

Ultrasound-Assisted Synthesis of High-Entropy Materials for Enhanced Oxygen Evolution Electrocatalysis

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Electrocatalytic measurements

Electrochemical measurements of the as-synthesized samples were performed with a CHI760E electrochemistry workstation (CH Instruments, Inc.) using a standard three-electrode electrochemical cell with a Pt sheet and Hg/HgO as the counter electrode and the reference electrode, respectively. The catalyst ink for OER tests was prepared as follows: 5 mg of catalysts was dispersed into a mixed solution of 300 μL of a 0.5% Nafion ethanol solution and 200 μL of deionized water, and then ultrasonicated for 30 min to obtain a homogeneous solution. In total, 50 μL of the catalysts' ink was loaded on a clean NF electrode (NF, 0.5 cm^2) by drop coating to form the electrode, and dried slowly at room temperature. The final mass loading of the catalysts on the electrode is around 1 mg cm^{-2} .

The electrochemical measurements were all performed at room temperature, and the potential was referenced to that of the reversible hydrogen electrode (RHE). For the RHE calibration, the potential difference between Hg/HgO and RHE was measured in a 99.999%-pure- H_2 -saturated 1.0 M KOH aqueous solution. During the measurement, high-purity H_2 is bubbled into the electrolyte to saturate the electrolyte and fix the

reversible hydrogen potential.

The TOF values are calculated via the following equation:

$$\text{TOF} = \frac{jA}{4 \times F \times n} \quad (\text{S1})$$

Where j is the current density at an overpotential of 300 mV during the LSV measurement in the 1.0 M KOH solution. A stands for the area of the electrode (0.5 cm^2) and F is the Faradaic constant (96485 C mol^{-1}). The 4 accounts for the electrons consumed to form an O_2 molecule from water ($4e^-$ for OER). n represents the quantity of active sites, and n can be calculated as follows. If it is assumed that all metal ions are involved in the electrocatalytic reaction, then the n value can be calculated from the XPS results:

$$n = \frac{(m_{\text{catalyst}} \times C_{\text{wt}\% - M})}{M} \quad (\text{S2})$$

Where m_{catalyst} is the catalyst loading on the NF electrode (0.5 mg); $C_{\text{wt}\%}$ is the concentration of metal derived from XPS.

Mass activity (MA)

Mass activity ($\text{A} \cdot \text{g}^{-1}$) equals the measured current density j (mA cm^{-2}) at

$\eta=300 \text{ mV}$ divided by the catalyst loading amount 1 mg cm^{-2} .

$$\text{Mass activity} = \frac{j}{m} \quad (\text{S3})$$

Intrinsic activity (SA)

The area-specific activity, then, represents the current density per unit of electrochemically active area, per unit mass of the catalyst. It can be calculated according to the following formula:

