

## Electronic Supplementary Information

### Borate-Deficient Decavanadoborate Additives Boost Nafion Membrane Performance: Enhanced Proton Conductivity and Power Density in Humidified PEMFCs

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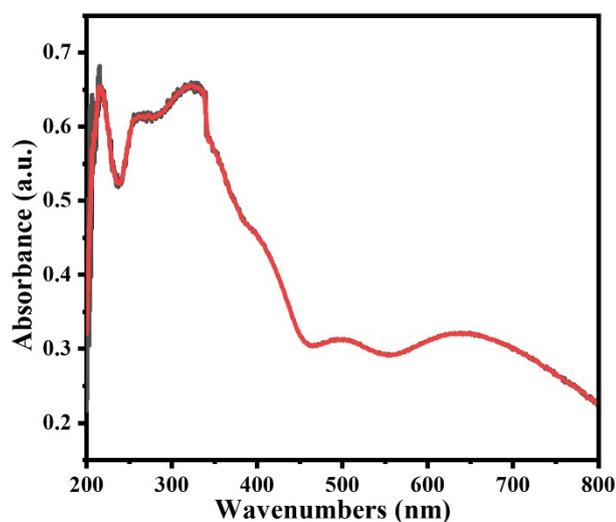


Fig. S1. UV-vis spectrum of VB-7.

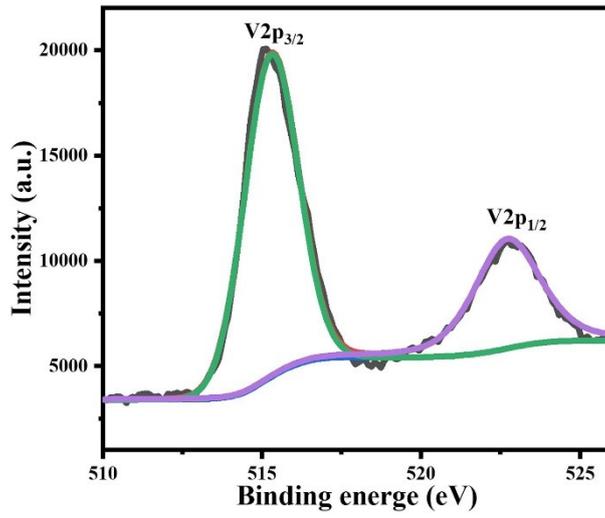


Fig. S2. XPS spectrum of vanadium in VB-7.

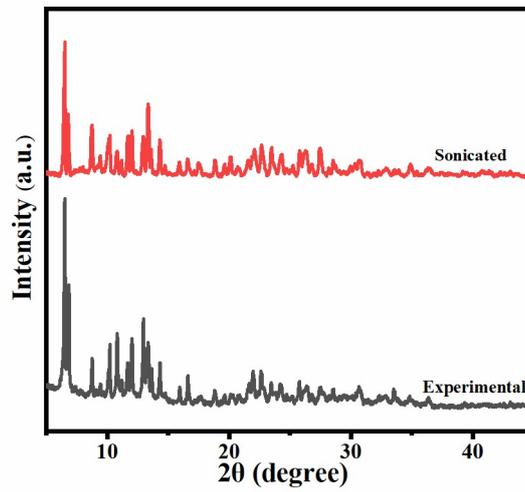


Fig. S3. PXRD of VB after and before ultrasonic treatment.

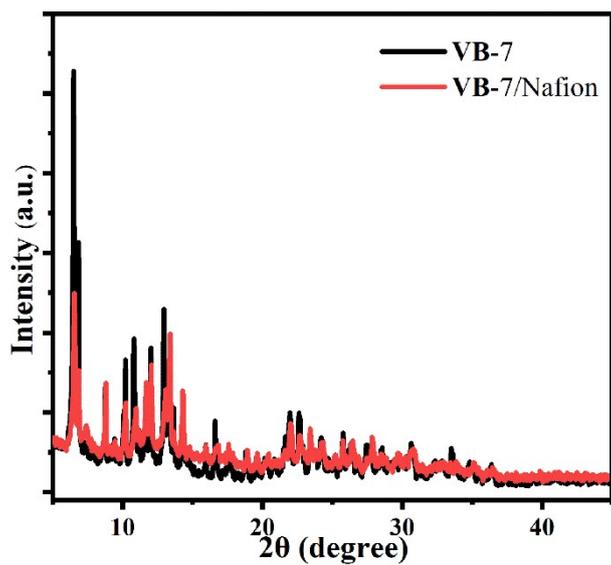


Fig. S4. A comparison of the peak intensities of PXR D patterns of VB-7 in experimental and Nafion-composited conditions.

