Supporting information for

Controllable Electrodeposition of Binary Metal Films from Deep Eutectic Solvent as Efficient and Durable Catalyst for Oxygen Evolution Reaction

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Table S1 Composition of DES-based plating bath

Sample	$NiCl_2.6H_2O$ (mmole)	$FeCl_3.6H_2O$ (mmole)	Choline chloride (mmole)	Ethylene glycol (mmole)
Ni	7.0	-		
$Ni_{25}Fe_{75}$	1.75	5.25		
$Ni_{50}Fe_{50}$	3.5	3.5	145	290
$Ni_{75}Fe_{25}$	5.25	1.75		
Fe	-	7.0		

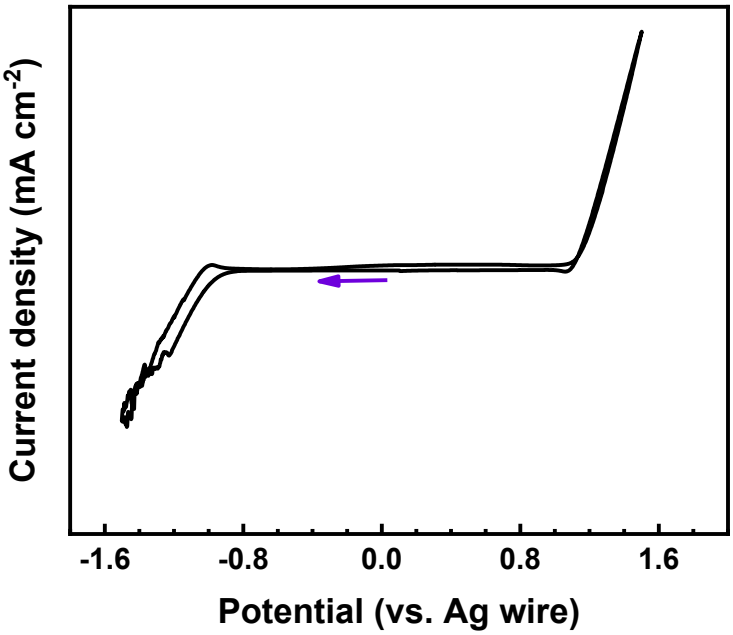

Figure S1 Cyclic voltammetry of the neat DES with a 5 mV s^{-1} scan rate at 60 °C.

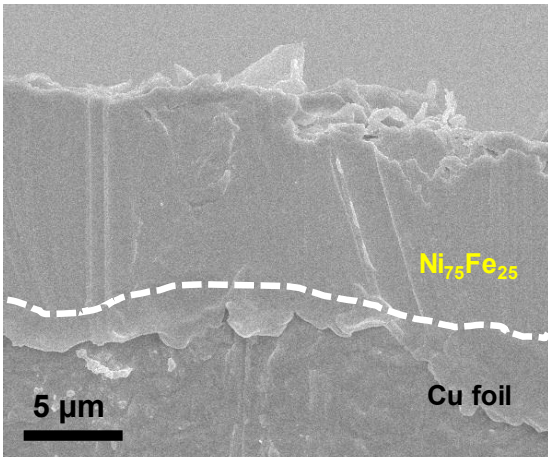

Figure S2 Cross-sectional SEM image of $Ni_{75}Fe_{25}$ film

Interplanar spacing can be calculated easily according to Bragg's Law:

Order of Reflection (n) × Wavelength (λ) = 2 × Interplanar spacing (d) × sinθ

The lattice parameter *a* could be calculated from the *d*-spacing values of major intensity peaks as given by $a = \sqrt{3}d$ for an fcc structure and $a = \sqrt{2}d$ for a bcc structure.

Table S2 Variation of *d*-spacing and lattice parameter, for different Ni and NiFe alloys.

Sample	d-spacing (Å)	Lattice parameter (Å)
Ni	0.203	3.516
$Ni_{75}Fe_{25}$	0.206	3.568
$Ni_{50}Fe_{50}$	0.207	3.585
$Ni_{25}Fe_{75}$	0.204	3.521

