Supplementary Data

In-situ desalination-coupled electrolysis with concurrent one-step-synthesis

of value-added chemicals

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Table S1. Elemental (metal) fractions of NiFe-LDH and NiMo catalysts estimated by XPS, SEM-EDS, and STEM.

Sample	Element	Elemental fraction (%)		
		XPS	SEM-EDS	STEM
As-deposited NiFe-LDH	Ni	73.6	31.2	68.4
	Fe	26.4	68.8	31.6
Post-anodized NiFe-LDH	Ni	66.9	34.3	70.2
	Fe	33.1	65.7	29.8
NiMo	Ni	81.7	89.6	-
	Mo	18.3	10.4	-

Table S2. Performance comparison of water splitting catalysts in literature.

Catalysts (anode || cathode)	Support (anode || cathode)	Conditions (anolyte || membrane || catholyte)	E_{cell} (V)	Durability (h)	Reference
NiFe-LDH || NiMo	**Ni foam || Ni foam**	**1 M KOH || BPM || 1 M H$_2$SO$_4$**	**1.57 @10 mA cm^{-2}**	**20**	**This work**
NiFe-LDH || CoP	Ni foam || Ti foil	1 M KOH || BPM || 0.5 M H$_2$SO$_4$	1.63 @10 mA cm^{-2}	90	[1]
Co-Ni-P || Co-Ni-P	Ni foam || Ni foam	1 M NaOH || BPM || 0.5 M H$_2$SO$_4$	1.55 @10 mA cm^{-2}	25	[2]