$$SA = \frac{MA}{ECSA} \quad (S4)$$

$$ECSA = \frac{C_{dl}}{C_s} \quad (S5)$$

Supporting Figures

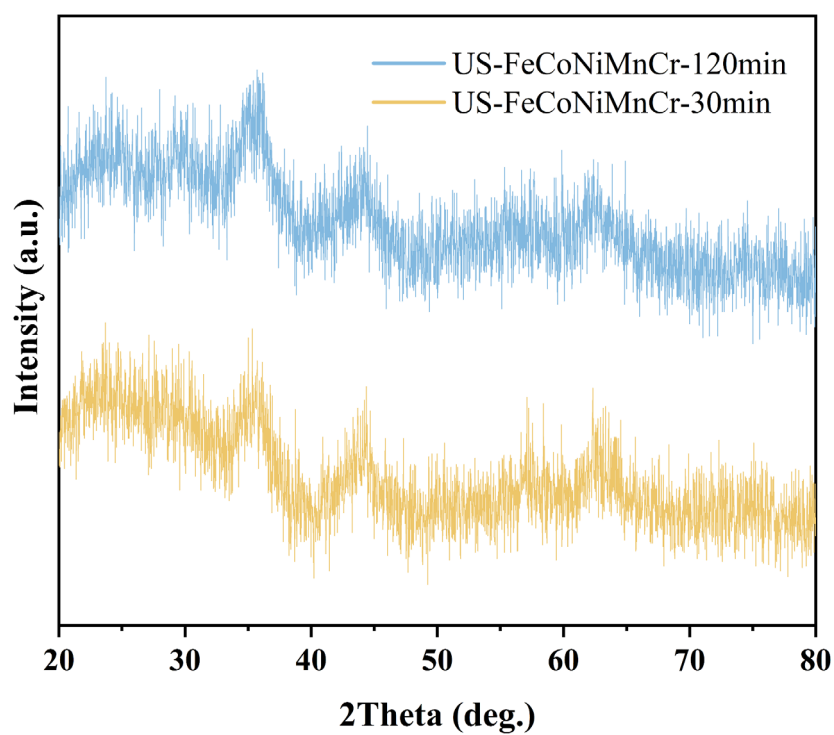


Figure S1. XRD patterns of US-FeCoNiMnCr-30min and US-FeCoNiMnCr-120min.

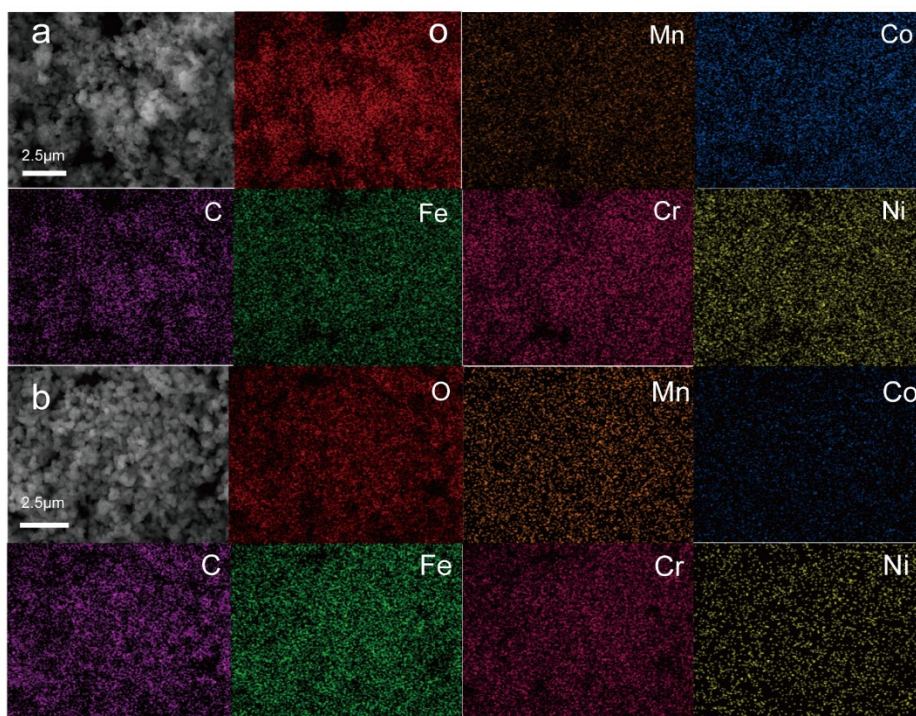


Figure S2. The EDS image of (a) FeCoNiMnCr and (b) US-FeCoNiMnCr.

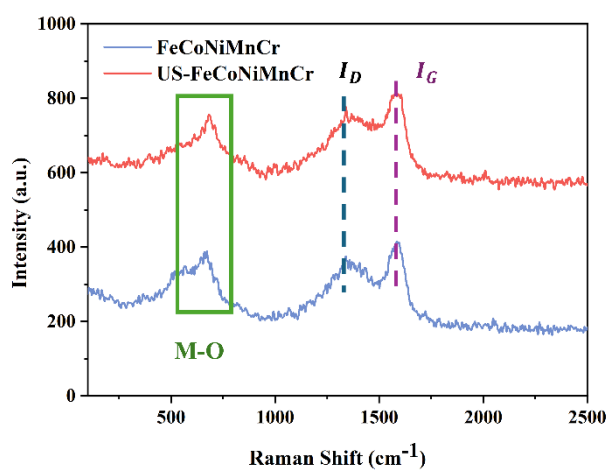


Figure S3. Raman spectra of FeCoNiMnCr and US-FeCoNiMnCr.

