru@NC(1:2).

Fig. S6. EDS image of Ru@NC(1:10).

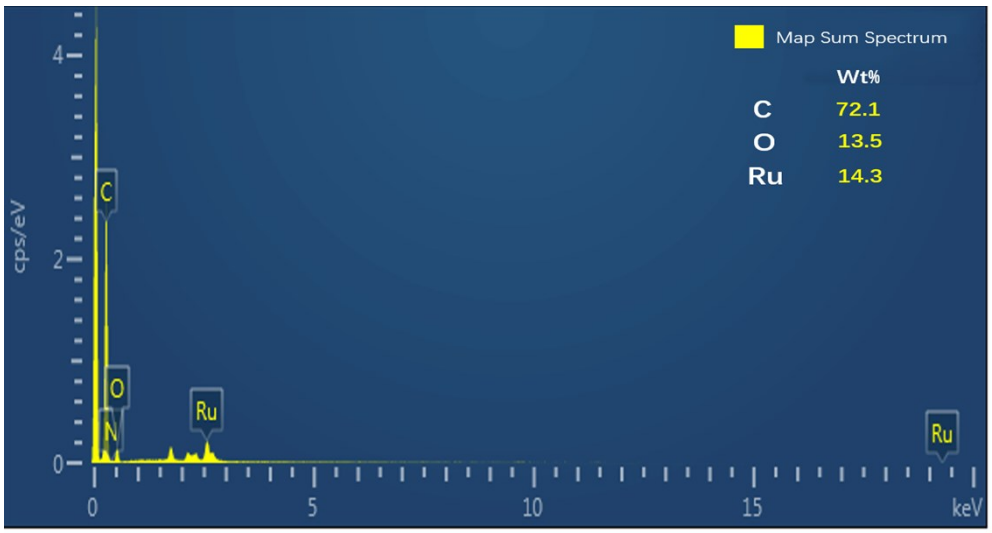
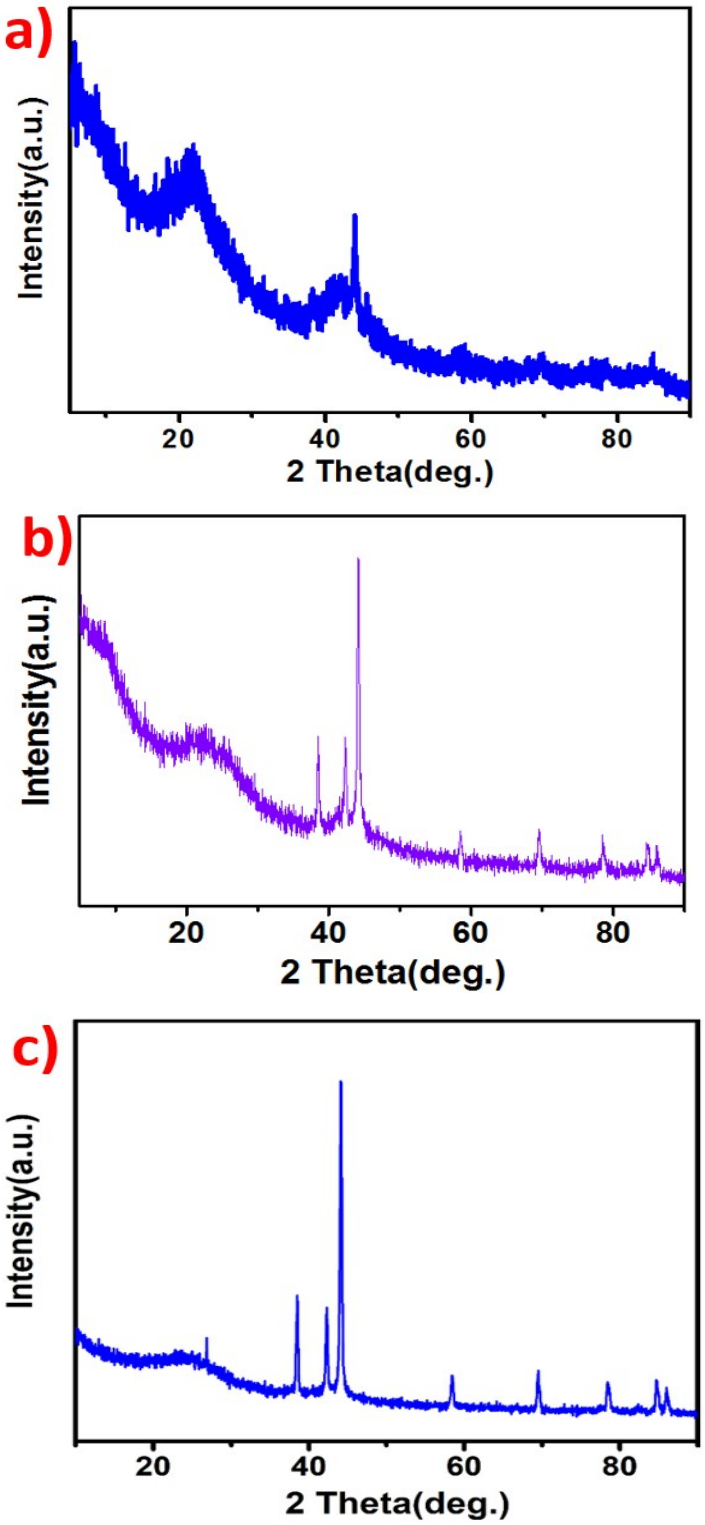


