

Amorphous ruthenium nanosheets for efficient hydrazine-assisted water splitting

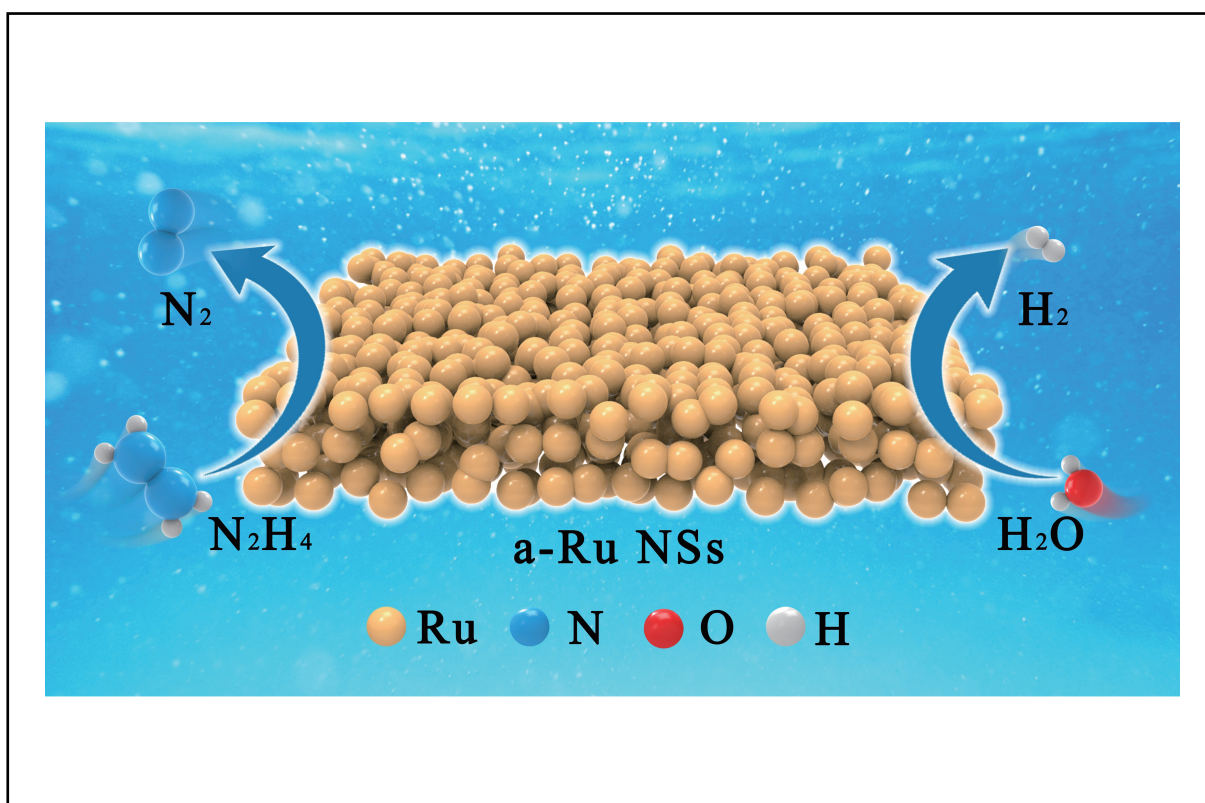
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Graphical abstract



Amorphization endows ruthenium nanosheets with enhanced activity in hydrazine-assisted water splitting.

Public summary

- The amorphous ruthenium nanosheets (a-Ru NSs) exhibit outstanding performance, with an ultralow overpotential of 17 mV for the hydrogen evolution reaction (HER) and a working potential of −76 mV for the hydrazine oxidation reaction (HzOR) to reach a current density of 10 mA·cm^{−2}.
- The a-Ru NSs show superior performance for overall hydrazine splitting (OH_zS) in a two-electrode catalytic system, which only require ultrasmall cell voltages of 28 mV and 207 mV to achieve current densities of 10 mA·cm^{−2} and 100 mA·cm^{−2}, respectively.
- The a-Ru NSs with abundant active sites can efficiently accelerate the cleavage of N–N bonds, which is beneficial for promoting the dehydrogenation of the *NH₂ intermediate and the final formation of nitrogen.

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Supporting Information

Abstract: The hydrazine oxidation reaction (HzOR) has garnered significant attention as a feasible approach to replace sluggish anodic reactions to save energy. Nevertheless, there are still difficulties in developing highly efficient catalysts for the HzOR. Herein, we report amorphous ruthenium nanosheets (a-Ru NSs) with a thickness of approximately 9.6 nm. As a superior bifunctional electrocatalyst, a-Ru NSs exhibited enhanced electrocatalytic performance toward both the HzOR and hydrogen evolution reaction (HER), outperforming benchmark Pt/C catalysts, where the a-Ru NSs achieved a working potential of merely -76 mV and a low overpotential of only 17 mV to attain a current density of $10 \text{ mA} \cdot \text{cm}^{-2}$ for the HzOR and HER, respectively. Furthermore, a-Ru NSs displayed a low cell voltage of 28 mV at $10 \text{ mA} \cdot \text{cm}^{-2}$ for overall hydrazine splitting in a two-electrode electrolyzer. In situ Raman spectra revealed that the a-Ru NSs can efficiently promote N–N bond cleavage, thereby producing more $^*\text{NH}_2$ and accelerating the progress of the reaction.

