

Size-controllable PtPdRuRhCuMo high-entropy alloy nanoparticles for electrocatalytic ethylene glycol oxidation

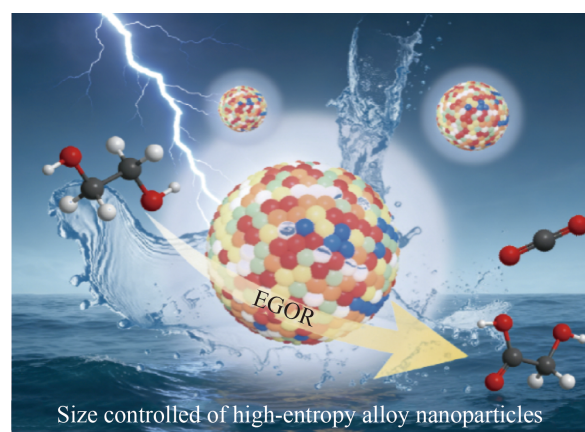
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Abstract: High-entropy alloy nanoparticles (HEA NPs), with unique multi-element synergistic effects and excellent structural stability, are promising core materials for high-efficiency electrocatalysts, but precise regulation of their morphology and composition remains a critical challenge. In this study, cetyltrimethylammonium bromide (CTAB) and cetyltrimethylammonium chloride (CTAC) were used as structure-directing agents to controllably synthesize PtPdRuRhCuMo HEA NPs *via* solvothermal method, and their regulatory effects on material properties were systematically investigated. Structural characterizations showed HEA-1 (CTAB-prepared) had superior dispersibility, uniform particle size and high crystallinity, while HEA-2 (CTAC-prepared) exhibited agglomeration and uneven particle size. Electrochemical tests revealed HEA-1 achieved a mass activity of 2.4 A/mg in alkaline EGOR, significantly higher than HEA-2 and commercial Pt/C (1.25 A/mg) and retained 50% initial activity after 10 h continuous testing, showing certain stability. Structure-directing agents determine HEA NPs' structural characteristics and thus their electrocatalytic performance; HEA-1's excellence is attributed to CTAB-mediated regular structure and strong multi-metal electronic interactions. This work provides a new strategy for precise regulation of HEA NPs and lays a foundation for their practical application in energy conversion systems.

Key words: high-entropy alloy; electrocatalysis; ethylene glycol oxidation; morphology regulation



The relentless depletion of global fossil energy reserves, coupled with the pressing challenge of exorbitant extraction costs, renders the development of novel, clean, efficient, and sustainable energy conversion technologies an urgent imperative^[1–3]. Among the numerous strategies addressing the energy crisis, direct ethylene glycol fuel cells (DEGFCs) stand out due to their unique advantages^[4–5]. As the core fuel for DEGFCs, ethylene glycol exhibits a host of notable characteristics. It can be sustainably synthesized *via* the catalytic conversion of biomass-derived compounds such

as glycerol and cellulose. This synthetic approach endows the feedstock with renewability, thereby ensuring the sustainability of energy supply at the source^[6]. Furthermore, ethylene glycol possesses a high theoretical capacity of 4.8 A·h/mL, which provides a robust material basis for the construction of high-efficiency energy systems^[7]. Nevertheless, the cleavage of the C–C bond along with the incomplete electrooxidation of ethylene glycol (EG) poses substantial hurdles to the practical commercialization of DEGFCs.

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The electrocatalytic ethylene glycol oxidation reaction (EGOR), a critical process in DEFCs, directly influences overall cell efficiency and application prospects. However, this reaction system faces complex challenges. Structurally, EG contains two hydroxyl groups, resulting in an intricate oxidation pathway^[8]. In general, the oxidation process of EGOR proceeds *via* two distinct pathways, C₁ and C₂. Specifically, the C₁ pathway involves a complete cleavage of the C–C bond accompanied by a 10 e[−] transfer, yielding CO₂ and its derivatives (CO₃^{2−}, HCO₃[−], HCOO[−]). In contrast, the C₂ pathway corresponds to the incomplete oxidation of ethylene glycol with a 4e[−]/8e[−] transfer, where the primary products are glycolic acid or oxalic acid^[9–10]. Notably, the C₁ pathway exhibits more pronounced advantages over the C₂ pathway, releasing higher electrical energy. Alloying active noble metals (Pd, Pt) with other noble metals (Ir, Ag, Ru) or non-noble metals is a feasible strategy to promote the C₁ pathway, as it optimizes the catalyst's electronic structure and facilitates C–C bond oxidative cleavage for C1 product formation^[11]. However, conventional Pt-based and Pd-based EGOR catalysts suffer from suboptimal active site distribution, low product selectivity, and poor stability^[12–13].