Fig. S7. EDS image of Ru@NC(1:20).



'**Figure S8.** XRD spectra of a) Ru@NC(1:2), b) Ru@NC(1:10) and c) Ru@NC(1:20).

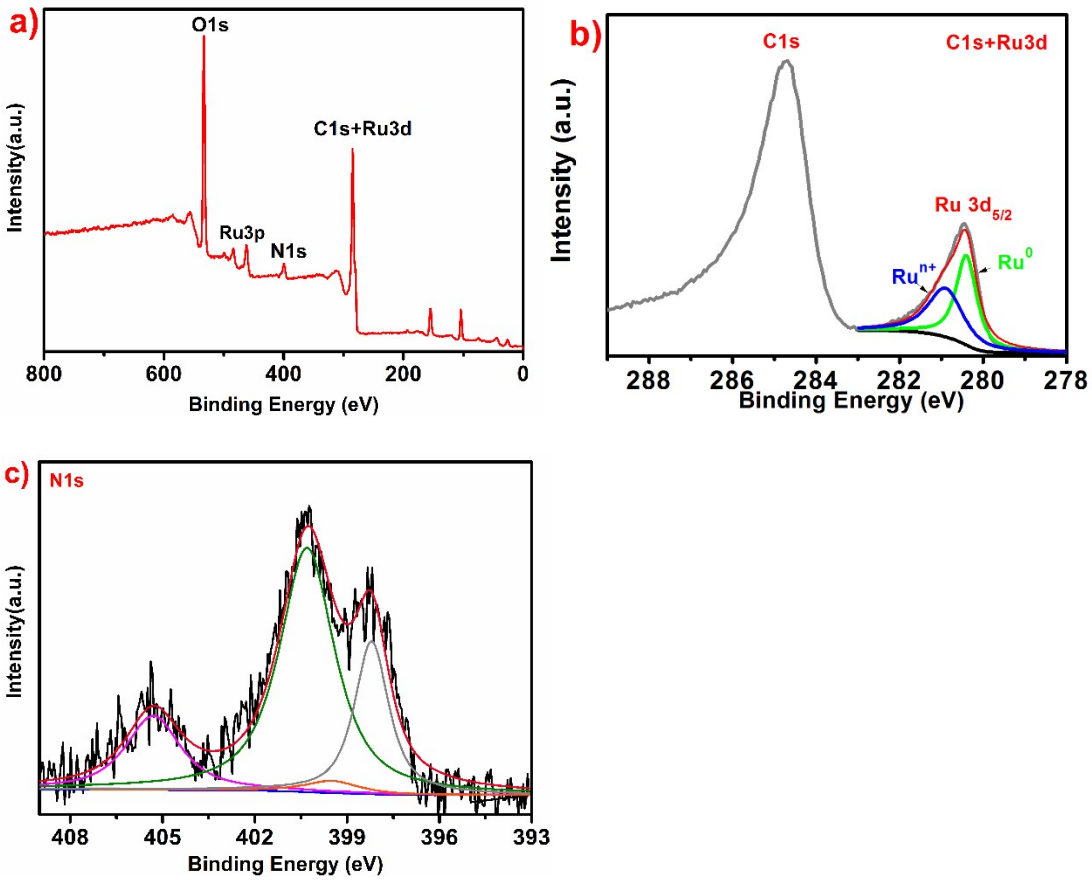


Figure S9. XPS analysis of the catalyst Ru@NC(1:2). a) XPS spectrum. b) High-resolution XPS spectrum of C 1s and Ru 3d. **c**) High-resolution XPS spectrum of N 1s.