Keywords: amorphous structure; Ru nanosheets; hydrogen evolution reaction; hydrazine oxidation reaction; $^*\text{NH}_2$ adsorption

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1 Introduction

Electrochemical water splitting is considered an environmentally friendly and carbon-neutral method capable of producing high-purity hydrogen (H_2)^[1], but it is hindered by the sluggish kinetics of the oxygen evolution reaction (OER : $4\text{OH}^- = \text{O}_2 + 2\text{H}_2\text{O} + 4\text{e}^-$, 1.23 V vs. reversible hydrogen electrode (RHE))^[2,3]. In this respect, replacing the oxygen evolution reaction (OER) with the thermodynamically more favorable electrocatalytic oxidation of small molecules has emerged as an effective strategy to accelerate hydrogen production^[4–8], including urea^[9], methanol^[10], 5-hydroxymethylfurfural^[11,12], and hydrazine^[13]. Among these alternative reactions, the oxidation of hydrazine (HzOR: $\text{N}_2\text{H}_4 + 4\text{OH}^- \rightarrow \text{N}_2 + 4\text{H}_2\text{O} + 4\text{e}^-$, -0.33 V vs. RHE) stands out because of its low theoretical oxidation potential and significant reduction in energy consumption^[14–16], making it a focal point of research^[17,18]. Generally, there is a need to design two different types of catalysts for the hydrazine oxidation reaction (HzOR) and hydrogen evolution reaction (HER) in practical applications. Designing bifunctional electrocatalysts presents a strategic solution, simplifying the construction of catalytic electrodes and reducing costs^[19]. Unfortunately, achieving high-performance bifunctional electrocatalysts remains challenging, primarily because the active sites might differ for each catalytic process.

Noble metal nanomaterials hold significant potential for applications in the HER and HzOR. For example, partially exposed surface RuP_2 nanoparticles exhibit exceptional HER and HzOR activity in alkaline media^[20]. However, the scarcity and high cost of noble metals limit their widespread practical application. To construct more cost-effective noble metal

catalysts, there is a continuous effort to optimize their intrinsic activity, including alloying^[21], interface engineering^[22], and composition adjustment^[23]. Among these methods, amorphization endows electrocatalysts with abundant unsaturated coordination sites and defects, further enhancing their electrocatalytic performance^[24,25]. Therefore, it is rational to infer that designing amorphous noble metal catalysts represents a feasible approach to enhance the performance of the HER and HzOR but has rarely been achieved because of the strong metallic bonds between noble metal atoms^[26,27].

Here, we presented amorphous ruthenium nanosheets (a-Ru NSs) with a thickness of approximately 9.6 nm through the annealing of a mixture of acetylacetonate ruthenium and inorganic salts. The a-Ru NSs not only demonstrated excellent activity for HzOR ($-76 \text{ mV}@10 \text{ mA} \cdot \text{cm}^{-2}$), HER ($-17 \text{ mV}@10 \text{ mA} \cdot \text{cm}^{-2}$) and overall hydrazine splitting ($28 \text{ mV}@10 \text{ mA} \cdot \text{cm}^{-2}$) but also exhibited favorable durability across these electrocatalytic reactions. Furthermore, in situ Raman spectra demonstrated that the surface of the a-Ru NSs contained a relatively high concentration of $^*\text{NH}_2$ during the HzOR process, which is conducive to the reaction process of the HzOR.

2 Materials and methods

2.1 Chemicals

Ruthenium acetylacetonate ($\text{Ru}(\text{acac})_3$) was purchased from Adamas-Beta. Potassium bromide (KBr), absolute ethanol (99.5%), and hydrazine hydrate were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

Deionized (DI) water from a Milli-Q system (Millipore, Billerica, MA) was used in all our experiments. Nafion solution (5 wt%) was purchased from Aldrich. All chemicals were used as received without further purification.

2.2 Synthesis of a-Ru NSs

In the typical synthesis process of a-Ru NSs, 10 mg of Ru(acac)₃ and 30 mg of KBr were dissolved in a 10 mL ethanol–deionized water mixture (ethanol to water volume ratio of 3 : 1) under stirring. The solution was then dried in an oven at 70 °C to yield a mixed powder. The mixed powder was subsequently calcined at 280 °C for 90 min in an air atmosphere. After cooling to room temperature, the as-obtained products were washed several times with a DI water–ethanol mixture.

2.3 Synthesis of c-Ru NSs and RuO₂ NSs

In a typical synthesis of crystalline ruthenium nanosheets (c-Ru NSs), 15 mg of a-Ru NSs were annealed at 420 °C under an argon atmosphere for 90 min. The as-obtained products were washed several times with a DI water–ethanol mixture. The RuO₂ NSs were obtained via the same process as the a-Ru NSs, with the distinction of increasing the annealing temperature to 400 °C.

2.4 Characterization

Transmission electron microscopy images were captured using a Hitachi-7700 instrument at a working voltage of 100 kV. Additionally, high-resolution transmission electron microscopy and high-angle annular dark-field scanning transmission electron microscopy images of the samples were recorded on a JEOL JEM-2010 LaB₆ high-resolution transmission electron microscope at a working voltage of 200 kV. Scanning electron microscopy images were acquired with a Gemini SEM 500 microscope. Atomic force microscopy images were taken using a Bruker Dimension ICON instrument equipped with a Nano Scope V controller. Powder X-ray diffraction measurements of the samples were conducted on a Rigaku Miniflex-600 instrument. X-ray photoelectron spectroscopy data were collected via a PHI 5000 VersaProbe scanning X-ray microprobe (manufacturer: ULVAC-PHI, Inc.). In situ Raman spectra were generated via a laser Raman spectrometer (LabRAM HR Evolution) equipped with a 532 nm laser.

2.5 Electrochemical measurements

HER and HzOR tests were conducted indoors via a three-electrode setup and a CHI760E electrochemical workstation from Shanghai Chenhua. The working electrode employed was a rotating ring-disk electrode, with a platinum wire as the counter electrode and a mercury/mercuric oxide (Hg/HgO) electrode used for reference. Additionally, a two-electrode system was used for both overall water splitting (OWS) and overall hydrazine splitting (OHZS) assessments.