CoP-CoTe$_2$ || CoP-CoTe$_2$	Carbon paper || carbon paper	1 M KOH || BPM || 0.5 M H$_2$SO$_4$	1.72 @10 mA cm^{-2}	100	[3]
LSC/K-MoSe$_2$ || LSC/K-MoSe$_2$	Ni foam || Ni foam	1 M KOH	1.59 @10 mA cm^{-2}	- (LSV)	[4]
NiFe-LDH@NiCoP || NiFe-LDH@NiCoP	Ni foam || Ni foam	1 M KOH	1.57 @10 mA cm^{-2}	100	[5]
Ni$_2$P@NiFe hydroxide || Ni$_2$P@NiFe hydroxide	Ni foam || Ni foam	1 M KOH	1.51 @10 mA cm^{-2}	100	[6]
Ni/NiS/NC || Ni/NiS/NC	Glassy carbon || glassy carbon	1 M KOH	1.61 @10 mA cm^{-2}	25	[7]
MoS$_2$@Ni$_{0.96}$S || MoS$_2$@Ni$_{0.96}$S	Ni foam || Ni foam	1 M KOH	1.68 @10 mA cm^{-2}	15	[8]
RuTe$_2$ || RuTe$_2$	Glassy carbon || glassy carbon	1 M KOH	1.57 @10 mA cm^{-2}	20	[9]
RuCu || RuCu	Glassy carbon || glassy carbon	1 M KOH	1.50 @10 mA cm^{-2}	12	[10]
NiSe$_2$-Ni$_2$P || NiSe$_2$-Ni$_2$P	Ni foam || Ni foam	1 M KOH	1.50 @10 mA cm^{-2}	- (LSV)	[11]

$NiCo_2O_4@FeOOH$ || $NiCo_2O_4@FeOOH$	Ni foam || Ni foam	1 M KOH	1.52 @10 mA cm^{-2}	- (LSV)	[12]
$NiCo_2O_4$ || $NiCo_2O_4$	Ni foam || Ni foam	1 M NaOH	1.65 @10 mA cm^{-2}	32 h	[13]