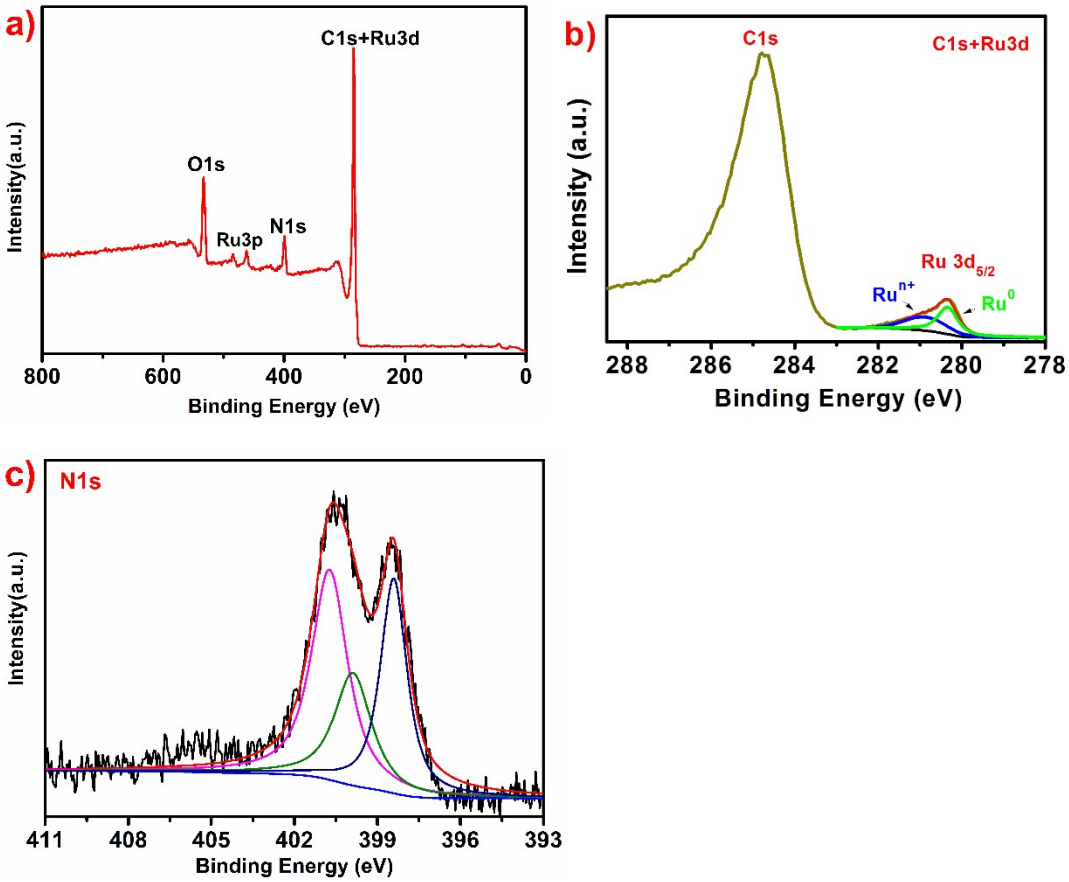


Figure S10. XPS analysis of the catalyst Ru@NC(1:10). a) XPS spectrum. b) High-resolution XPS spectrum of C 1s and Ru 3d. **c**) High-resolution XPS spectrum of N 1s.