3 Results and discussion

3.1 Structural characterization

a-Ru NSs were synthesized through the annealing of

Ru(acac)₃ and KBr at 280 °C in air. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) images (Fig. 1a, b) demonstrated that a-Ru NSs have a two-dimensional flake-like morphology with a lateral size within the micrometer range. As indicated by the atomic force microscopy (AFM) image (Fig. 1c), the thickness of the a-Ru NSs was approximately 9.6 nm. Furthermore, high-angle annular dark-field scanning transmission electron microscopy and corresponding energy-dispersive X-ray spectroscopy (STEM-EDS) elemental mapping (Fig. 1d) revealed a uniform distribution of Ru, C, and O within the a-Ru NSs. As shown in Fig. 1e, the high-resolution transmission electron microscopy (HRTEM) image revealed that a-Ru NSs possessed a disordered atomic structure lacking regular lattice periodicity, which is in good agreement with the results of the fast Fourier transform (FFT) image with a halo pattern (Fig. 1f). Furthermore, the X-ray diffraction (XRD, Fig. 1g) pattern of a-Ru NSs lacked distinct diffraction peaks, further corroborating the amorphous structure. Together, the analysis of the HRTEM image, FFT image, and XRD pattern provided coherent evidence for the amorphous feature of a-Ru NSs.

The surface chemical state of a-Ru NSs was investigated via X-ray photoelectron spectroscopy (XPS) characterization. The XPS survey spectrum indicated the presence of C, O, and Ru in the amorphous nanosheets (Fig. S1 in Supporting information). As shown in the Ru 3d XPS spectrum of a-Ru NSs (Fig. 1h), the peaks located at 281.4 eV and 285.7 eV originated from Ru_{5/2} and Ru_{3/2} in the metallic state (Ru⁰). Additionally, the peaks at 282.5 eV and 286.9 eV correspond to the presence of Ru_{5/2} and Ru_{3/2} in the oxidative state (Ru⁴⁺), which may have been caused by metal Ru bonding with carbon or oxygen^[28]. According to the results of the Ru XPS spectra, the valence state of Ru present in a-Ru NSs was predominantly in the metallic state.

3.2 Electrocatalytic performance of a-Ru NSs for the HzOR and HER

The HzOR performance of a-Ru NSs was studied by employing a three-electrode system. For comparison, c-Ru NSs and RuO₂ NSs were synthesized (Figs. S2 and S3), and their electrocatalytic activities were measured. Initially, the HzOR activity of a-Ru NSs was tested under various concentrations of hydrazine (Fig. S4). The HzOR current of a-Ru NSs sharply increased with increasing concentrations of N₂H₄ and eventually remained constant at 0.3 M, demonstrating the potential of a-Ru NSs as efficient catalysts for the HzOR^[29]. Furthermore, linear sweep voltammetry (LSV) curves and Tafel slopes were employed to verify the HzOR activity of a-Ru NSs. As shown in Fig. 2a, the a-Ru NSs only required a working potential of −76 mV to achieve a current density of 10 mA·cm^{−2}, outperforming the c-Ru NSs (−51 mV), RuO₂ NSs (232 mV) and commercial Pt/C catalysts (130 mV). Moreover, the corresponding Tafel slope of a-Ru NSs was 31.58 mV·dec^{−1}, surpassing that of c-Ru NSs (50.65 mV·dec^{−1}), RuO₂ NSs (118.05 mV·dec^{−1}), and commercial Pt/C catalysts (34.05 mV·dec^{−1}), demonstrating that a-Ru NSs possess favorable kinetics for the HzOR (Fig. 2b). The combination of LSV and Tafel slope data indicates that a-Ru NSs exhibit outstanding intrinsic activity toward the HzOR. In ad-