In this context, high-entropy alloys (HEAs), as an innovation in materials science, present new opportunities for EGOR. Typically composed of five or more principal elements in near-equiatomic ratios (5%–35%), HEAs exhibit lattice distortion, sluggish diffusion, and a “cocktail” effect, which confer unique properties and demonstrate potential for applications in fields such as catalysis^[14–17]. For instance, Liu et al.^[18] prepared defect-rich FeCoNiMnRu HEAs *via* rapid Joule heating. Han et al.^[19] reported a facile co-reduction method to synthesize ultrathin PtPdRuFeCoNi HEA nanowires (NWs) with an average diameter of only 1.36 ± 0.05 nm, which exhibited notable catalytic performance in HER and MOR. Wang's team designed a class of PtIrRhCoFeNiCu HEA nanodendrites using a simple wet-chemical method; the introduction of transition metals enhanced lattice distortion and facilitated complete C–C bond cleavage during ethanol oxidation^[20]. Existing studies indicate that HEA synthesis often requires elevated temperatures to overcome kinetic limitations, while the material morphology is constrained by the incorporated metals and morphology-directing agents^[21].

In this work, leveraging the morphology-directing effect of CTAB and CTAC, PtPdRuRhCuMo high-entropy alloy nanoparticles (HEA NPs) were successfully synthesized *via* a facile solvothermal method. These nanoparticles exhibit the characteristics of excellent dispersibility, uniform size distribution and high charge separation efficiency, which provide a structural basis for enhancing the reaction activity and stability. The as-prepared electrocatalyst demonstrates outstanding electrocatalytic performance toward the EGOR in an alkaline medium, which is significantly superior to that of the conventional commercial Pt/C catalyst. Among the synthesized samples, HEA-1 (using CTAB as the surfactant) achieves the optimal catalytic activity with a mass activity of 2.4 A/mg for EGOR. More importantly,

HEA-1 exhibits remarkable durability in the long-term stability test, which is attributed to the strong interactions between different metals and the excellent dispersibility of the nanoparticles. This study provides a novel strategy for the precise morphology regulation of PtPdRuRhCuMo HEA NPs and expands their potential application scenarios in related fields.

1 Experimental

1.1 Materials

Pt(acac)₂ (purity ≥ 98%), Pd(acac)₂ (purity ≥ 98%), Cu(acac)₂ (purity ≥ 98%), Ru(acac)₃ (purity ≥ 97%) and Rh(acac)₃ (purity ≥ 97%) were purchased from Sinopharm Chemical Reagent Co., Ltd. Commercial Pt/C (20%, Pt loading: 20%, carbon support: XC-72, purity ≥ 99.5%), cetyltrimethylammonium bromide (CTAB, purity ≥ 99%), cetyltrimethylammonium chloride (CTAC, purity ≥ 99%), glucose (purity ≥ 99.5%), Mo(CO)₆ (purity ≥ 98%), and OAm (C₁₈ content: 80%–90%) were all purchased from Shanghai Aladdin Reagent Co., Ltd. 5% Nafion solution (purity ≥ 99%) was purchased from the DuPont company. Ethanol (analytical reagent, AR, purity ≥ 99.7%) and *n*-hexane (analytical reagent, AR, purity ≥ 99.5%) were purchased from Sinopharm Chemical Reagent Co., Ltd.

1.2 Material characterizations

The morphology of as-prepared samples was studied by using transmission electron microscopy (TEM) on a Talos F200X, USA with an energy-dispersive spectrometer (EDS) detector and high-angle angular dark-field scanning transmission electron microscopy (HAADF-STEM) on a ARM200F. X-ray diffraction patterns (XRD) patterns of samples were examined by X-ray diffraction on a Rigaku MLtina IV-185 X-ray diffractometer using Cu Kα radiation in a region of 2θ = 10°–90°, operating with a scanning rate of 10(°)/min. X-ray photoelectron spectroscopy (XPS) measurements were carried out on a ESCALAB-210 using an Al Kα X-ray source. The mass loadings of metal on the UCN were measured by inductively coupled plasma-optical emission spectroscopy (ICP-OES) on a PerkinElmer 8300.