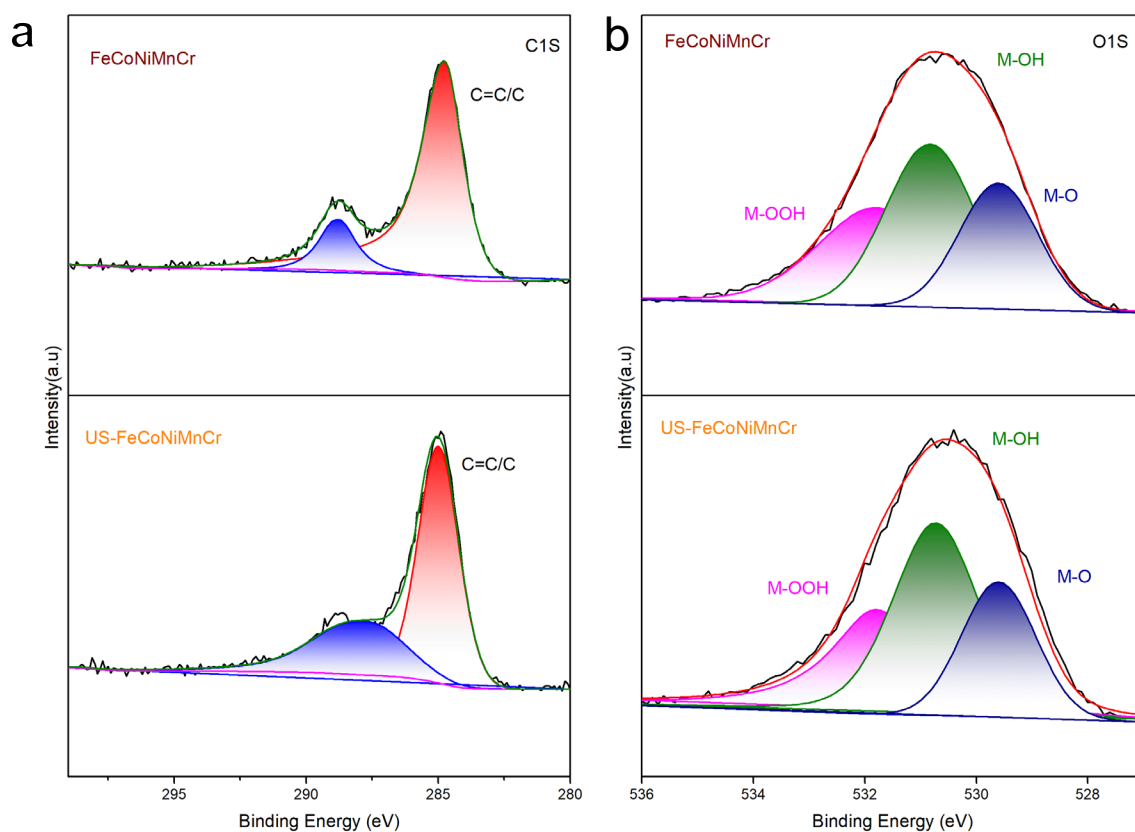


Figure S4. XPS (a) C 1s and (b) O 1s spectra of FeCoNiMnCr and US-FeCoNiMnCr.