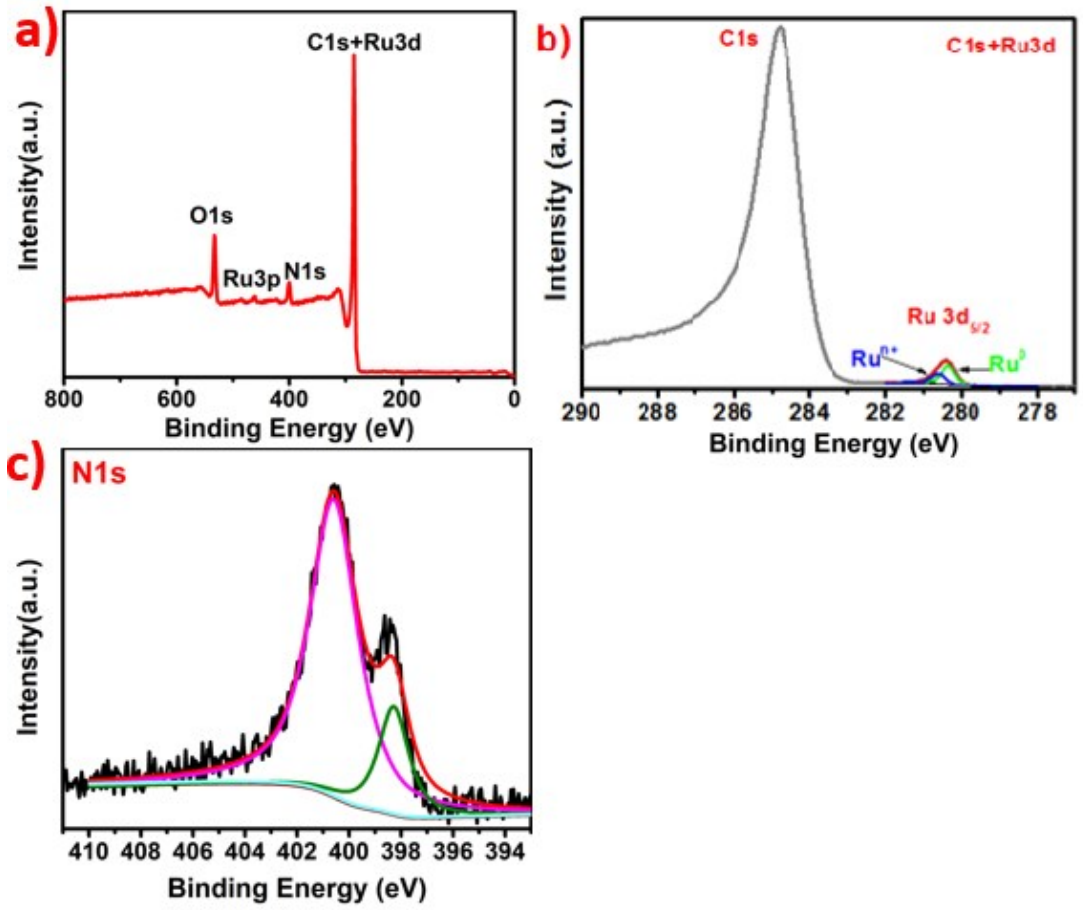


Figure S11. XPS analysis of the catalyst Ru@NC(1:20). a) XPS spectrum. b) High-resolution XPS spectrum of C 1s and Ru 3d. **c**) High-resolution XPS spectrum of N 1s, d) High-resolution XPS spectrum of Ru 3p.

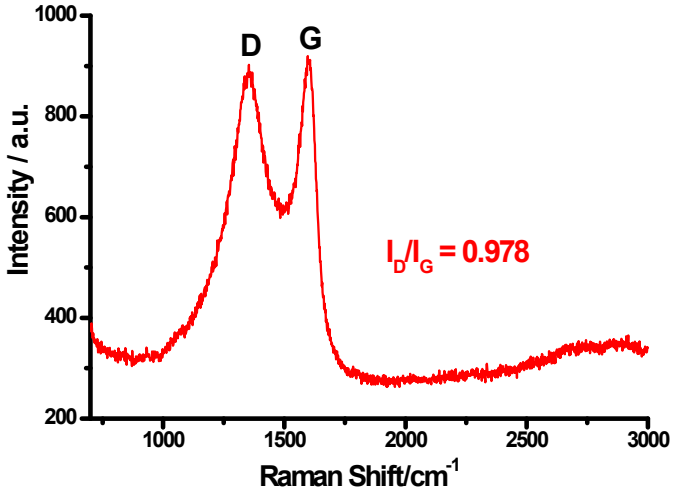


Fig. S12. Raman spectrum of Ru@NC(1:5).