$FeWO_4$-$Ni_3S_2@C$ || $FeWO_4$-$Ni_3S_2@C$	Ni foam || Ni foam	1 M KOH	1.51 @10 mA cm^{-2}	- (LSV)	[14]
Fe_2O_3/CuO || Fe_2O_3/CuO	Ni foam || Ni foam	1 M KOH	1.49 @10 mA cm^{-2}	10 h	[15]

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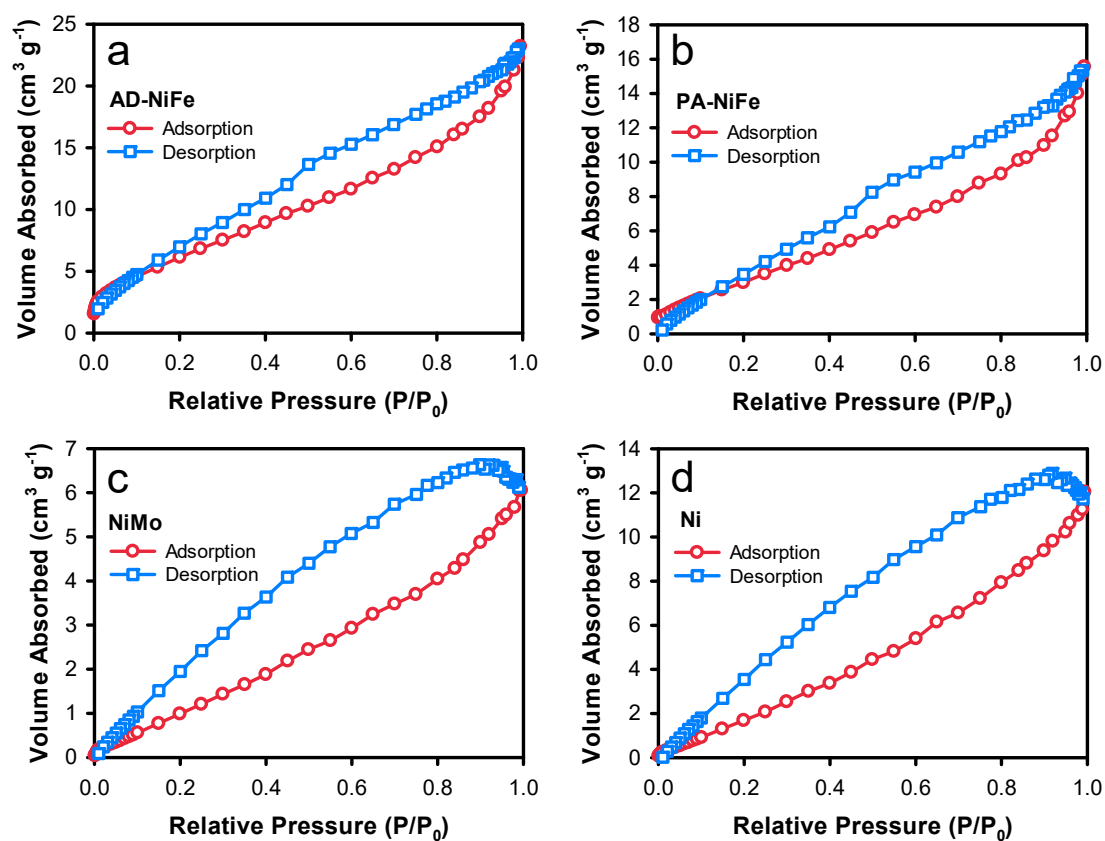
Figure S1. N_2 adsorption and desorption isotherms of (a) as-deposited NiFe-LDH (AD-NiFe), (b) post-anodized NiFe-LDH (PA-NiFe), (c) NiMo, and (d) Ni substrate.
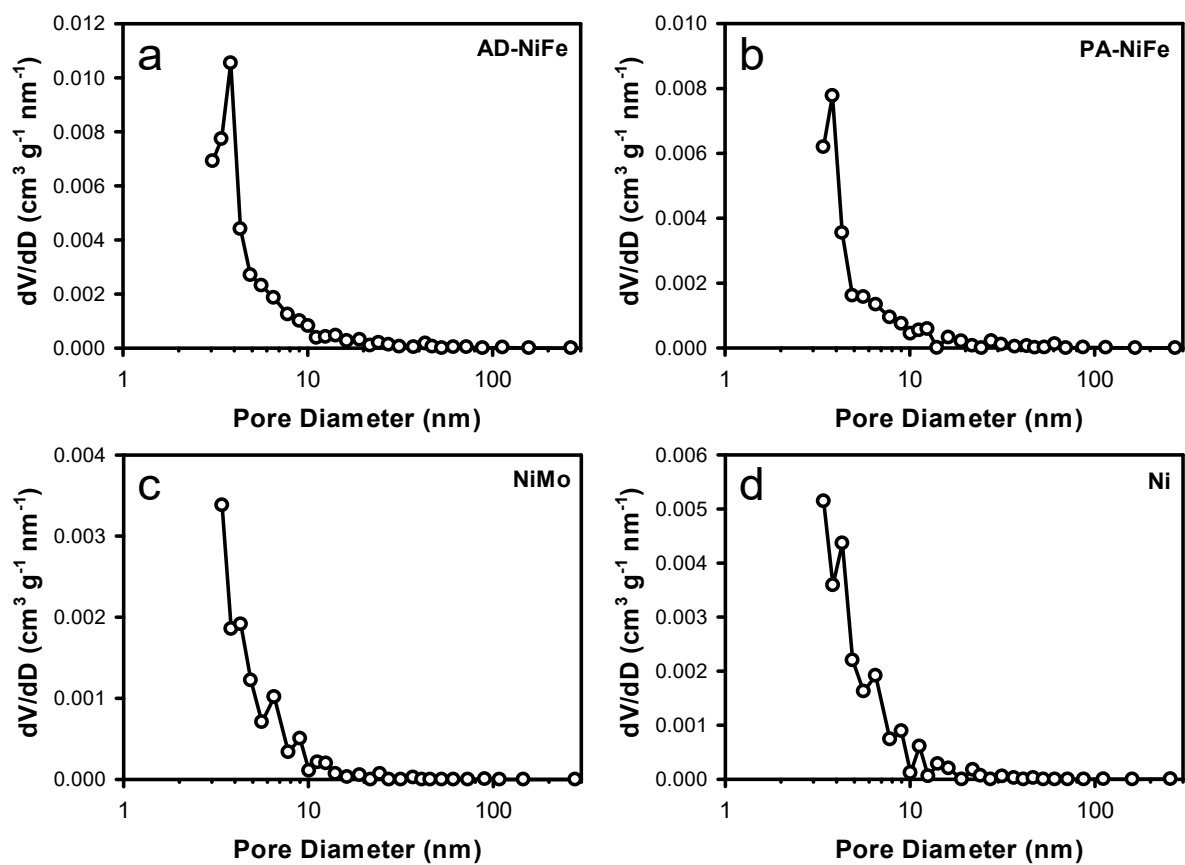
Figure S2. Pore diameter distributions of (a) as-deposited NiFe (AD-NiFe), (b) post-anodized NiFe (PA-NiFe), (c) NiMo, and (d) Ni substrate.
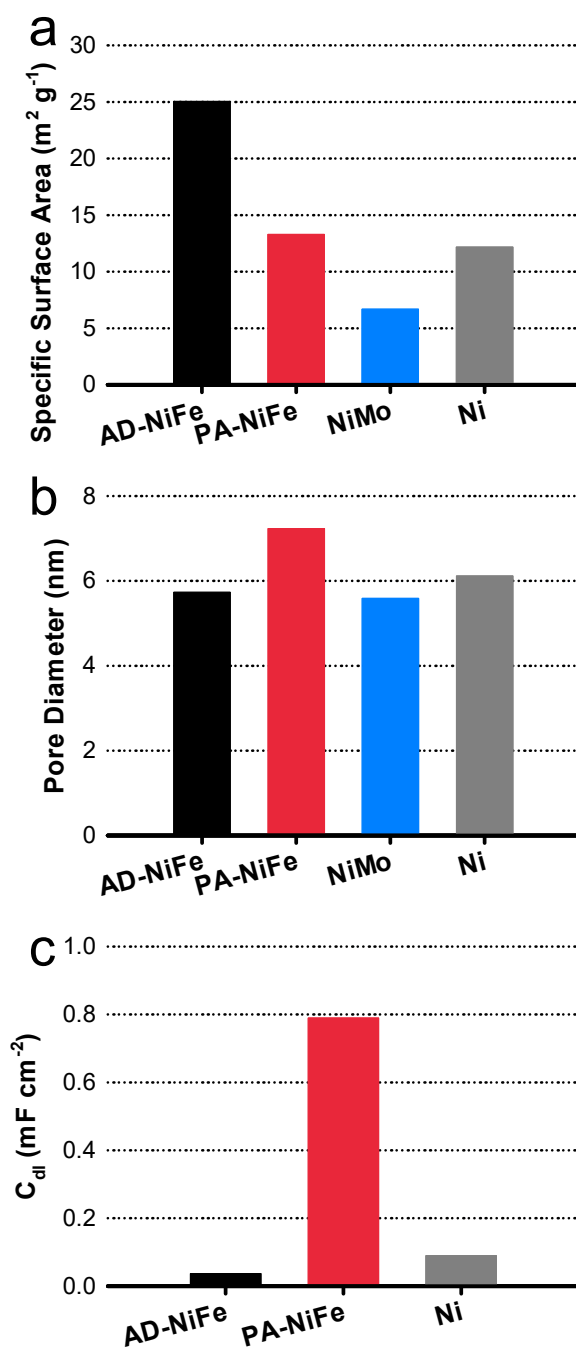
Figure S3. (a) Specific surface areas (BET method), (b) average pore diameters (BJH method), and (c) electrochemical double-layer capacitance (C_{dl}) of as-deposited NiFe-LDH (AD-NiFe), post-anodized NiFe (PA-NiFe), NiMo, and Ni substrate. Electrolytes for NiFe and Ni substrate: 1 M KOH. Electrolyte for NiMo: 1 M H_2SO_4. C_{dl} of NiMo was not obtained due to high Faradaic currents in the acidic solution.

