Supplementary Information (SI) for Sustainable Energy & Fuels.
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Supporting Information

Boosted Hydrogen Production via Binder-Assisted Immobilization of Hydrogen Boride Sheets on Cathodes

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Contents of Supporting Information

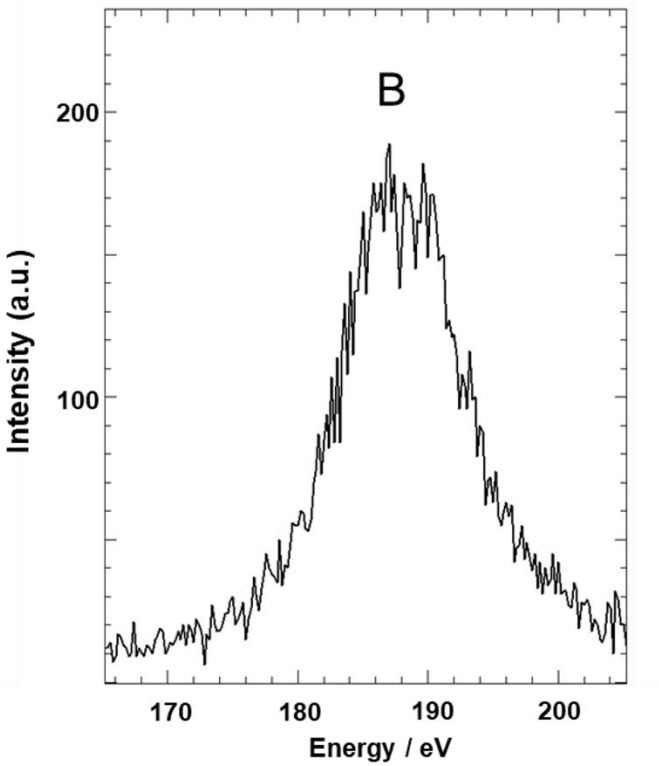

Figure S1. Electron probe microanalyzer (EPMA) spectra of the HB+aramid electrodes with HB/aramid weight ratio of 1/0.25, confirming the presence of boron species on the electrode.

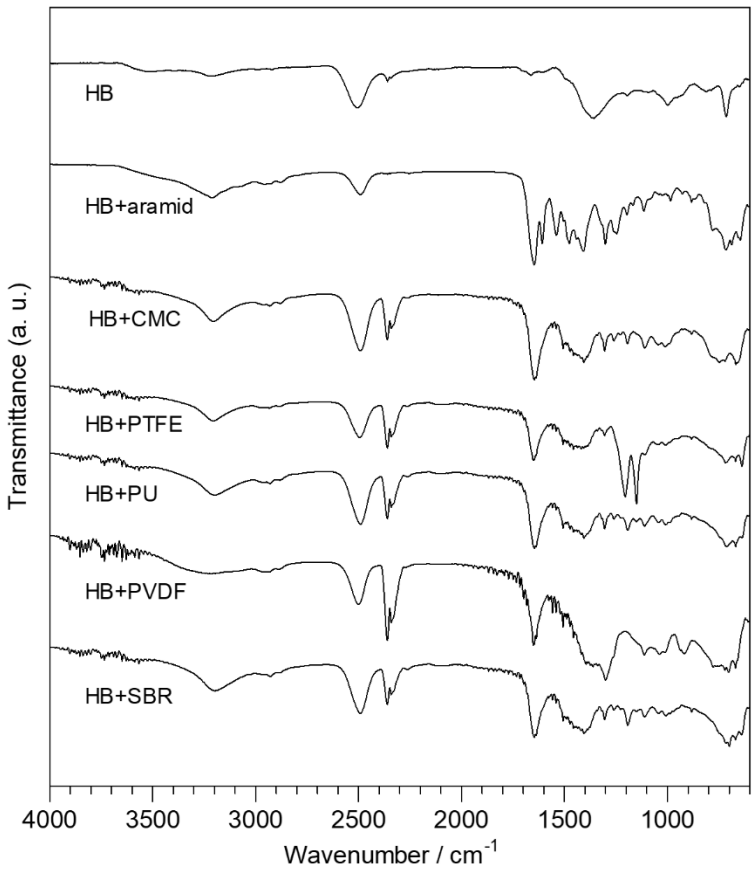

Figure S2. FT–IR spectra of bare HB sheets and HB-immobilized electrodes.