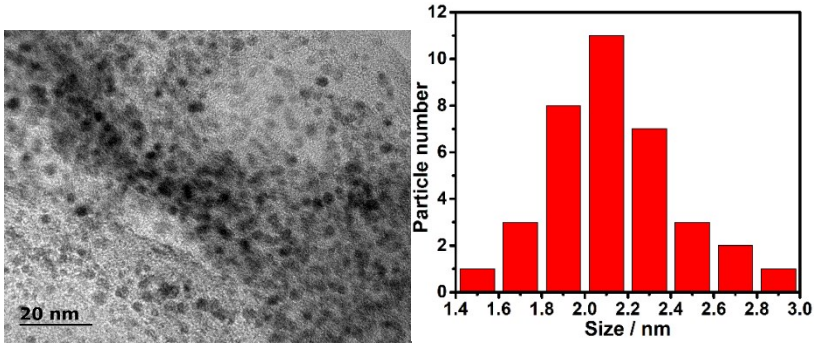


Figure S13. TEM image (left) and the corresponding particle size distribution of the Ru nanoparticles of Ru@NC(1:5) (right) after long-term stability test in 1.0 M KOH.

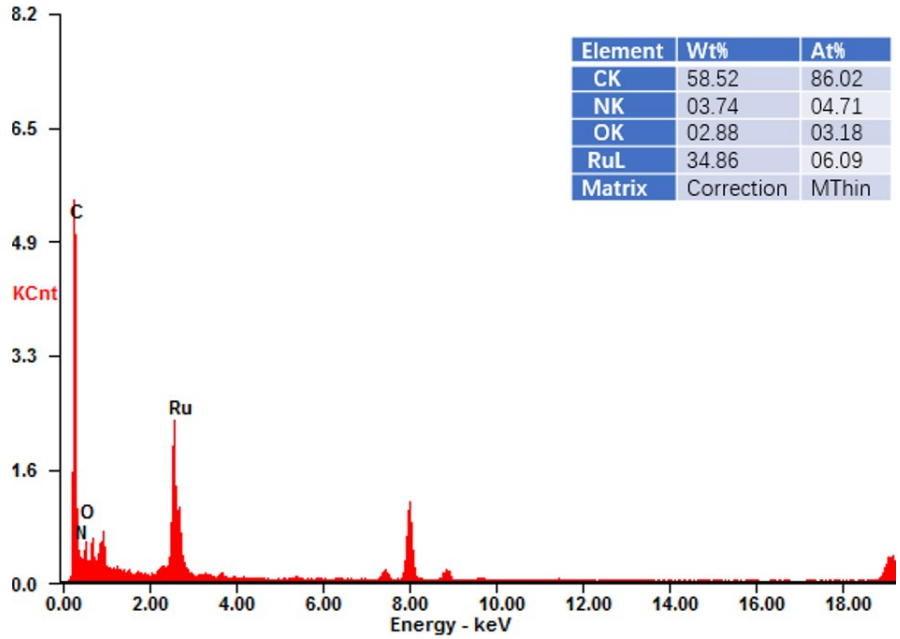


Figure S14. TEM-EDS image of Ru@NC(1:5) after long-term stability test in 1.0 M KOH.

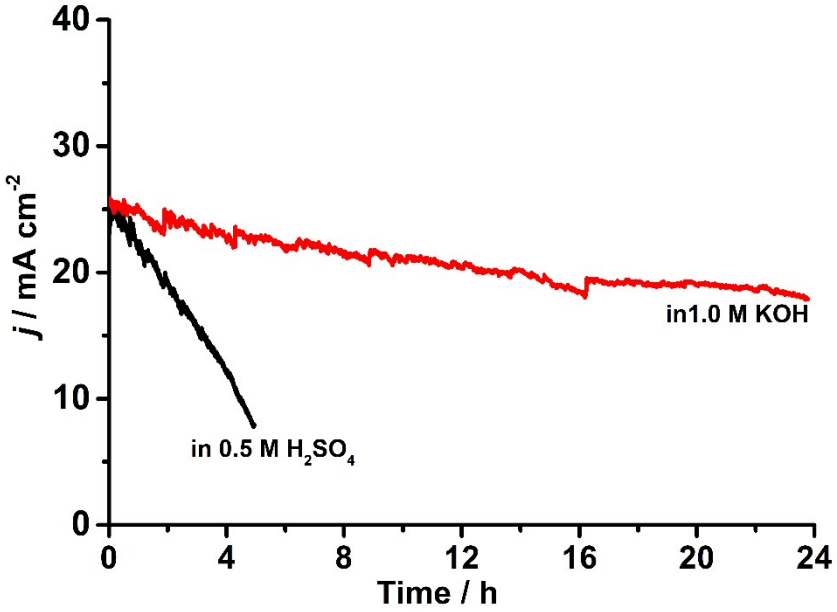


Figure S15. Time-dependent current density curves for Pt/C at -0.058 V vs RHE in 0.5 M H_2SO_4 and at -0.054 V vs RHE in 1.0 M KOH.