1.3 Preparation of the HEA-1 and HEA-2

The catalyst was synthesized using an improved hydrothermal method. First, 100 mg CTAB was dissolved in 25 mL of OAm solution and heated with stirring in a 60 °C oil bath until the solution became completely transparent. Subsequently, 120 mg glucose was added, followed by 0.25 mmol Mo(CO)₆, 0.05 mmol Pt(acac)₂, 0.05 mmol Pd(acac)₂, 0.05 mmol Cu(acac)₂, 0.05 mmol Ru(acac)₃ and 0.05 mmol Rh(acac)₃. The mixture was continuously stirred and ultrasonically dissolved for 1 h. The resulting slurry was transferred to a Teflon-lined

autoclave (50 mL) and subjected to hydrothermal treatment at 160 °C for 3 h. The reaction was then accelerated by heating to 210 °C for 5 h. After complete cooling to room temperature, the product was washed multiple times with anhydrous ethanol and *n*-hexane, followed by 12 h of vacuum drying at 60 °C to obtain the HEA-1 catalyst.

According to the same preparation method, HEA-2 catalyst was prepared by substituting CTAC for CTAB.

1.4 Electrochemical tests

All electrochemical measurements were performed in a standard three-electrode system using a CHI 760E electrochemical workstation. The working electrode was a glassy carbon electrode (GCE, diameter: 4 mm) loaded with the catalyst, the counter electrode was a carbon rod, and Hg/HgO electrode as reference electrode in 1.0 mol/L KOH, respectively. Notably, the ink was prepared as follows: 2 mg of the prepared catalyst and 8 mg of XC-72 treated with HNO₃ are ground and mixed, then a certain amount of anhydrous ethanol (10 mL) and Nafion (0.5 mL, 5%) solution are added for ultrasonic treatment to form a homogeneous solution. Finally, 6 µL ink was coated on the surface of the glassy carbon electrode polished with Al₂O₃ powder and dried in air for use.

The electrocatalytic ethylene glycol oxidation performance was evaluated by cyclic voltammetry (CV), and all measurements were conducted in a nitrogen-saturated electrolyte at room temperature. In this method, a triangular wave potential scan is applied to the working electrode at a constant scan rate over time to complete one cycle of the redox process, and the recorded current-voltage curve is designated as a cyclic voltammogram. In addition, chronoamperometry (CA) was employed to assess the stability of the catalyst, where a constant potential is applied to the working electrode and the corresponding current-time response curve is recorded. All potentials measured in the alkaline medium were calibrated to the reversible hydrogen electrode (RHE) using the following equation:

$$E_{\text{RHE}} = E_{\text{Hg/HgO}} + 0.098 \text{ V} + 0.059 \text{ pH} \quad (1)$$

2 Results and discussion

2.1 Structure and morphology characterization

The preparation method of the HEA electrocatalyst was shown in Figure 1. In the synthesis, we employed Pd(acac)₂, Rh(acac)₃, Pt(acac)₂, Ru(acac)₃ and Cu(acac)₂ as the metal precursors. In the process of synthesizing nanostructures, CTAB or CTAC is often used as a structure-directing agent. It mainly prevents particle agglomeration through a charge dispersion mechanism, and finally forms a unit structure with a well-defined structure. At the same time, Mo(CO)₆ decomposes to generate CO at high temperatures. CO acts as a mild reducing agent, and its reduction mechanism involves the transfer of electrons from the carbon atom (with a lone pair of electrons) to the metal cations in the

precursors, reducing the metal cations to their metallic state^[22–23]. This process promotes the co-reduction of multi-metal ions and thus facilitates the synthesis of high-entropy alloys (HEA). In addition, ammonia generated by the thermal decomposition of oleylamine (OAm) under solvothermal conditions also acts as a reducing agent. Studies have demonstrated that PdRuPtCuRhMo high-entropy alloy nanoparticles with distinct sizes can be successfully synthesized by regulating the type of structure-directing agent (CTAB/CTAC).

The crystalline structure of the as-obtained samples was performed by X-ray powder diffraction (XRD), as depicted in Figure 2. Four distinct diffraction peaks were observed in all patterns at 40.4°, 46.8°, 68.4° and 82.4°, which correspond to the (111), (200), (220) and (311) crystal planes of the face-centered cubic (fcc) structure, respectively. This result confirmed the formation of a single-phase alloy without phase separation, and the slight shift in the diffraction peaks of different high-entropy alloys is associated with the difference in atomic sizes^[24]. Notably, compared with the diffraction peak intensity of HEA-2, the diffraction peaks of the HEA-1 sample were significantly sharper and stronger, indicating that the crystallinity of HEA-1 was remarkably superior to that of HEA-2^[25]. Inductively coupled plasma optical emission spectrometry (ICP-OES) analysis verified that the atomic ratio of Ru, Rh, Cu, Pt, Pd and Mo in the high-entropy alloy conformed to the characteristic of near-equiatomic composition for high-entropy alloys (Table 1).