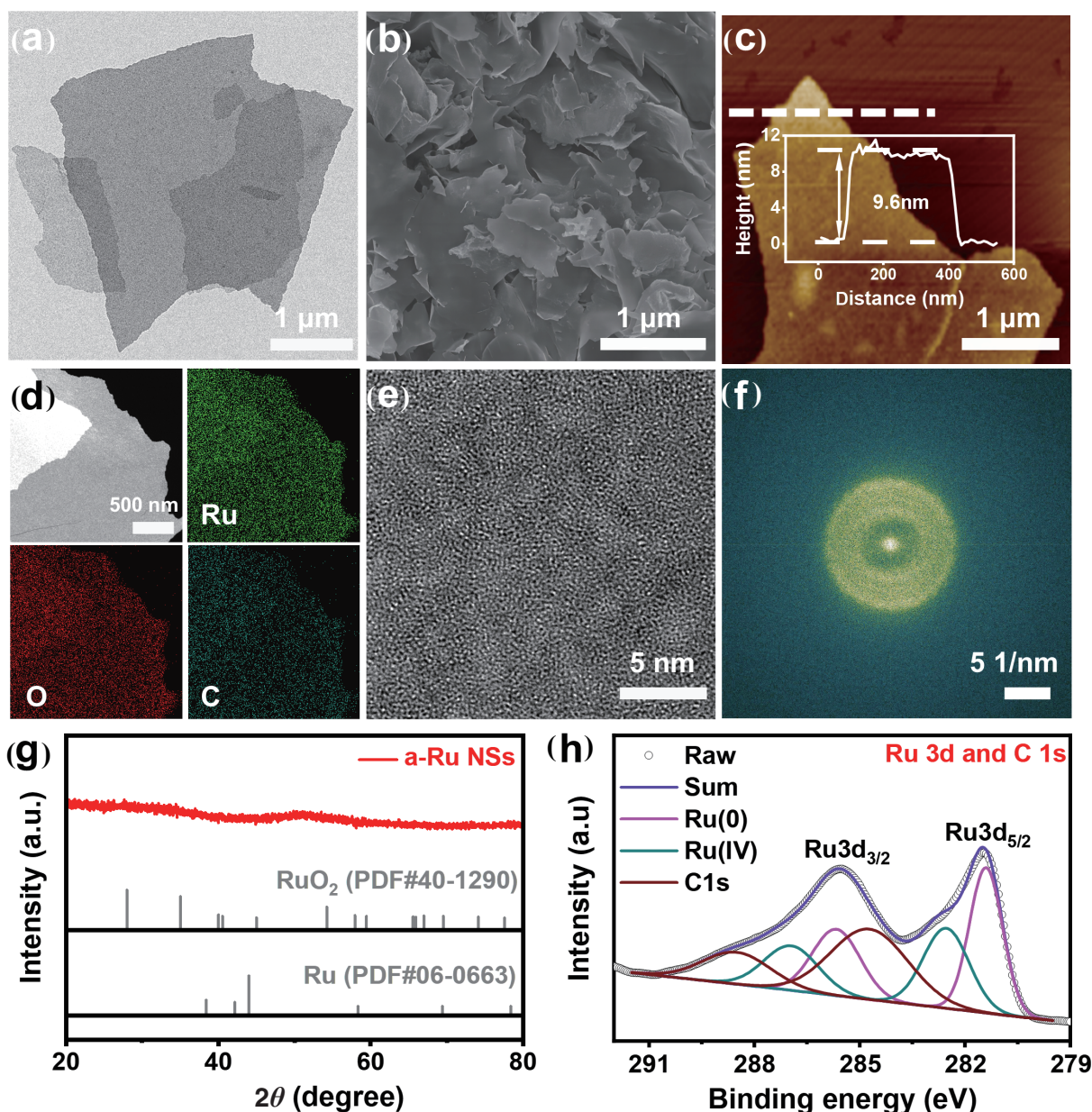


Fig. 1. (a) TEM image, (b) SEM image, (c) AFM image, (d) STEM-EDS elemental map, (e) HRTEM image, and (f) corresponding FFT image of a-Ru NSs. (g) XRD patterns and (h) Ru 3d XPS spectrum of a-Ru NSs.

dition to activity, durability is also a key factor in evaluating electrochemical performance. As a result, the electrochemical stability of a-Ru NSs was evaluated through chronoamperometry. As displayed in Fig. 2c, the current density of a-Ru NSs remained virtually stable over a 30-hour testing period, confirming the exceptional electrocatalytic stability of a-Ru NSs.

Furthermore, the HER performance of a-Ru NSs was also investigated through LSV, Tafel slope, and chronoamperometry. As shown in Fig. 2e and Fig. 2d, the a-Ru NSs exhibited the lowest overpotential (17 mV@10 mA·cm⁻²) and the smallest Tafel slope (49.26 mV·dec⁻¹) among all the tested catalysts, suggesting that the a-Ru NSs possessed remarkable activity for the HER. In addition, chronoamperometry

(Fig. 2f) and long-term cycling stability tests were performed (Fig. S5). The performance of a-Ru NSs remained unchanged at 10 mA·cm⁻² for at least 20 h and exhibited only slight attenuation after 5000 cycles, revealing outstanding durability. Furthermore, the structural stability of a-Ru NSs was tested via TEM and XRD. As displayed in Fig. S6, there were nearly no changes in a-Ru NSs after the stability test, confirming the structural stability of a-Ru NSs. In addition, the turnover frequency (TOF) values of the tested catalysts were calculated at different applied potentials (Fig. 2g and Fig. S7). a-Ru NSs exhibited a high TOF value of 2.72 s⁻¹ at an overpotential of 100 mV, which was approximately 1.73 and 2.09 times greater than those of c-Ru NSs (1.57 s⁻¹) and commercial Pt/C catalysts (1.3 s⁻¹), respectively. Furthermore, the