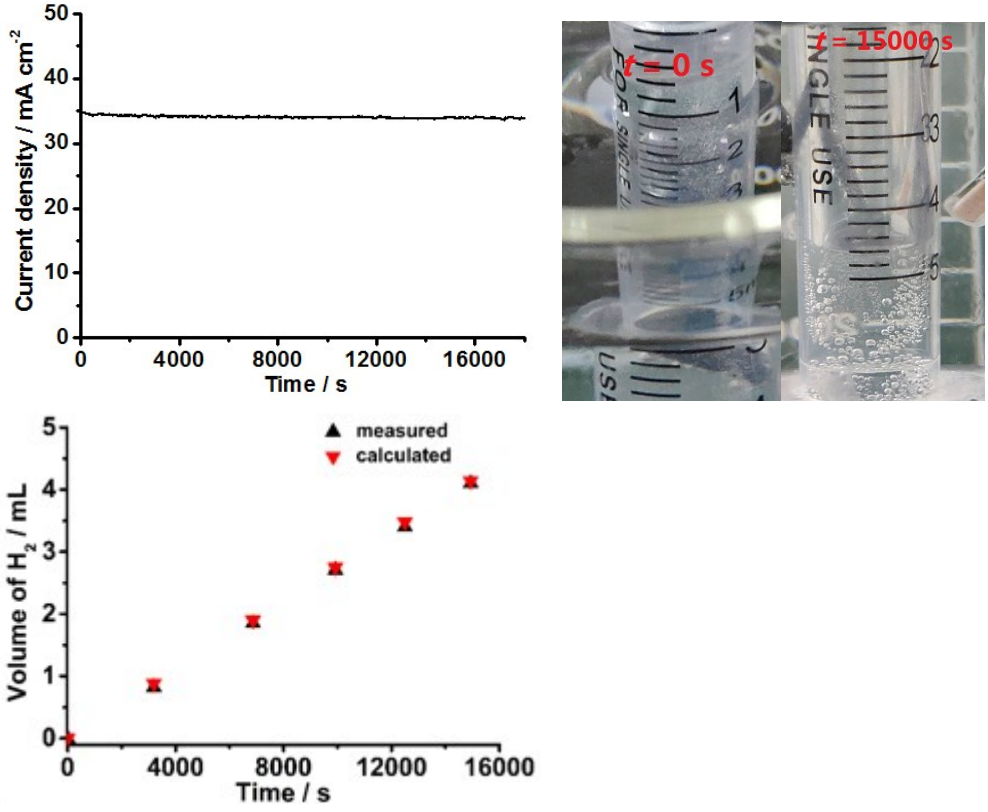


Figure S16. Time-dependent current density curve with the current density at 34 mA cm^{-2} in 1.0 M KOH solution (top). Amount of hydrogen theoretically calculated and experimentally measured versus time for Ru@NC(1:5) with the current density at 34 mA cm^{-2} in 1.0 M KOH solution (bottom).