Table S1. Net mass loss of HB sheets that were peeled from the HB+aramid electrodes. The top table shows the mass loss after 5 minutes of soaking. The bottom table shows the mass loss after 5 minutes of electrolysis.

	Aramid/HB ratio (w/w)	HB + binder loss/ mg	Binder loss/ mg	Net HB loss/ mg
Soaking for 5 minutes	0.125	2.5	0.0	2.5
	0.25	1.6	0.0	1.6
	0.5	1.4	0.1	1.3
	1	1.4	0.1	1.3
	2	1.0	0.3	0.7
	5	0.8	0.3	0.5
Electrolysis for 5 minutes	0.125	2.6	0.0	2.6
	0.25	1.8	0.1	1.7
	0.5	1.3	0.0	1.3
	1	1.4	0.1	1.3
	2	1.0	0.3	0.7
	5	0.7	0.3	0.4

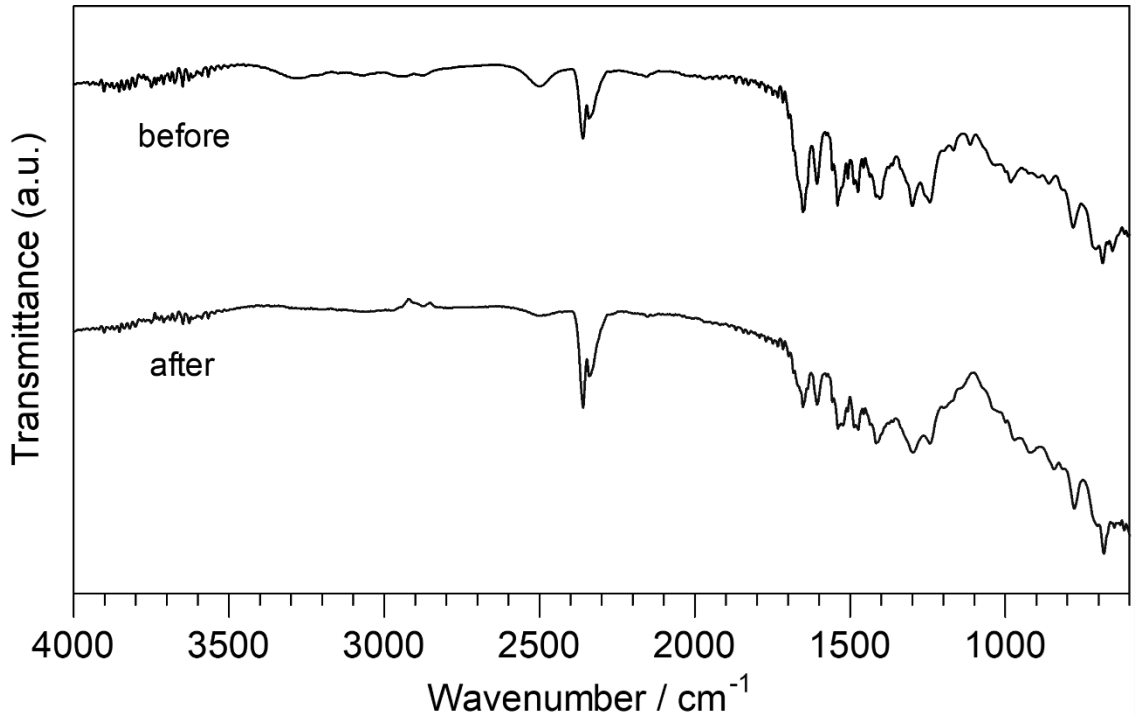

Figure S3. FT-IR spectra of the HB+aramid+graphene electrode before and after the electrolysis, where HB/aramid/graphene ratio is 1/2/1. The signal of B-H bond (2500 cm^{-1}) was reduced after the electrolytic hydrogen production.