Figure S4. Linear sweep voltammograms of NiFe-LDH electrodes deposited at various Ni/Fe ratios and deposition time.

Figure S5. η values with NiFe-LDH electrodes for OER at (a) $J = 10$ mA cm^{-2} and (b) $J = 100$ mA cm^{-2} (η_{10} and η_{100}, respectively).
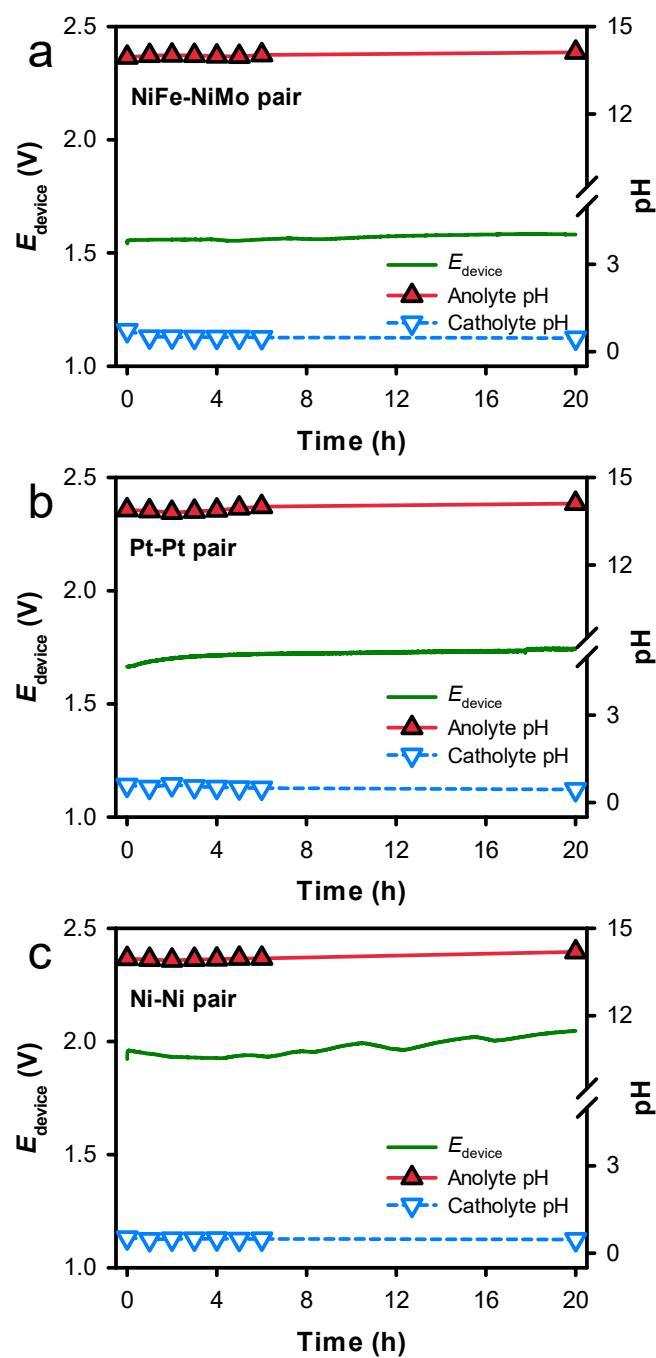
Figure S6. Changes in E_{device} and electrolyte pH values with time at $J = 10$ mA cm^{-2} in two-cell devices divided by BPMs with anolyte of 1 M KOH and catholyte of 1 M H$_2$SO$_4$. (a) NiFe-LDH anode and NiMo cathode pair, (b) Pt anode-Pt cathode pair, and (c) Ni anode-Ni cathode pair.