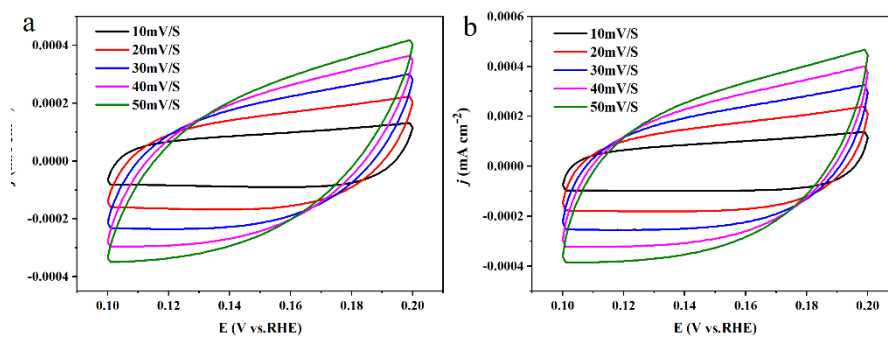


Figure S5. Cyclic voltammogram curves of (a) US-FeCoNiMnCr and (b) FeCoNiMnCr in 1.0 M KOH.

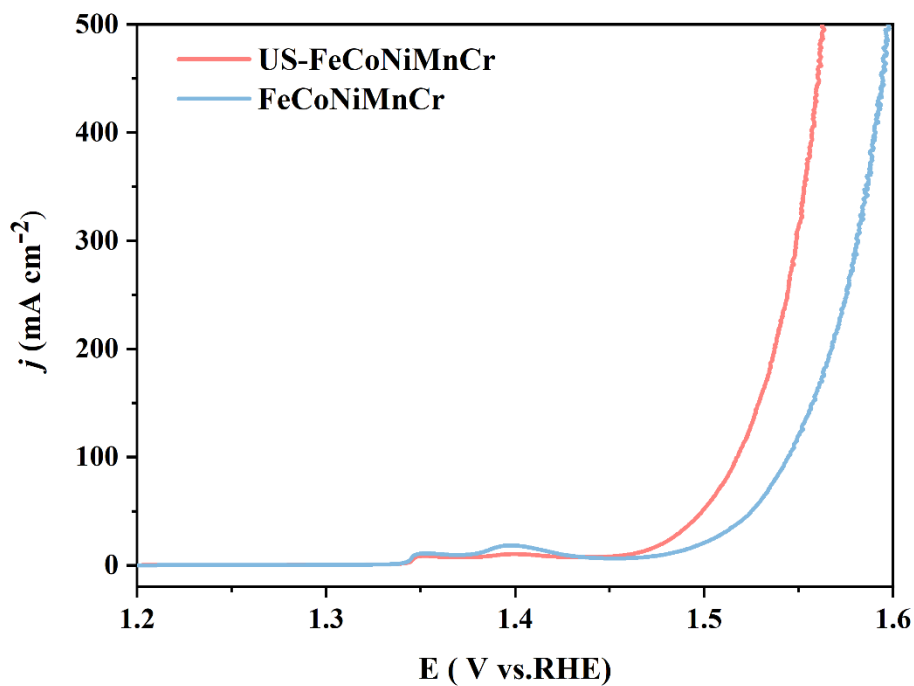


Figure S6. ECSA-normalized LSV curves of US-FeCoNiMnCr and FeCoNiMnCr catalysts.

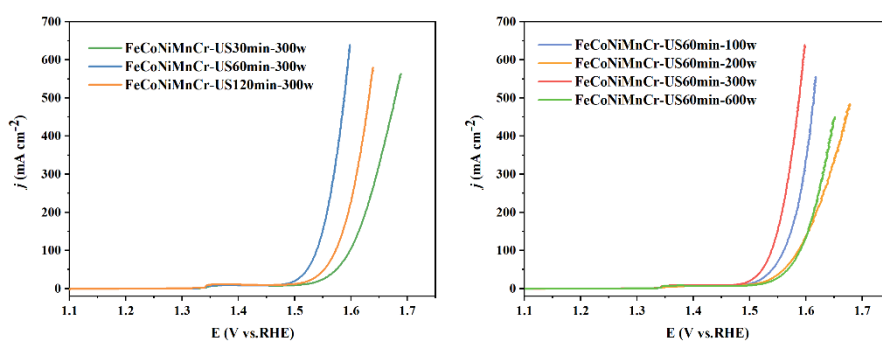


Figure S7. LSV curves of US-FeCoNiMnCr catalysts under (a) different ultrasound time, and (b) different ultrasonic powers in 1.0 M KOH.