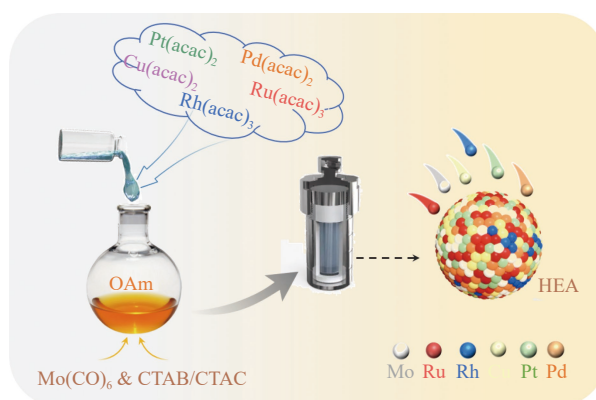


Fig. 1 Schematic illustration of the synthesis of PdRuPtCuRhMo HEA

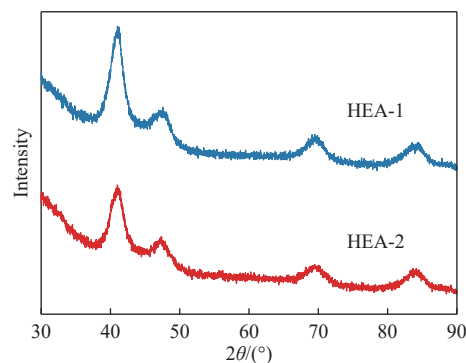


Fig. 2 XRD patterns of HEA-1 and HEA-2

Table 1 The element content of HEA catalysts determined from ICP-OES results

Sample	Ru/%	Rh/%	Cu/%	Pt/%	Pd/%	Mo/%
HEA-1	15.2	16.8	18.5	19.5	17.3	12.7
HEA-2	22.4	18.7	15.8	14.1	16.7	12.3

To further investigate the influence of surfactants on the structure and morphology of materials, transmission electron microscopy (TEM) measure was employed. As depicted in Figure 3(a), (b), HEA-1 and HEA-2 displayed similar spherical morphologies, differing only in particle size. The average particle size of HEA-1 was 4.77 nm, while that of HEA-2 was approximately 5.27 nm. Combined with high-resolution transmission electron microscopy (HRTEM) images (Figure 3(c), (f)), the lattice fringes of the PdRuPtCuRhMo HEA were clearly visible, reflecting the ordered crystal structure of the alloy.

The imaging results demonstrated that the as-synthesized HEA-1 nanoparticles exhibit a more regular structure and more uniform dispersion, indicating that CTAB is more conducive to metal aggregation and crystallinity enhancement during the synthesis process. The distinct lattice fringes with interplanar spacings of 0.223 and 0.216 nm observed in Figure 3(d), (g) correspond to the (111) crystal plane of the face-centered cubic (fcc) structure. Compared with the standard lattice spacing of palladium (0.22 nm), these values confirm the presence of significant tensile/compressive strain inside the material. As shown in Figure 3(e), (h), the internal characteristics of the nanoparticles were investigated *via* geometric phase analysis (GPA) based on high-resolution transmission electron microscopy (HRTEM) images. The combined results of HRTEM and GPA characterizations verify that the high-entropy alloys were mainly featured by prominent

strain distribution characteristics. High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) coupled with elemental mapping results (Figure 3(i)) reveal that the six elements (Rh, Pt, Pd, Ru, Cu and Mo) were uniformly distributed throughout the HEA-1 nanoparticles.

The surface electronic state of two HEA NPs were meticulously examined through X-ray photoelectron spectroscopy (XPS) analysis. The XPS survey spectrum indicated that the elements Pd, Rh, Pt, Ru, Cu and Mo coexist in the HEA. Among these elements, the chemical states of the noble metals (Pt, Pd and Rh) were predominantly metallic, albeit with the presence of a small amount of oxidation. Considering the overlap in binding energy between Ru 3d_{3/2} and C 1s, we only analyzed the Ru 3d_{5/2} XPS spectrum as shown in Figure 4(a). It can be observed from the spectrum that ruthenium primarily exists in the metallic state (Ru⁰) on the material surface^[26–27]. For the high-resolution Cu 2p spectrum (Figure 4(b)), two distinct peaks can be clearly observed at approximately 931.8 and 951.7 eV, which correspond to Cu⁺ or Cu⁰ respectively^[28–29]. Given that XPS was a surface-sensitive technique and the XRD pattern matched the alloy phase very well, the detected Cu⁺ signal was mainly attributed to the surface oxidation of Cu⁰ in the alloy during air exposure after synthesis, as well as the electronic interaction between Cu and other metal components.