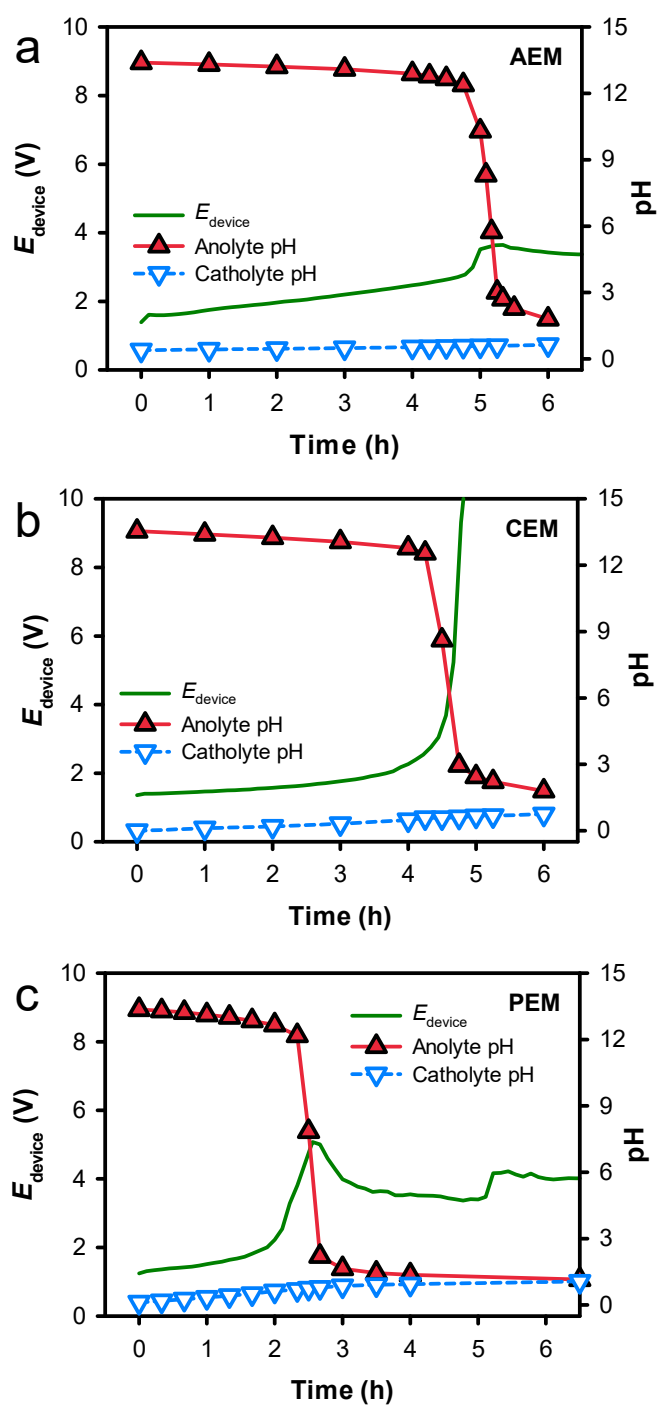
Figure S7. Changes in E_{device} and electrolyte pH values with NiFe-LDH anode and NiMo cathode pairs in two-cell devices divided by (a) an AEM, (b) a CEM, and (c) a PEM with anolyte of 1 M KOH and catholyte of 1 M H_2SO_4 at $J = 100$ mA cm^{-2}.