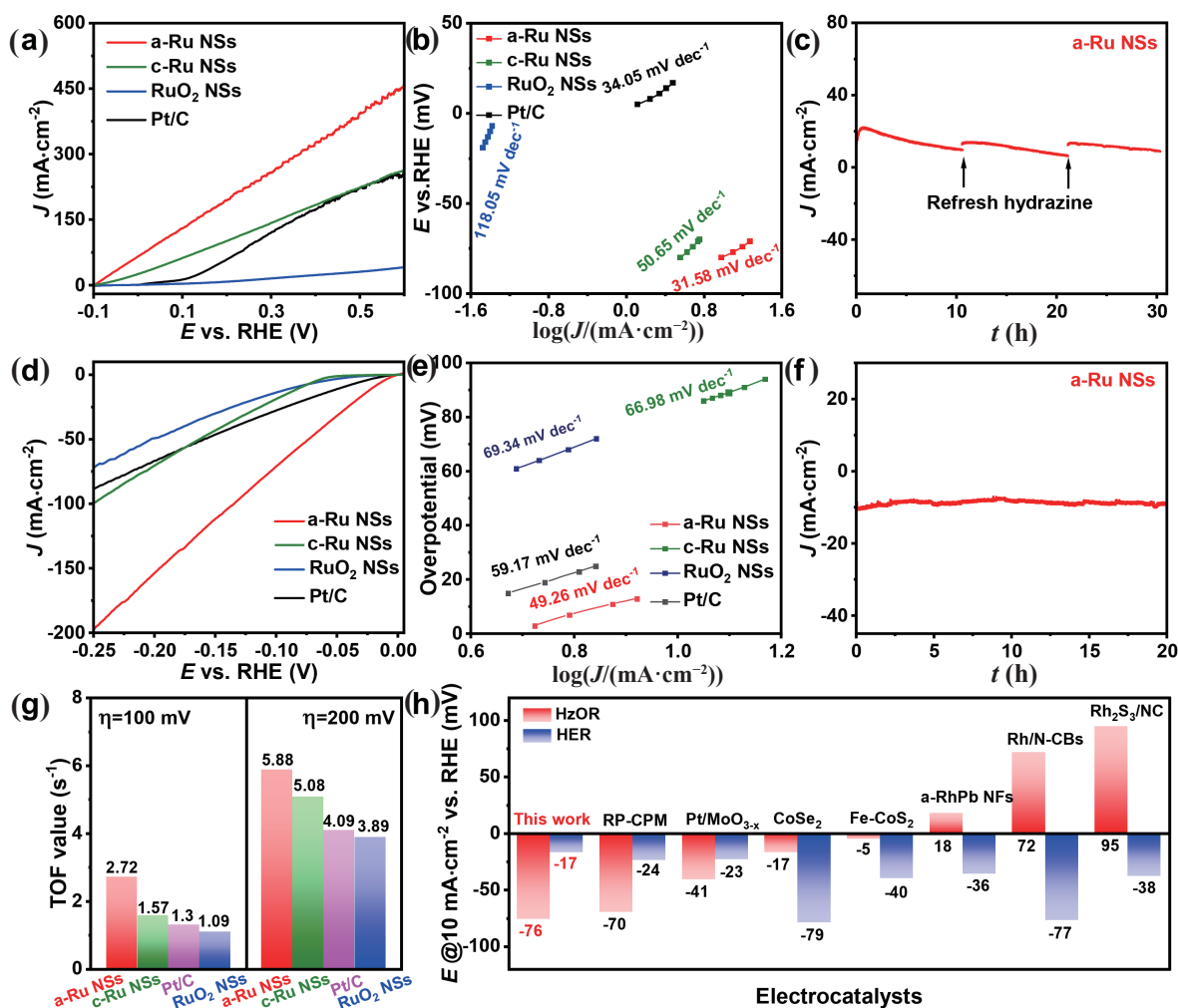


Fig. 2. (a) LSV curves and (b) corresponding Tafel plots of a-Ru NSs, c-Ru NSs, RuO₂ NSs, and commercial Pt/C catalysts toward the HzOR. (c) Chronoamperometric curve of a-Ru NSs for the HzOR. (d) LSV curves and (e) corresponding Tafel plots of a-Ru NSs, c-Ru NSs, RuO₂ NSs, and commercial Pt/C catalysts for the HER. (f) Chronoamperometric curve of a-Ru NSs for the HER. (g) TOFs of catalysts at different applied potentials. (h) Comparison of the working potentials of a-Ru NSs and various other electrocatalysts at a current density of 10 mA·cm⁻² for both the HzOR and HER.

Nyquist plots revealed that a-Ru NSs had the lowest charge transfer resistance among the tested catalysts (Fig. S8). The combination of the results of LSV, Tafel slope and TOF confirmed that a-Ru NSs possessed substantially enhanced intrinsic activity for the HER. Among all the tested electrocatalysts, a-Ru NSs exhibited the highest mass activity for the HzOR (3100.7 A per gram of Ru) and HER (1383.5 A per gram of Ru), suggesting a notable advantage for the HzOR and HER (Fig. S9). Importantly, a-Ru NSs delivered the optimal working potential at a current density of 10 mA·cm⁻² among the recently reported bifunctional electrocatalysts for both the HzOR and HER (Fig. 2h and Table S1 in Supporting information).

To elucidate the superior HzOR performance of a-Ru NSs, electrochemically active surface area (ECSA) and electrochemical impedance spectroscopy (EIS) analyses were conducted. The ECSA of the electrocatalysts was obtained via the double-layer capacitance (C_{dl}) method. As revealed in Fig. 3a and Fig. S10, the C_{dl} of a-Ru NSs was 51.33 mF·cm⁻², significantly greater than those of c-Ru NSs (7.98 mF·cm⁻²) and

commercial Pt/C catalysts (2.38 mF·cm⁻²). Accordingly, the ECSA of a-Ru NSs was 6.4 times greater than that of c-Ru NSs (Fig. 3b), demonstrating that a-Ru NSs exposed more active sites. Moreover, the polarization curves of a-Ru NSs at different scan rates were investigated (Fig. S11). At varying scan rates, the polarization curves showed no obvious change, confirming the excellent charge transfer of a-Ru NSs for the HzOR. This result coincided well with the results of the Nyquist plots (Fig. 3c), in which a-Ru NSs had the lowest charge-transfer resistance among the tested catalysts, further confirming the faster charge-transfer process of a-Ru NSs for the HzOR.

To further clarify the catalytic reaction mechanism of a-Ru NSs toward the HzOR, in situ Raman spectra were obtained. As shown in Fig. 3d and Fig. S12, a-Ru NSs presented two distinctive vibrational peaks at 1351 and 1603 cm⁻¹ at a minimal working potential of -0.05 V during the HzOR, which corresponded to the adsorbed *NH₂ intermediates. Indeed, as the tested potential increased, the vibration peak of the adsorbed *NH₂ intermediates became more pronounced,