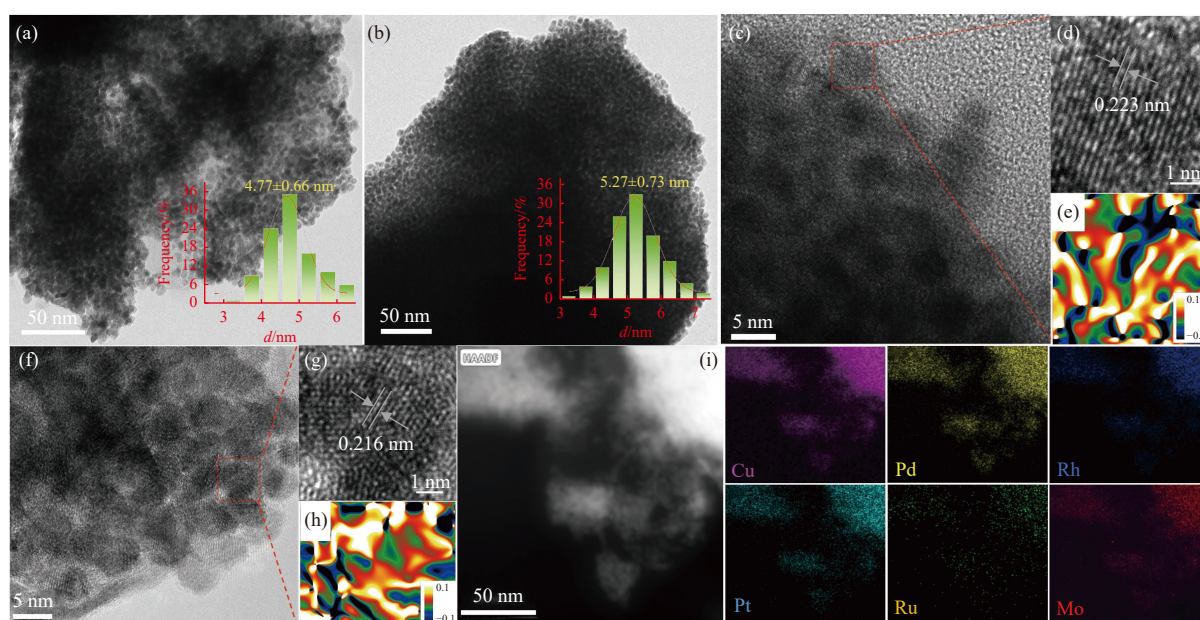


Fig. 3 TEM images and size distributions of (a) HEA-1 and (b) HEA-2; HRTEM images of (c) HEA-1 and (f) HEA-2; (d), (g) Enlarged selected of HEA-1 and HEA-2, respectively; (e), (h) Geometric phase analysis (GPA) for the lattice strain distribution taken from the areas in (c) and (f); (i) HAADF-TEM of HEA-1 and corresponding elemental mapping of Rh, Pt, Pd, Ru, Cu and Mo