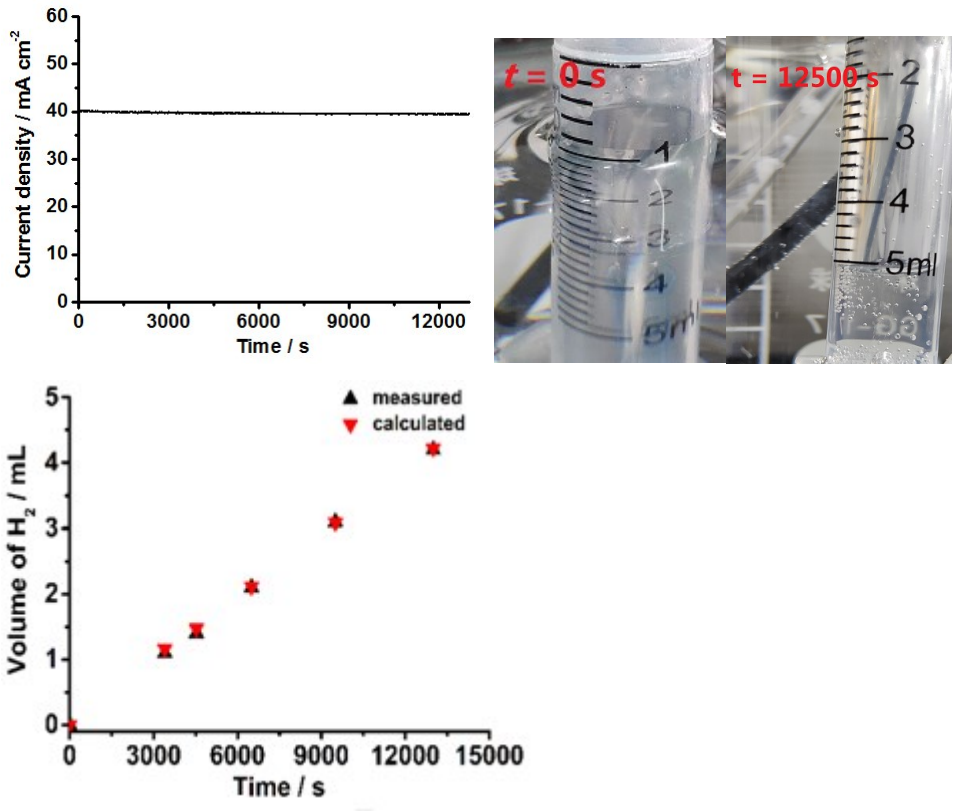


Figure S17. Time-dependent current density curve with the current density at 40 mA cm^{-2} in 0.5 M H_2SO_4 solution (top). Amount of hydrogen theoretically calculated and experimentally measured versus time for Ru@NC(1:5) with the current density at 40 mA cm^{-2} in 0.5 M H_2SO_4 solution (bottom).

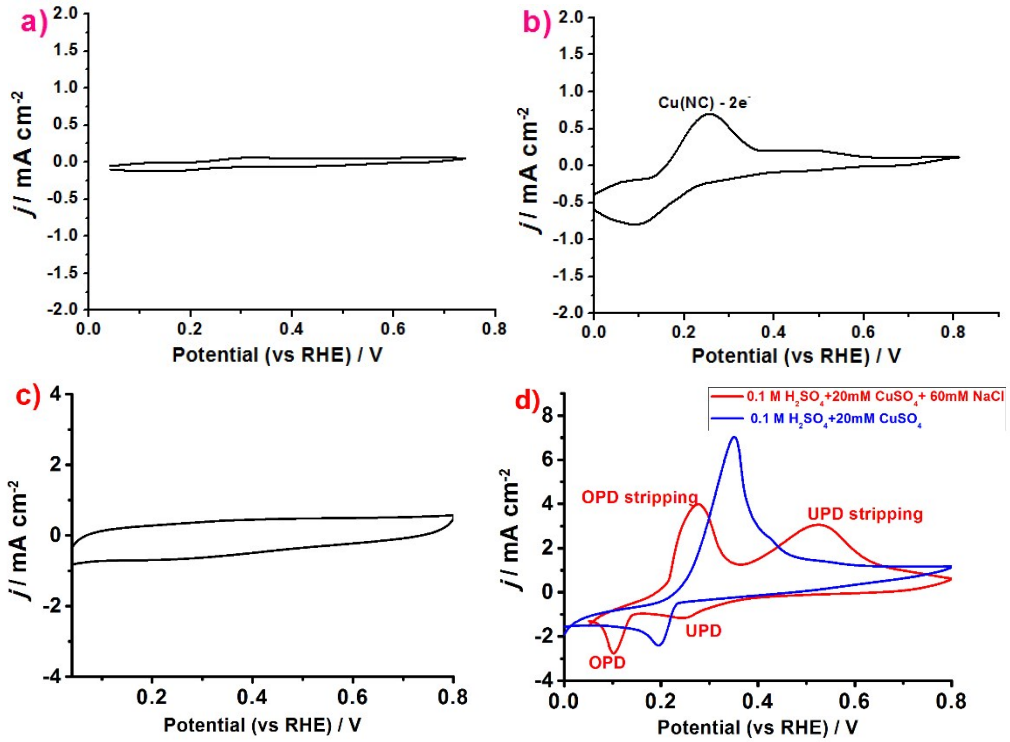


Figure S18. The current–voltage scan at 0.01 V/s in different solutions：a) NC in 0.1 M H_2SO_4; b) NC in 0.1 M H_2SO_4+20 mM $CuSO_4$; c) Ru@NC(1:5) in 0.1 M H_2SO_4; d) Ru@NC(1:5) in 0.1 M H_2SO_4+20 mM $CuSO_4$ (blue line) and in 0.1 M H_2SO_4+20 mM $CuSO_4$ +60 mM NaCl (red line).