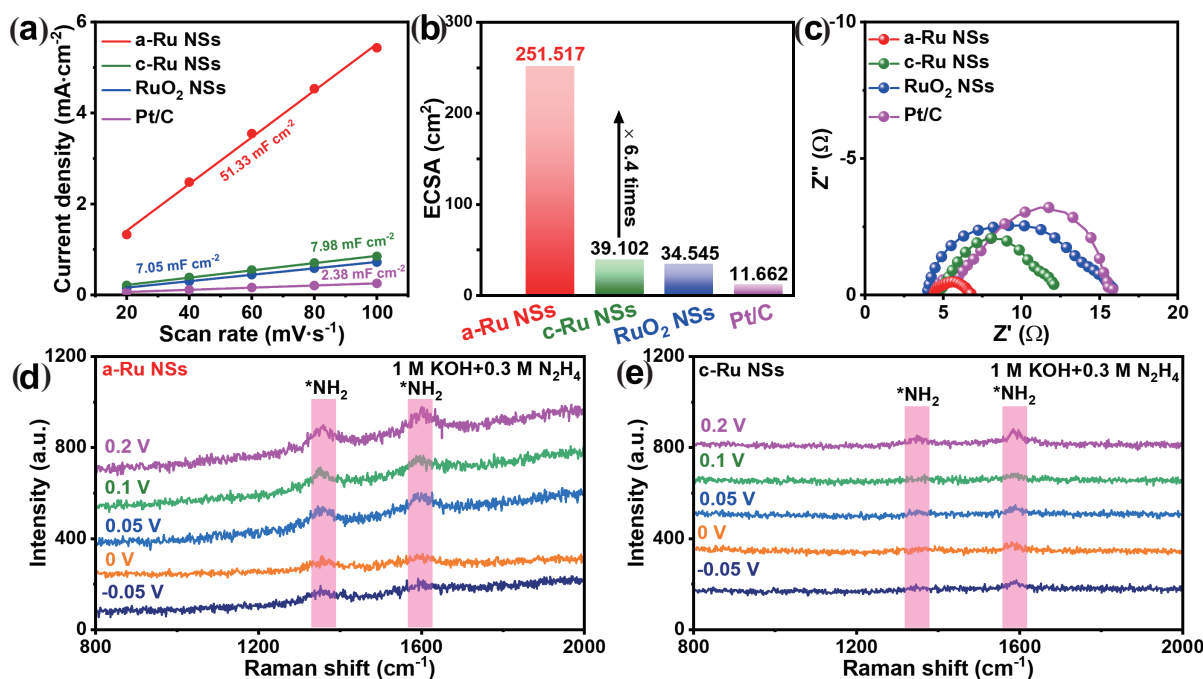


Fig. 3. (a) C_{dl} values, (b) ECSA values, and (c) Nyquist plots of a-Ru NSs, c-Ru NSs, RuO₂ NSs, and Pt/C, respectively. In situ Raman spectra of (d) a-Ru NSs and (e) c-Ru NSs toward the HzOR.

indicating the accumulation of the *NH₂ intermediate. Moreover, as shown in Fig. 3e, the variation trend of the absorption *NH₂ peak observed for the c-Ru NSs was almost identical to that of the a-Ru NSs. Remarkably, the intensity of the *NH₂ adsorbed peaks on the a-Ru NSs was significantly greater than that on the c-Ru NSs at the same tested potential (Fig. S13), indicating greater coverage of *NH₂ on the surface of the a-Ru NSs. According to the analysis of in situ Raman spectra, the HzOR pathways of a-Ru NSs followed the N–N bond cleavage process, in which the N₂H₄ molecules adsorbed on the a-Ru NSs surface first underwent intramolecular N–N bond cleavage and then each *NH₂ intermediate gradually dehydrogenated to form nitrogen^[30]. Combining the data of the ECSA, EIS, and in situ Raman spectra, a reasonable explanation for the superior HzOR performance is that a-Ru NSs with high ECSAs and abundant active sites could efficiently accelerate the cleavage of N–N bonds, which was beneficial for promoting the dehydrogenation of the *NH₂ intermediate and the final formation of nitrogen.

3.3 Electrocatalytic performance of a-Ru NSs toward OHzS in a two-electrode system

Owing to the impressive performance of a-Ru NSs for both the HzOR and HER, we employed a-Ru NSs as bifunctional electrodes in a two-electrode catalytic system to investigate the OHzS performance (Fig. S14). As exhibited in the LSV curves (Fig. 4a), the OHzS performance of a-Ru NSs was superior to that of commercial Pt/C catalysts. Specifically, a-Ru NSs achieved ultralow voltages of 28 mV and 207 mV at current densities of 10 and 100 mA·cm⁻², significantly greater than the 144 mV and 529 mV required by commercial Pt/C catalysts (Fig. 4b), which were also better than those of recently reported OHzS catalysts (Fig. 4c, Table S2).

Additionally, a comparison of the polarization curves of OHzS and overall water splitting (OWS) was conducted (Fig. 4d), revealing a notable reduction in electric power for H₂ production through a hydrazine-assisted water splitting system. For a comprehensive comparison, as displayed in Fig. 4e, the overpotentials for OHzS of a-Ru NSs only required overpotentials of 358, 437, and 537 mV to reach 10, 50, and 100 mA·cm⁻², respectively, which were much smaller than those for OWS at the above current densities (1237, 1494, and 1646 mV, respectively). Additionally, the OHzS of a-Ru NSs also exhibited remarkable durability (Fig. 4f), and the current density of OHzS remained almost unchanged throughout the 30-hour stability test, indicating that a-Ru NSs have significant potential in OHzS applications.