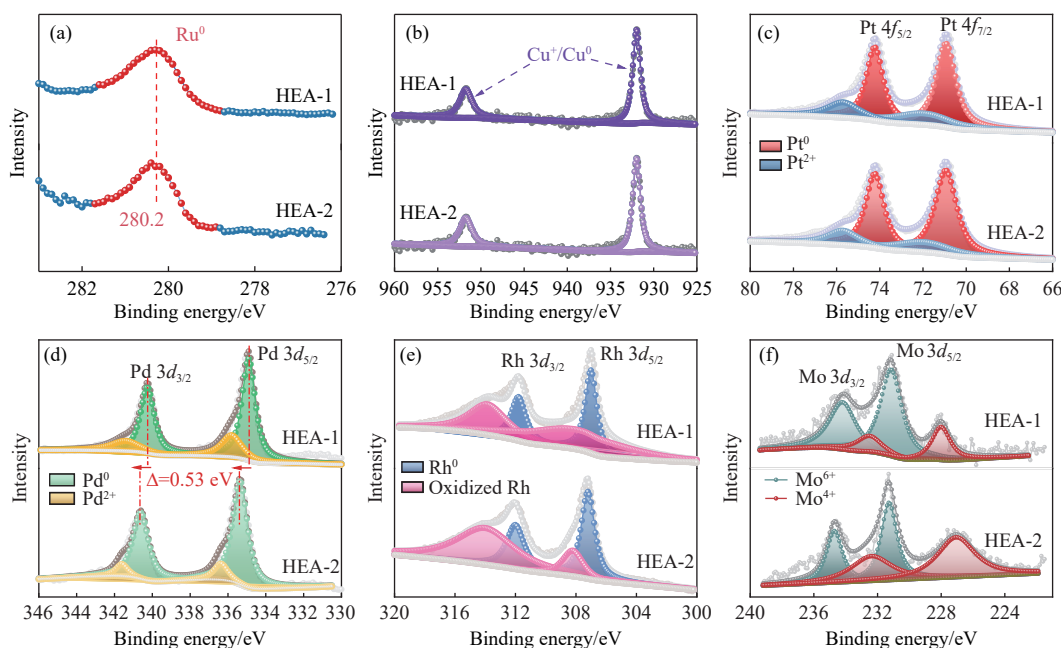


Fig. 4 High resolution Ru 3d (a), Cu 2p (b), Pt 4f (c), Pd 3d (d), Rh 3d (e) and Mo 3d (f) XPS spectra of the HEA

Furthermore, Figure 4(c) showed a doublet of Pt $4f_{7/2}$ and $4f_{5/2}$ in the Pt 4f region, where Pt existed in the forms of Pt²⁺ and Pt⁰. Specifically, the peaks at 71.0 and 74.2 eV belong to Pt⁰, while the peaks appearing at 75.7 and 72.0 eV are attributed to Pt²⁺, indicating that part of Pt²⁺ was reduced to form Pt⁰. The Pd 3d XPS spectrum revealed the predominance of metallic Pd with a minor fraction of metal oxides on the material surface (Figure 4(d)). Specifically, the peaks centered at 334.9 and 340.2 eV were assigned to Pd⁰ $3d_{5/2}$ and Pd⁰ $3d_{3/2}$, respectively, whereas those at 335.8 and 341.4 eV correspond to Pd²⁺ $3d_{5/2}$ and Pd²⁺ $3d_{3/2}$.

Notably, the two Pd 3d peaks in HEA-1 displayed a positive binding energy shift relative to their counterparts in HEA-2. Specifically, the Pd $3d_{5/2}$ peak of HEA-1 was located at 334.9 eV, while that of HEA-2 was at 334.37 eV, corresponding to a positive shift of ~0.53 eV (as indicated by arrows in Figure 4(d)). That indicated a stronger alloying interaction between Pd and other metallic components in HEA-1 and characterized by more pronounced electron transfer. This phenomenon is concurrently accompanied by a smaller grain size, a more homogeneous elemental distribution, and lattice tensile strain^[30]. Similarly, most Rh existed as a zero-valent substance, while a portion existed as an oxidized substance (Figure 4(e)). In particular, the high-resolution Rh 3d spectrum of HEA consisted of four fitted peaks: Rh⁰ $3d_{5/2}$ (307.0 eV), Rh⁰ $3d_{3/2}$ (311.8 eV), Rh³⁺ $3d_{5/2}$ (308.2 eV) and Rh³⁺ $3d_{3/2}$ (313.9 eV)^[29]. In the Mo 3d region (Figure 4(f)), the peaks of the HEA could be deconvoluted into two components corresponding to Mo⁴⁺ and Mo⁶⁺ species. Mo⁶⁺ was derived from the decomposition of Mo(CO)₆, whereas Mo⁴⁺ is generated *via* the reduction of Mo⁶⁺ by Cu^[31]. Notably, the characteristic peaks of HEA-2 exhibit a relatively higher content of Mo⁴⁺ compared with those of HEA-1, indicating that a greater amount of Cu participates in the reaction with Mo⁶⁺ in HEA-2^[32–33].

2.2 Electrochemical measurements

Before the test, all catalysts were activated *via* cyclic voltammetry (CV) measurements in 1.0 mol/L KOH solution, which was saturated with N₂. Figure 5(a) illustrated the normalized CV curves of these catalysts in a 1.0 mol/L KOH solution with the scan rate of 50 mV/s from 0 to 1.50 V (vs. RHE). The voltammograms exhibited well-established features of adsorption and desorption of the H species on Pd and Pt sites^[34]. Furthermore, the formation of HEAs and hybrid structures can significantly enhance electrochemical activity and metal utilization efficiency. The electrocatalytic performance of these samples toward EGOR was tested in 1.0 mol/L KOH + 0.5 mol/L EG solution at the scan rate of 50 mV/s, where the PGM-based current density was calculated from the contents of noble metals (Pd, Pt, Rh and Ru) in the material (Figure 5(b)). Two characteristic oxidation peaks were observed from CV curves, in which the peak in the forward scan was associated with ethylene glycol oxidation, while the peak in the reverse scan originated from further oxidation or removal of the poisoning carbonaceous intermediate.