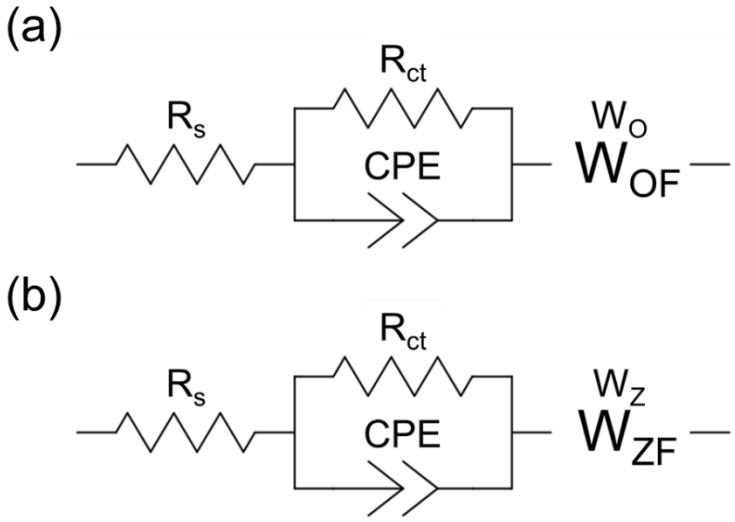

Figure S4. Equivalent circuits used for the fitting of (a) the system using HB+aramid+graphene electrode, (b) the system using dispersed HB sheets and without them.

Table S2. Impedance of circuit elements.

Name	Symbol	Impedance (Ω)
Solution resistance	R_s	$Z = R_s$
Charge transfer resistance	R_{ct}	$Z = R_{ct}$
Constant phase element	CPE	$Z = \frac{1}{T(j\omega)^p}$
Warburg element (semi-infinite)	W_Z	$Z = \frac{(1-j)\sigma}{\sqrt{\omega}}$
Warburg element (open)	W_O	$Z = R_N \frac{\coth(jwT)^p}{(j\omega T)^p}$

Table S3. Each parameter of circuit elements obtained from the fittings.

(a) The system using the HB+aramid+graphene electrode.

Circuit element	Parameters
R_s	$R_s = 37.8\ \Omega$
R_{ct}	$R_{ct} = 65.8\ \Omega$
CPE	$p = 0.661$
	$T = 2.16 \times 10^{-4}\ \mathrm{F\ s^{-0.339}}$
W_O	$R_N = 0.840\ \Omega$
	$p = 0.331$
	$T = 1.17\ \times 10^{-3}\ \mathrm{s}$

(b) The system using the aramid+graphene electrode with dispersed HB sheets.

Circuit element	Parameters
R_s	$R_s = 42.1\ \Omega$
R_{ct}	$R_{ct} = 5.20 \times 10^2\ \Omega$
CPE	$p = 0.737$
	$T = 5.36 \times 10^{-5}\ \mathrm{F\ s^{-0.263}}$
W_Z	$\sigma = 4.32 \times 10^2\ \Omega\ \mathrm{s^{-1/2}}$

(c) The system using the aramid+graphene electrode without dispersed HB sheets.

Circuit element	Parameters
R_s	$R_s = 38.9\ \Omega$
R_{ct}	$R_{ct} = 7.05 \times 10^2\ \Omega$
CPE	$p = 0.746$
	$T = 6.94 \times 10^{-5}\ \mathrm{F\ s^{-0.254}}$
W_Z	$\sigma = 3.87 \times 10^2\ \Omega\ \mathrm{s^{-1/2}}$