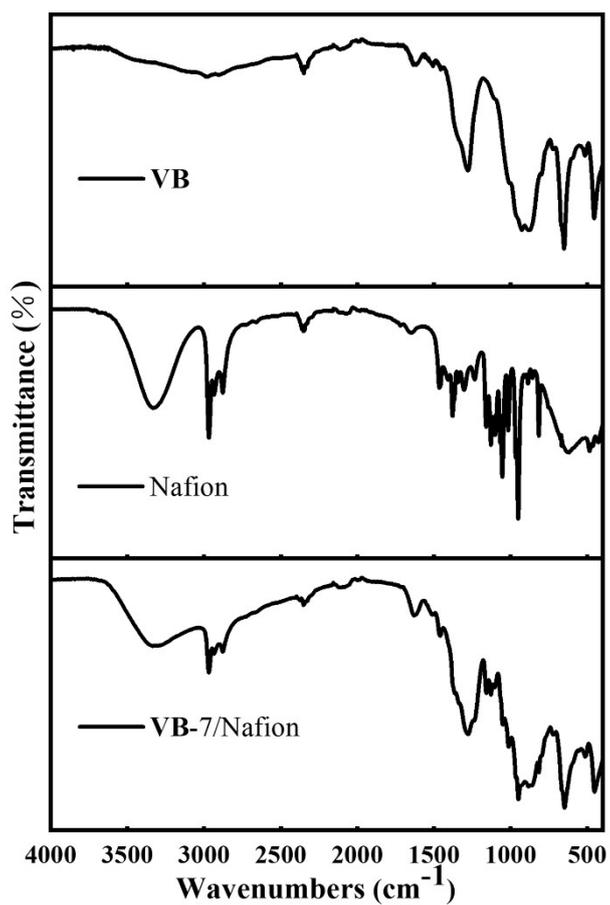
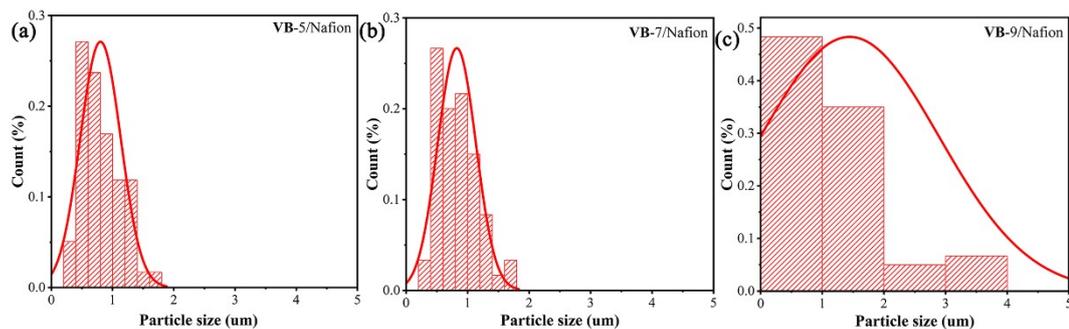
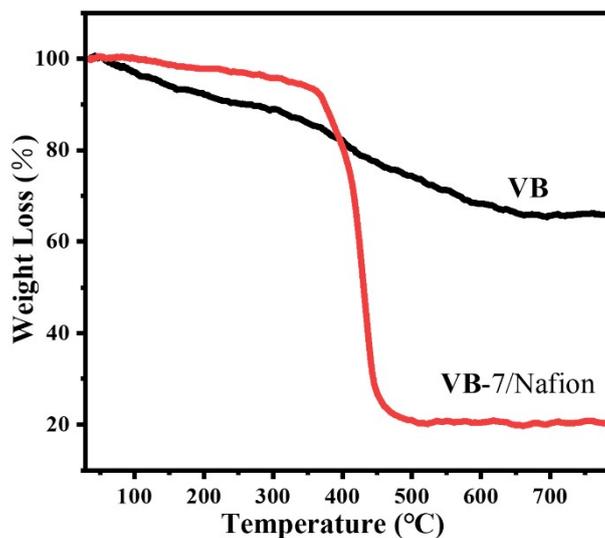


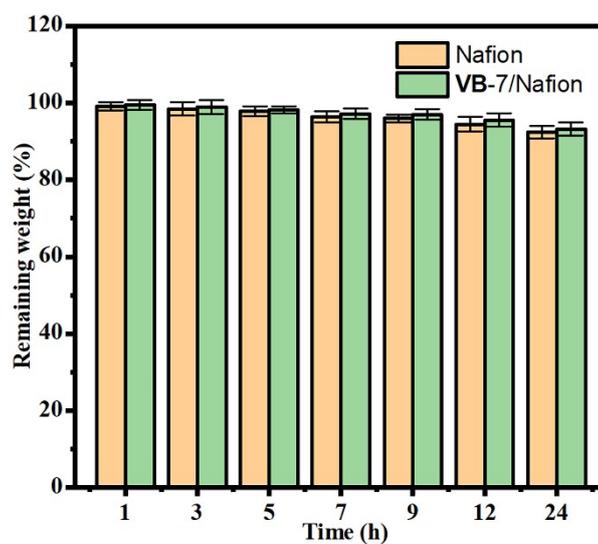
Fig. S5. FT-IR spectra of VB, pure Nafion and VB-7/Nafion composite membranes.