Firstly, we compared the performance of HEA-1 and HEA-2. HEA-2 exhibited a relatively low oxidation peak current (< 2.0 A/mg), indicating its inability to effectively enhance the electrochemical EGOR efficiency. In contrast, HEA-1 demonstrated superior EGOR performance, with a peak current density as high as 2.4 A/mg, which was 1.5 times the activity of HEA-2 and 1.8 times that of the commercial Pt/C (20%) catalyst, which exhibited distinct advantages compared with various recently reported catalysts (Table S1). This fully confirms that the synergistic interactions between different elements in HEA are beneficial for boosting catalytic activity. Although HEA-1 showed only slight morphological differences from HEA-2,

such differences (e.g., in dispersity, size, and crystallinity) can affect catalytic properties. Thus, the role of surfactants in improving catalytic activity should not be overlooked. In fact, as we subsequently observed through comparing the ethylene glycol concentration dependence and stability of different catalysts, the material structure plays a pivotal role in promoting the EGOR. Notably, as the ethylene glycol concentration was continuously increased, the current density of HEA-1 decreased significantly more slowly than that of HEA-2. When the ethylene glycol concentration reached 2.0 mol/L, the current density achieved by HEA-1 was twice that of HEA-2 (Figure 5(c)). By fitting the Nyquist plots in Figure 5(d) with the equivalent circuit (inset of Figure 5(d)), the charge transfer resistance (R_{ct}) of HEA-1 is determined to be 13.65 Ω , which is much lower than that of HEA-2 (37.15 Ω) and commercial Pt/C (47.48 Ω). This indicates a faster charge transfer process between the HEA-1 nanoparticles and the electrolyte, favoring the EGOR reaction.

In addition to catalytic activity, durability constitutes another crucial parameter determining the potential practical application of EGOR electrocatalysts. As depicted in Figure 5(e), the stability tests conducted *via* cyclic

voltammetry (CV) curves demonstrate that HEA-1 exhibits higher mass activity than HEA-2 and commercial Pt/C over 500 scanning cycles during the EGOR. Furthermore, we characterized the durability of the HEA *via* chronoamperometry ($i-t$) measurements in 1.0 mol/L KOH solutions containing 0.5 mol/L EG (Figure 5(f)). Under identical experimental conditions, the HEA-1 nanoparticles retained an excellent 50% of their initial activity even after a prolonged $i-t$ test of 10 h, indicating superior electrocatalytic stability. Characterization of the materials after the stability test (Figure S1–S4) revealed that although HEA-1 exhibited partial deactivation, its deactivation degree was significantly lower than that of commercial Pt/C and HEA-2. This is attributed to the multimetallic synergistic effect and excellent structural stability of the material, which effectively suppress severe agglomeration and deep oxidation. However, when the EG concentration was increased, a significant decay in the current density of the HEA nanoparticle catalyst was observed, suggesting that the catalyst's anti-poisoning capability in high-concentration ethylene glycol requires further improvement (Figure S5).