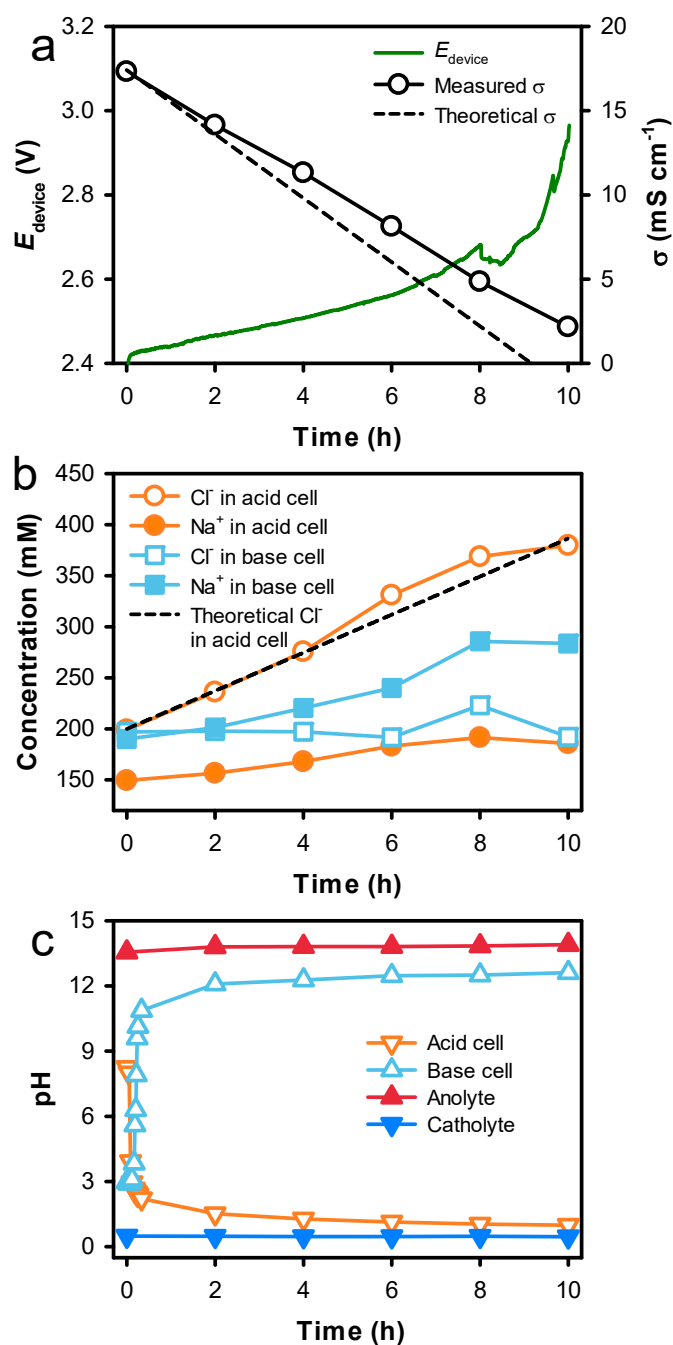
Figure S8. A desalination-coupled electrocatalytic unit device with NiFe-LDH anode and NiMo cathode at $J = 10$ mA cm^{-2} (Case Study II in Table 1). The device configuration is the same as that of Case Study I, except for acid cell (0.2 M NaCl) and base cell (0.2 M NaCl). For the device construction, see Scheme 1b. (a) Changes in E_{device} and ionic conductivity (σ) of saline water with electrolysis time. (b) Changes in concentrations of desalted ions (Cl$^-$ and Na$^+$). (c) Changes in pH values in the solutions in the acid and base cells, and electrolytes.
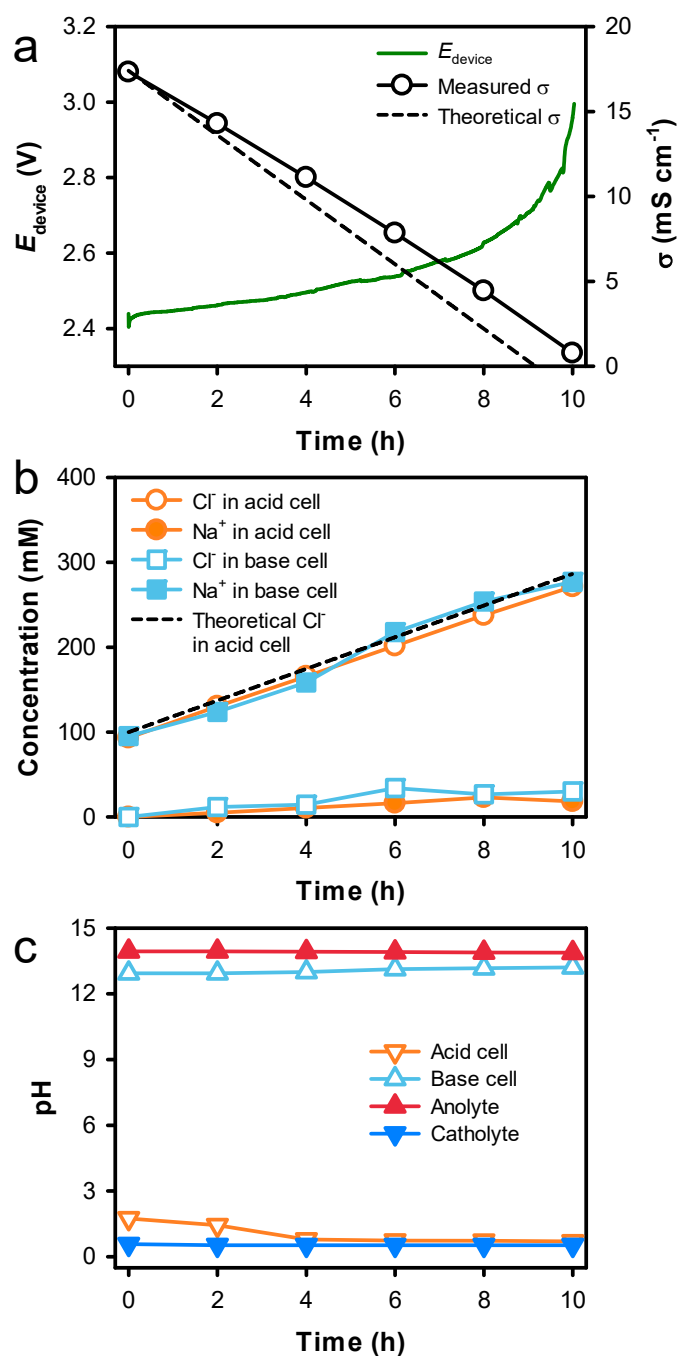
Figure S9. A desalination-coupled electrocatalytic unit device with NiFe-LDH anode and NiMo cathode at $J = 10$ mA cm^{-2} (Case Study III in Table 1). The device configuration is the same as that of Case