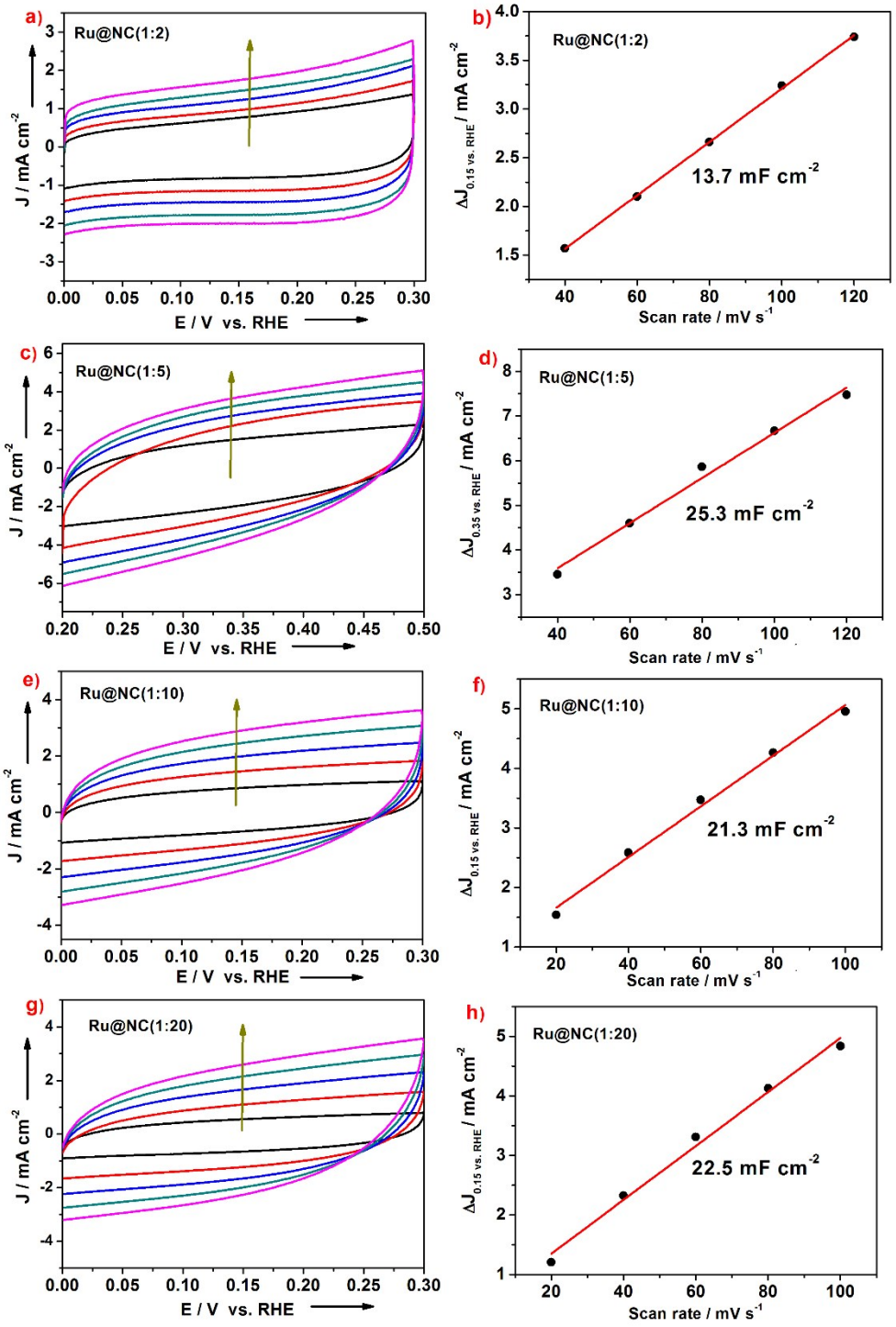


Figure S19. a, c, e, g) Cyclic voltammetry curves of Ru@NC(1:2, 1:5, 1:10, 1:20), respectively. The inset arrow indicates the increase of scan rate. b, d, f, h) Plot of current density variation (ΔJ=Ja-Jc) at the corresponding overpotentials vs. scan rate. Cdl was obtained from the linear plot, where the slope is equal to 2Cdl.