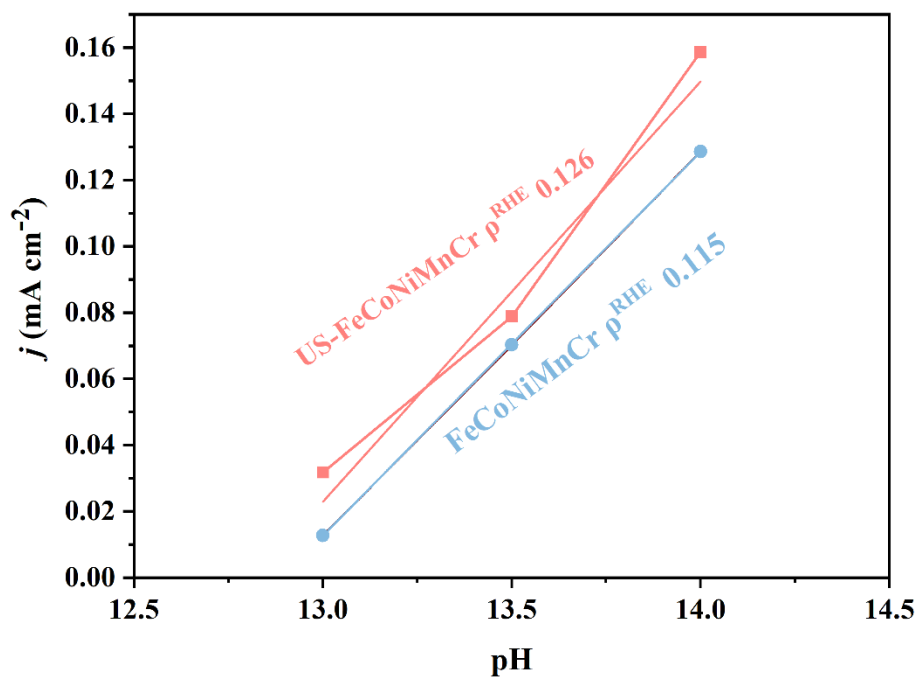


Figure S8. j plots as a function of pH on a logarithmic scale for RHE at 1.55V.

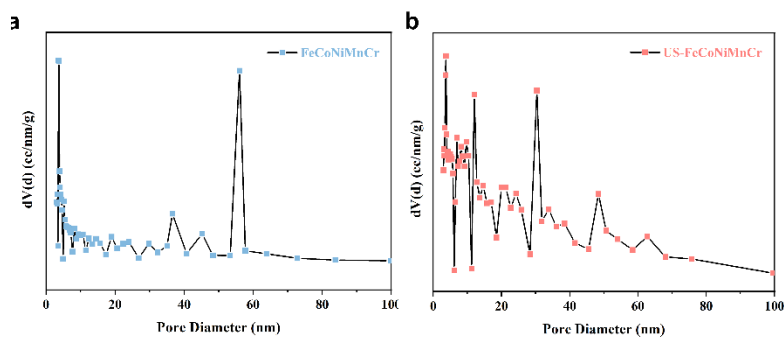


Figure S9. Pore size distribution of the (a) FeCoNiMnCr and (b) US-FeCoNiMnCr catalysts.

Supporting Table

Table S1. Fe, Co, Ni, Mn, and Cr contents of the two catalysts measured by ICP-OES.

Sample	Fe(wt%)	Co(wt%)	Ni(wt%)	Mn(wt%)	Cr(wt%)
FeCoNiMnCr	12.53%	13.12%	15.05%	6.65%	11.60%
US-FeCoNiMnCr	20.85%	3.63%	4.75%	1.63%	18.31%