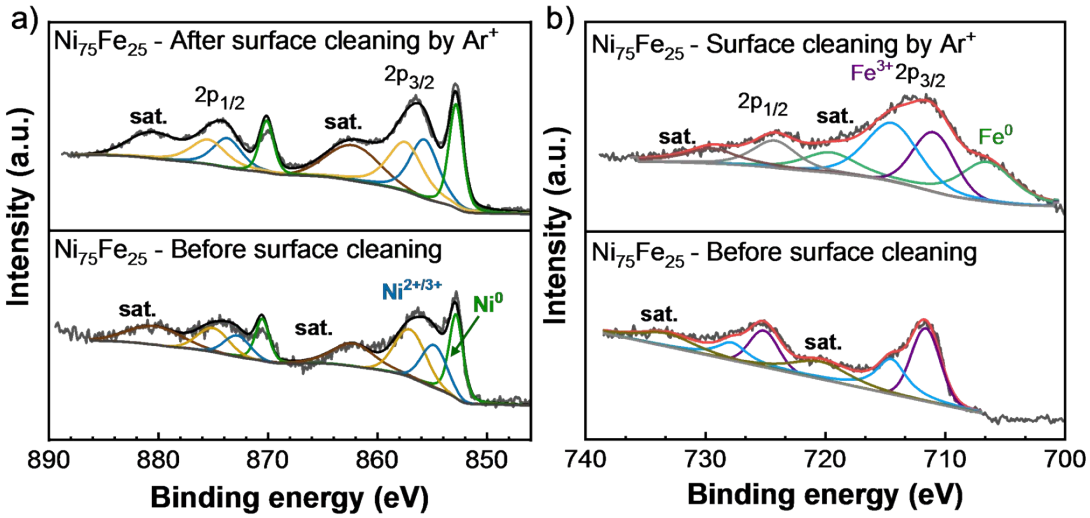

Figure S3 (a) Ni 2p and (b) Fe 2p XPS spectra of mixed $Ni_{75}Fe_{25}$ films electrodeposited in DES before and after surface cleaning by Ar^+ ion.

Electrochemical active surface area (ECSA) The active surface area of each catalyst was measured from their electrochemical capacitances in a non-faradic region using a simple cyclic voltammetry method. The double layer current is equal to the product of the scan rate and the capacitance, which is expected to be linearly proportional to the active surface area of electrode. By plotting the capacitive currents (J_{anodic} - $J_{cathodic}$) versus scan rate, the capacitance can be estimated as half of the slope.

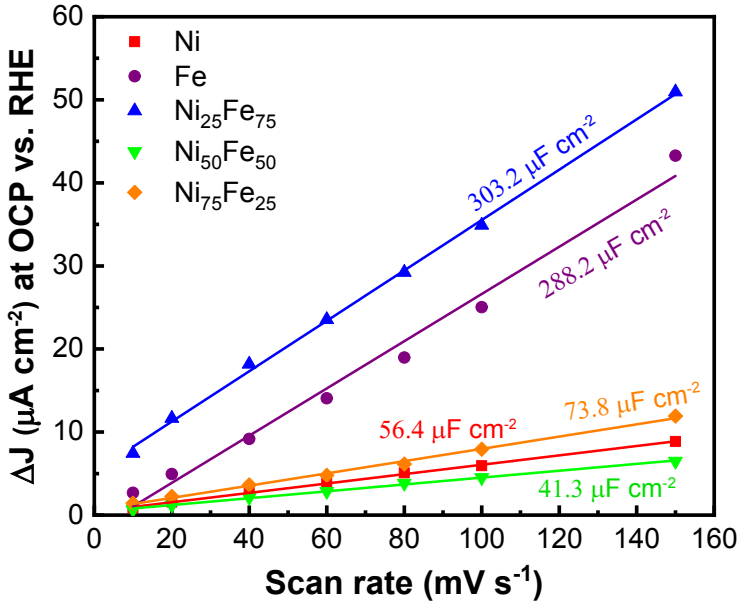

Figure S4 Pots of the current density differences vs. the scan rate for various electrodes

Table S3 EIS parameter derived from fitting with Randle circuit of mixed Ni-Fe films electrodeposited in DES with varying catalyst composition.

Sample	**R_s (Ω)**	**R_{ct} (Ω)**
Ni	13.4	11000
$Ni_{75}Fe_{25}$	14.1	8
$Ni_{50}Fe_{50}$	14.6	36
$Ni_{25}Fe_{75}$	14.9	51
Fe	14.2	12600