4 Conclusions

In summary, we successfully synthesized a-Ru NSs with a thickness of approximately 9.6 nm, which exhibited excellent bifunctional electrocatalytic performance for the HER and HzOR. Impressively, the a-Ru NSs achieved an overpotential of 17 mV for the HER and a working potential of -76 mV for the HzOR to reach a current density of 10 mA·cm⁻². Additionally, in situ Raman spectra revealed that a-Ru NSs were capable of effectively promoting N–N bond cleavage and enhancing the adsorption of *NH₂ intermediates in comparison to c-Ru NSs. Furthermore, for OHzS in a two-electrode catalytic system, a-Ru NSs also exhibit superior performance, requiring ultrasmall cell voltages of 28 mV and 207 mV to achieve current densities of 10 mA·cm⁻² and 100 mA·cm⁻², respectively. This work provides novel insight into the exploration and design of highly efficient bifunctional electrocatalysts for hydrazine-assisted water splitting.

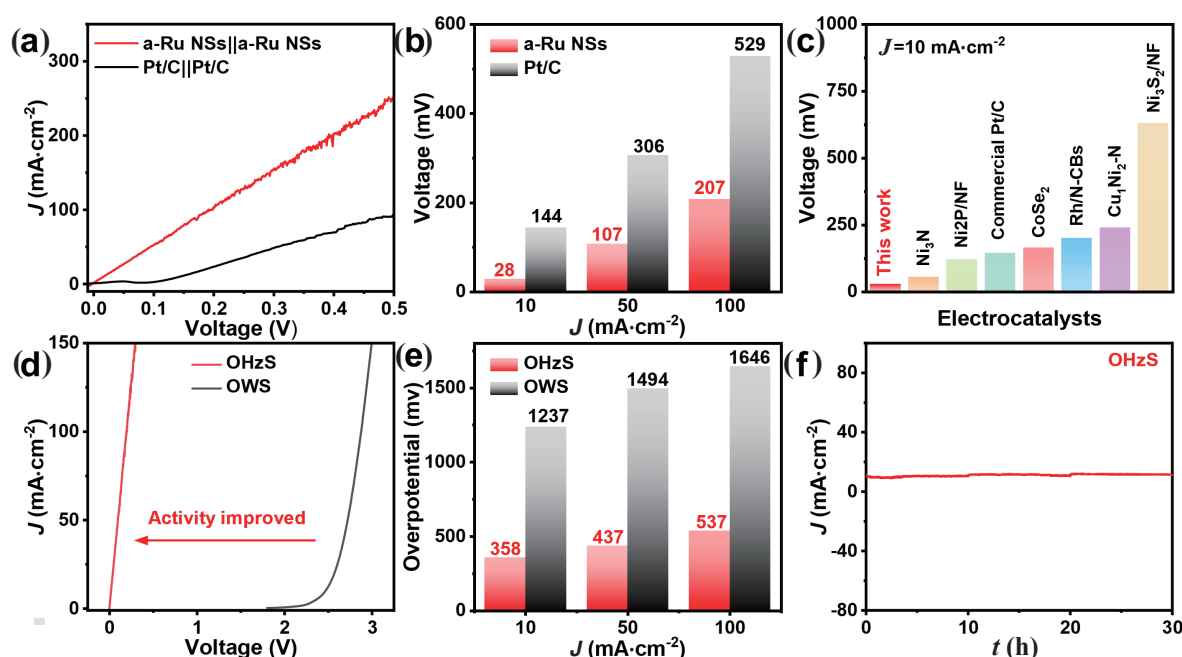


Fig. 4. (a) Comparison of the LSV curves of the a-Ru NSs||a-Ru NSs and Pt/C||Pt/C commercial catalyst couples and (b) the corresponding cell voltages at 10 mA·cm⁻², 50 mA·cm⁻², and 100 mA·cm⁻². (c) Comparison of the cell voltages for a-Ru NSs||a-Ru NSs couples and recently reported OHzS catalysts at a current density of 10 mA·cm⁻². (d) Polarization curves and (e) compared overpotentials of a-Ru NSs||a-Ru NSs couples for OWS and OHzS. (f) Chronoamperometric curve of a-Ru NSs||a-Ru NSs coupled with OHzS.

Supporting information

The supporting information for this article can be found online at <https://doi.org/10.52396/JUSTC-2024-0080>. The supporting information includes fourteen supporting figures (Figs. S1–S14) and two supporting tables (Tables S1 and S2).

Acknowledgements

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Conflict of interest

The authors declare that they have no conflict of interest.

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References

- [1] Dinh C T, Jain A, de Arquer F P G, et al. Multi-site electrocatalysts for hydrogen evolution in neutral media by destabilization of water molecules. *Nature Energy*, **2018**, 4: 107–114.
- [2] Huang W, Su C, Zhu C, et al. Isolated electron trap-induced charge accumulation for efficient photocatalytic hydrogen production. *Angewandte Chemie International Edition*, **2023**, 62 (25): e202304634.
- [3] Zhang H, An P, Zhou W, et al. Dynamic traction of lattice-confined platinum atoms into mesoporous carbon matrix for hydrogen evolution reaction. *Science Advances*, **2018**, 4 (1): eaao6657.
- [4] Chen F Y, Wu Z Y, Adler Z, et al. Stability challenges of electrocatalytic oxygen evolution reaction: From mechanistic understanding to reactor design. *Joule*, **2021**, 5 (7): 1704–1731.
- [5] Liu X, Meng J S, Zhu J X, et al. Comprehensive understandings into complete reconstruction of precatalysts: synthesis, applications, and characterizations. *Advanced Materials*, **2021**, 33 (32): 2007344.
- [6] Wu Y Y, Li Y, Yuan M K, et al. Operando capturing of surface self-reconstruction of Ni₃S₂/FeNi₂S₄ hybrid nanosheet array for overall water splitting. *Chemical Engineering Journal*, **2022**, 427: 131944.
- [7] Xie X H, Du L, Yon L T, et al. Oxygen evolution reaction in alkaline environment: material challenges and solutions. *Advanced Functional Materials*, **2022**, 32 (21): 2110036.
- [8] Wang Z Y, Xu L, Huang F Z, et al. Copper–nickel nitride nanosheets as efficient bifunctional catalysts for hydrazine-assisted electrolytic hydrogen production. *Advanced Energy Materials*, **2019**, 9 (21): 1900390.
- [9] Geng S K, Zheng Y, Li S Q, et al. Nickel ferrocyanide as a high-