Study I, except for acid cell (0.1 M HCl) and base cell (0.1 M NaOH). For the device construction, see Scheme 1b. (a) Changes in E_{device} and ionic conductivity (σ) of saline water with electrolysis time. (b) Changes in concentrations of desalted ions (Cl$^-$ and Na$^+$). (c) Changes in pH values in the solutions in the acid and base cells, and electrolytes.
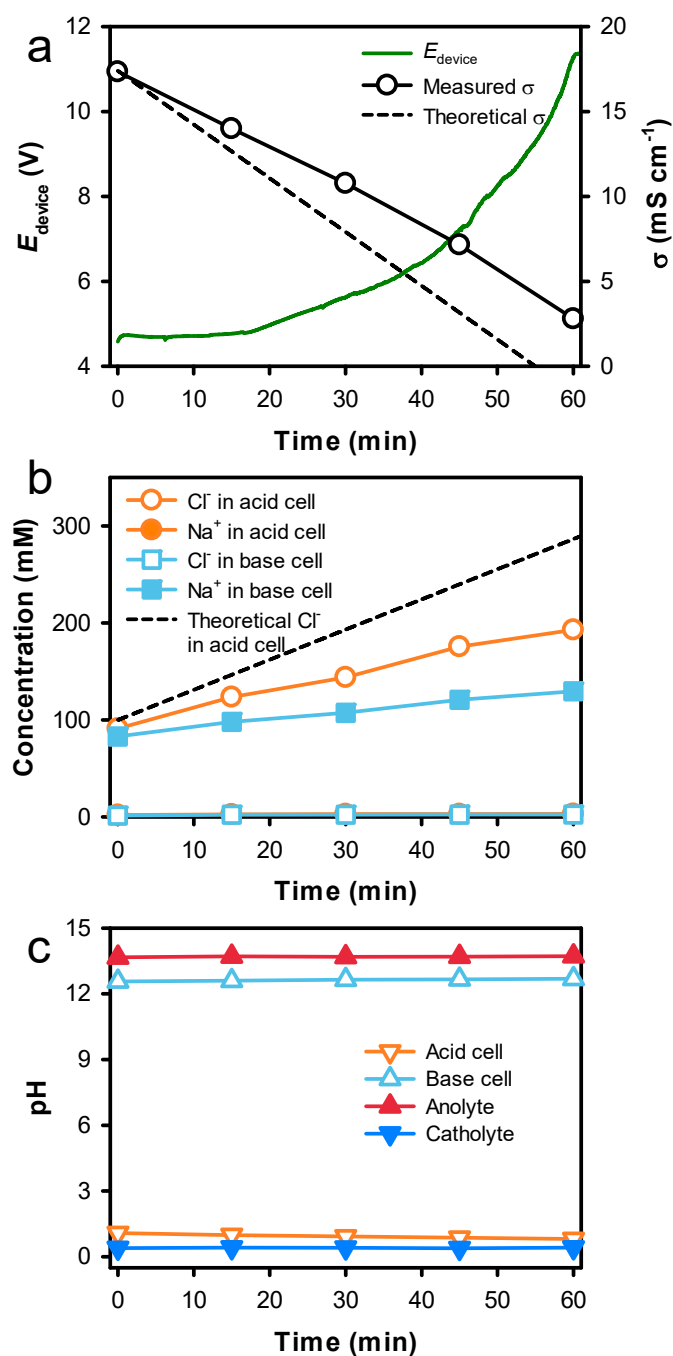
Figure S10. A desalination-coupled electrocatalytic unit device with NiFe-LDH anode and NiMo cathode at J = 100 mA cm^{-2} (Case Study IV in Table 1). The other conditions are the same as those in Figure S9. (a) Changes in E_{device} and ionic conductivity (σ) of saline water with electrolysis time. (b) Changes in concentrations of desalted ions (Cl$^-$ and Na$^+$). (c) Changes in pH values in the solutions in the acid and base cells, and electrolytes.