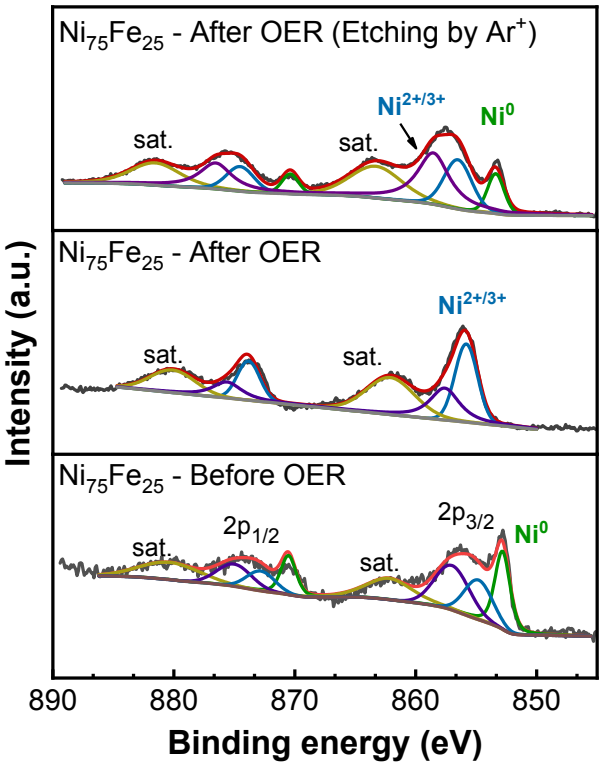

Figure S5 High-resolution XPS spectra of Ni 2p in $Ni_{75}Fe_{25}$ film before and after OER reaction

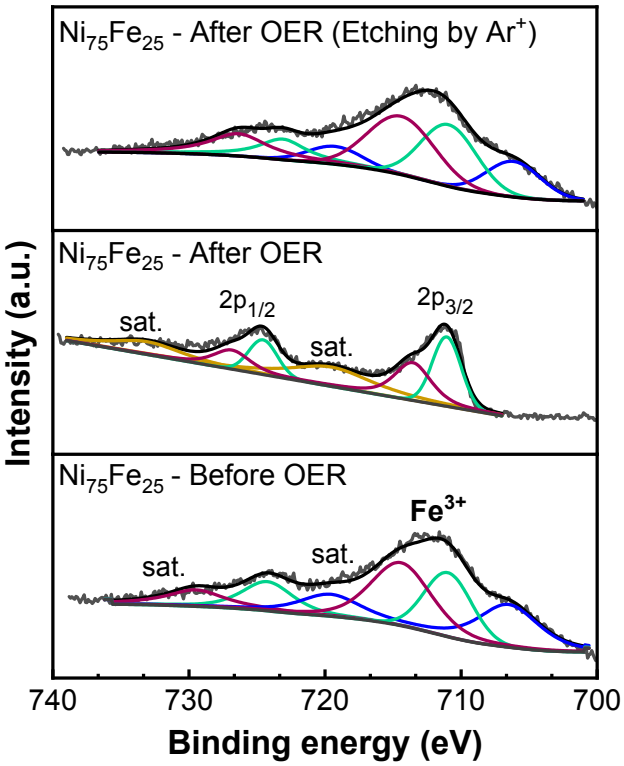

Figure S6 High-resolution XPS spectra of Fe 2p in $Ni_{75}Fe_{25}$ film before and after OER reaction

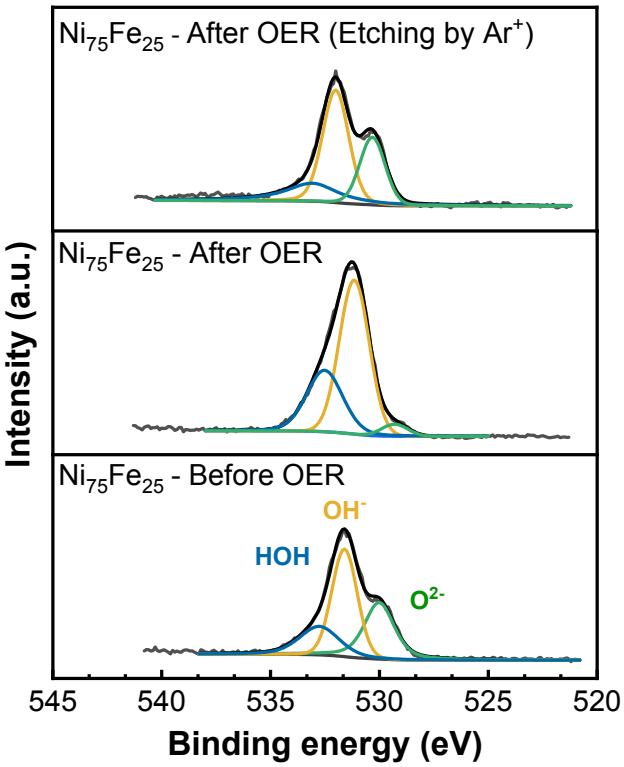

Figure S7 High-resolution XPS spectra of O 1s in $Ni_{75}Fe_{25}$ film before and after OER reaction