- p>performance urea oxidation electrocatalyst.
- Nature Energy*
- ,
- 2021**
- ,
- 6*
- : 904–912.
- [10] Li J S, Xing C C, Zhang Y, et al. Nickel iron diselenide for highly efficient and selective electrocatalytic conversion of methanol to formate. *Small*, **2021**, *17* (6): 2006623.
- [11] Jiang N, You B, Boonstra R, et al. Integrating electrocatalytic 5-hydroxymethylfurfural oxidation and hydrogen production via Co–P-derived electrocatalysts. *ACS Energy Letters*, **2016**, *1* (2): 386–390.
- [12] Kang M J, Park H, Jegal J, et al. Electrocatalysis of 5-hydroxymethylfurfural at cobalt based spinel catalysts with filamentous nanoarchitecture in alkaline media. *Applied Catalysis B: Environmental*, **2019**, *242*: 85–91.
- [13] Li Y P, Wang W T, Cheng M Y, et al. Arming Ru with oxygen-vacancy-enriched RuO₂ sub-nanometer skin activates superior bifunctionality for pH-universal overall water splitting. *Advanced Materials*, **2023**, *35* (24): 2206351.
- [14] Jose V, Do V H, Prabhu P, et al. Activating amorphous Ru metallenes through Co integration for enhanced water electrolysis. *Advanced Energy Materials*, **2023**, *13* (28): 2301119.
- [15] Wang J S, Guan X Y, Li H B, et al. Robust Ru–N metal-support interaction to promote self-powered H₂ production assisted by hydrazine oxidation. *Nano Energy*, **2022**, *100*: 107467.
- [16] Wang X H, Yuan R, Yin S B, et al. Ultrathin Co_{0.5}NiS nanosheets for hydrazine oxidation assisted nitrite reduction. *Advanced Functional Materials*, **2024**, *34* (8): 2310288.
- [17] Hu Y M, Chao T T, Li Y P, et al. Cooperative Ni(Co)–Ru–P sites activate dehydrogenation for hydrazine oxidation assisting self-powered H₂ production. *Angewandte Chemie International Edition*, **2023**, *62* (35): e202308800.
- [18] Li J C, Li Y, Wang J A, et al. Elucidating the critical role of ruthenium single atom sites in water dissociation and dehydrogenation behaviors for robust hydrazine oxidation-boosted alkaline hydrogen evolution. *Advanced Functional Materials*, **2022**, *32* (16): 2109439.
- [19] Liu Y, Zhang J H, Li Y P, et al. Realizing the synergy of interface engineering and chemical substitution for Ni₃N enables its bifunctionality toward hydrazine oxidation assisted energy-saving hydrogen production. *Advanced Functional Materials*, **2021**, *31* (35): 2103673.
- [20] Li Y P, Zhang J H, Liu Y, et al. Partially exposed RuP₂ surface in hybrid structure endows its bifunctionality for hydrazine oxidation and hydrogen evolution catalysis. *Science Advances*, **2020**, *6* (44): eabb4197.
- [21] Tian W J, Zhang X, Wang Z Q, et al. Amorphization activated RhPb nanoflowers for energy-saving hydrogen production by hydrazine-assisted water electrolysis. *Chemical Engineering Journal*, **2022**, *440*: 135848.
- [22] Liu Z Y, Li Y Y, Guo H R, et al. Interface engineering of amorphous/crystalline heterojunctions with synergistic Ru doping for efficient hydrazine oxidation assisted overall water splitting. *Journal of Materials Chemistry A*, **2023**, *11* (45): 24667–24677.
- [23] Luo F, Zhang Q, Yu X X, et al. Palladium phosphide as a stable and efficient electrocatalyst for overall water splitting. *Angewandte Chemie International Edition*, **2018**, *57* (45): 14862–14867.
- [24] Sun R B, Gao J Y, Wu G, et al. Amorphous metal oxide nanosheets featuring reversible structure transformations as sodium-ion battery anodes. *Cell Reports Physical Science*, **2020**, *1* (7): 100118.
- [25] Sun R B, Wei C, Huang Z X, et al. Cu₂₊₁O/CuO_x heterostructures promote the electrosynthesis of C₂₊ products from CO₂. *Nano Research*, **2023**, *16*: 4698–4705.
- [26] Wu G, Zheng X S, Cui P X, et al. A general synthesis approach for amorphous noble metal nanosheets. *Nature Communications*, **2019**, *10*: 4855.
- [27] Yu G, Li R L, Zhang Y D, et al. Constructing high coordination number of Ir–O–Ru bonds in IrRuO_x nanomeshes for highly stable acidic oxygen evolution reaction. *Nano Research*, **2024**, *17*: 5073–5079.
- [28] Han X, Wu G, Ge Y Y, et al. In situ observation of structural evolution and phase engineering of amorphous materials during crystal nucleation. *Advanced Materials*, **2022**, *34* (50): 2206994.
- [29] Meng G, Chang Z W, Zhu L B, et al. Adsorption site regulations of [W–O]-doped CoP boosting the hydrazine oxidation-coupled hydrogen evolution at elevated current density. *Nano-Micro Letters*, **2023**, *15*: 212.
- [30] Zhu L B, Huang J, Meng G, et al. Active site recovery and N–N bond breakage during hydrazine oxidation boosting the electrochemical hydrogen production. *Nature Communications*, **2023**, *14*: 1997.