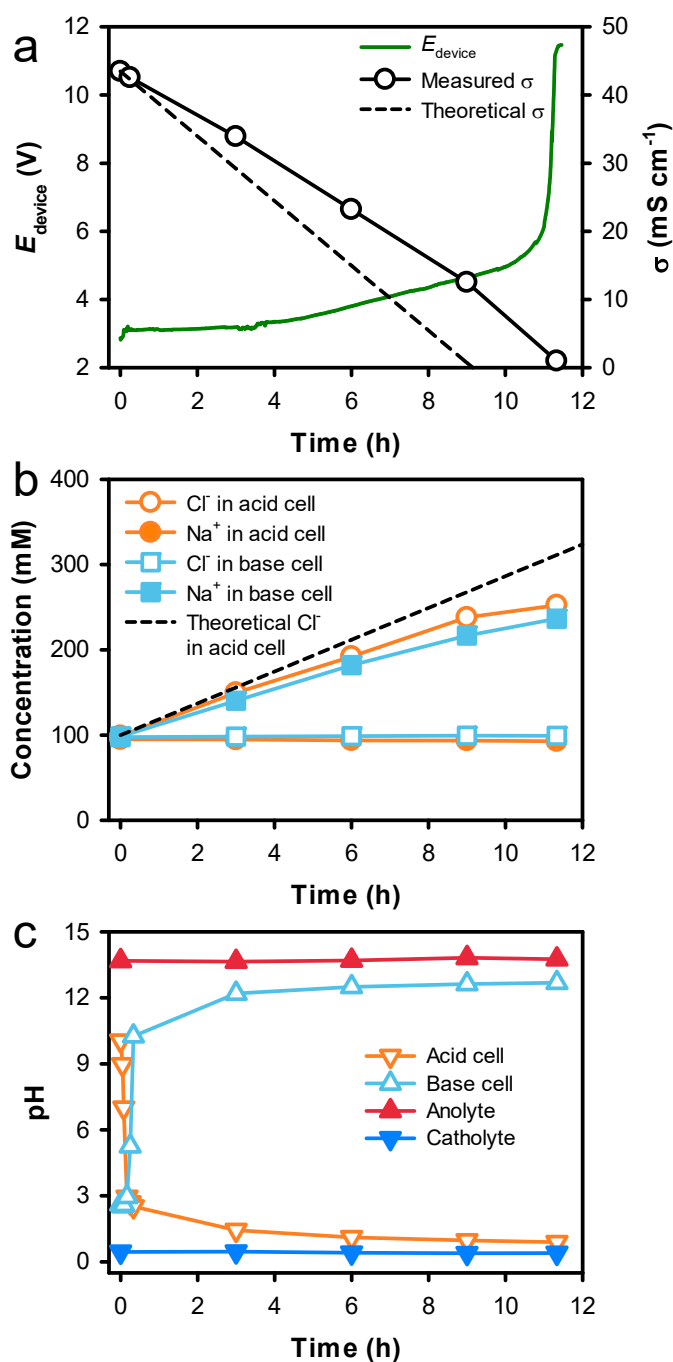
Figure S11. A five-desalination cell array-coupled electrocatalysis with desalination cell (seawater, salinity 36 g L^{-1}). For device configuration and conditions, refer to Case Study VI in Table 1. (a) Changes in E_{device} and ionic conductivity (σ) of saline water with electrolysis time. The dashed line represents the theoretical σ based on J. (b) Changes in concentrations of desalted ions (Cl$^-$ and Na$^+$). The dashed line represents the theoretical Cl$^-$ concentration in the acid cell. (c) Changes in pH values in the solutions in the acid and base cells, and electrolytes.