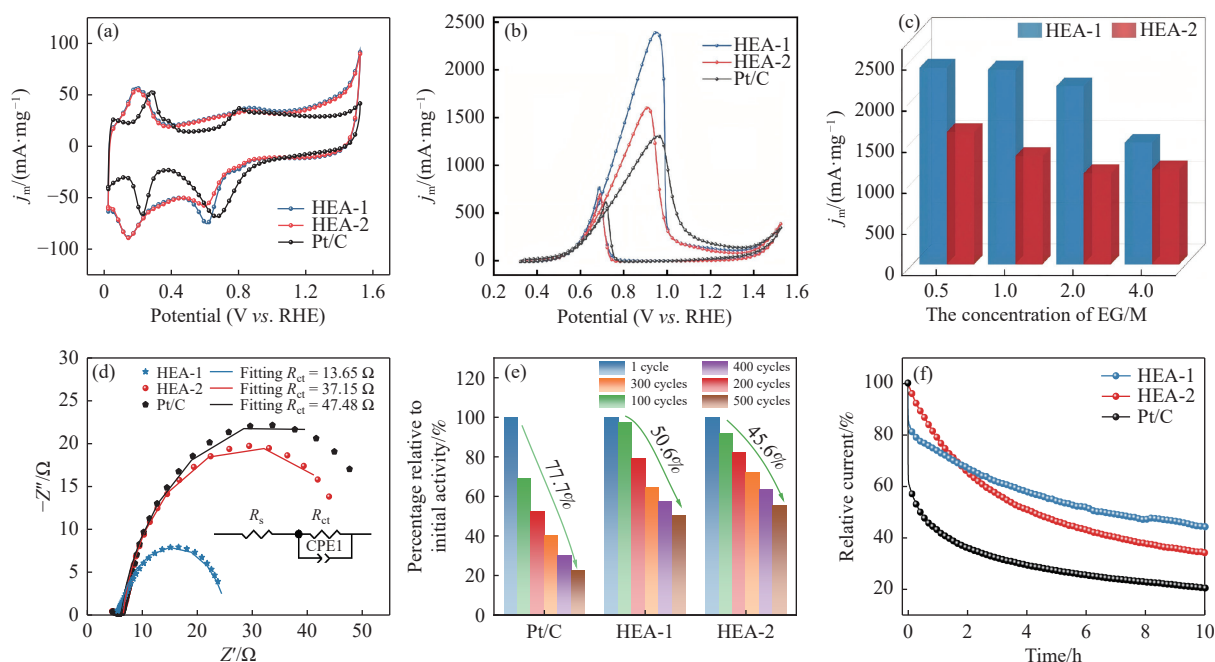


Fig. 5 CVs of the catalysts in (a) 1.0 mol/L KOH, (b) 1.0 mol/L KOH + 0.5 mol/L ethylene glycol solution, (c) The mass specific activity in different concentrations of EG + 1.0 mol/L KOH solution, (d) EIS Nyquist plots of EGOR on the catalysts in 1.0 mol/L KOH + 0.5 mol/L EG solution (Inset: The equivalent circuit used for fitting. R_s : solution resistance; R_{ct} : charge transfer resistance; CPE: constant phase element), (e) Cycling stability of electrocatalysts (The vertical axis represents the activity retention rate relative to the initial mass activity, tested in 1.0 mol/L KOH + 0.5 mol/L EG at a scan rate of 50 mV/s for 500 cycles), (f) Long-term chronoamperometry for EGOR at 0.90 V versus RHE (Tested in 1.0 mol/L KOH + 0.5 mol/L EG)

3 Conclusions

In summary, this study successfully fabricated well-defined PtPdRuRhCuMo HEA NPs *via* a facile solvothermal method, leveraging the morphology-directing

effects of CTAB and CTAC. The resulting catalyst demonstrated exceptional performance in the alkaline EGOR, exhibiting significantly superior mass activity and outstanding long-term stability compared to commercial Pt/C. Comprehensive characterization and analysis revealed

that the performance enhancement originates from the unique multi-element synergistic effects of the high-entropy alloy: on one hand, the coexistence of metals with different atomic radii induces considerable lattice tensile strain, optimizing the adsorption of reaction intermediates; on the other hand, the distinct electronegativity differences among components promote interfacial electron transfer. These factors collectively modulate the electronic structure of the active sites, thereby substantially enhancing the intrinsic activity. Concurrently, strong intermetallic interactions and the uniform nanostructure ensure structural integrity and resistance to aggregation during the reaction process. This work not only provides a new strategy for the microstructural regulation of multi-component alloys but also deepens the understanding of the structure-activity relationships in high-entropy systems.

Declaration of Competing Interest

The authors declare no conflict of interest.

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尺寸可控 PtPdRuRhCuMo 高熵合金纳米颗粒用于电催化乙二醇氧化

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摘 要: 高熵合金纳米颗粒(HEA NPs)凭借独特的多元素协同效应与优异的结构稳定性, 成为高效电催化剂设计中极具应用前景的核心材料, 但形貌与组成的精准调控仍是关键挑战。本研究分别以十六烷基三甲基溴化铵(CTAB)与十六烷基三甲基氯化铵(CTAC)为结构导向剂, 通过溶剂热法可控制备 PtPdRuRhCuMo HEA NPs, 探究其对材料形貌、分散性、粒径及结晶性的调控规律。结构表征表明, CTAB 制备的 HEA-1 分散性更优、粒径分布更均匀、结晶度更高, 而 CTAC 制备的 HEA-2 存在团聚与粒径不均现象。电化学测试表明, HEA-1 在碱性乙二醇氧化反应(EGOR)中质量活性达 2.4 A/mg, 优于 HEA-2 及商业 Pt/C 催化剂(1.25 A/mg), 且 10 h 连续测试后保留 50% 初始活性, 具有一定的结构稳定性。研究证实, 结构导向剂直接决定 HEA NPs 的结构特性, 进而影响其催化性能, HEA-1 的优异性能源于 CTAB 介导的规整结构与多金属电子相互作用。本研究为 HEA NPs 精准调控提供新策略, 也为其在能源转换系统中的实际应用奠定基础。

关键词: 高熵合金; 电催化; 乙二醇氧化; 形貌调控

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