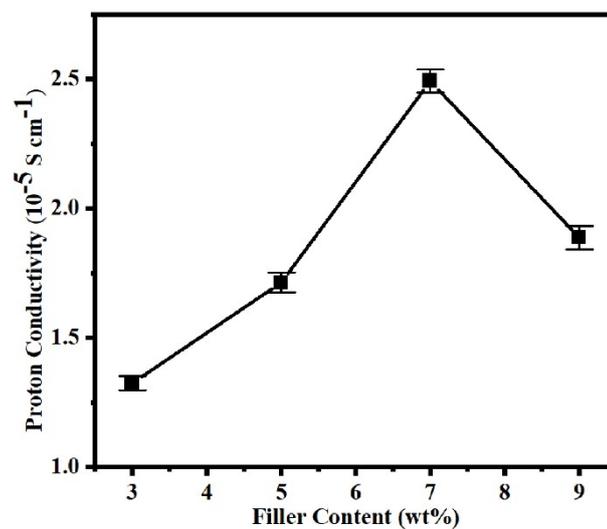
**Fig. S6.** Particle size distribution of VB in Nafion-based composite membranes: (a) VB-5/Nafion, (b) VB-7/Nafion, (c) VB-9/Nafion.



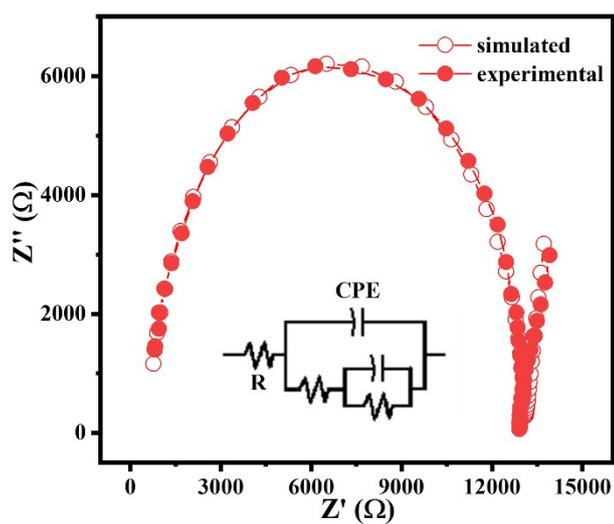
**Fig. S7.** TGA curves of VB and VB-7/Nafion in air.



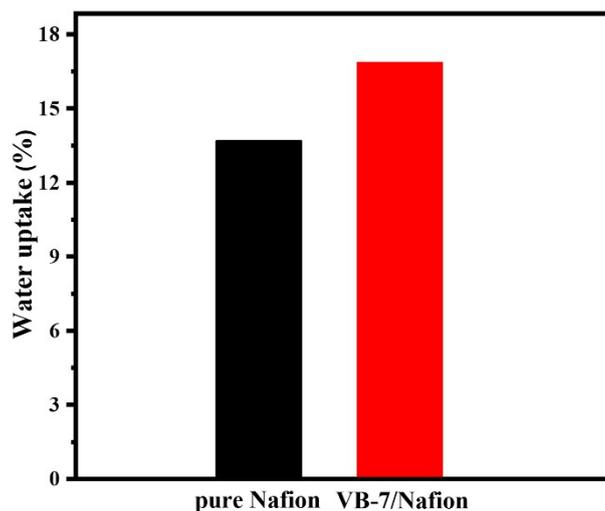
**Fig. S8.** Remaining weight of Nafion and VB-7/Nafion membranes in Fenton's reagent at Fenton's test at 80 °C for 24 h.



**Fig. S9.** Proton conductivity of VB-x/Nafion composite membranes with varying VB weight loadings (3, 5, 7, and 9 wt%) measured at 293 K under 100% relative humidity.



**Fig. S10.** Equivalent circuits used to fit the impedance data.



**Fig. S11.** Water uptake ratio of pure Nafion and VB-7/Nafion membranes in water.

**Table S1.** Bond lengths [Å] for VB

Atom-Atom	Length/Å	Atom-Atom	Length/Å
V1-V1 <sup>1</sup>	3.0601(15)	O9-B4	1.452(8)
V1-V2	2.9240(11)	O10-B4	1.484(7)
V1-O1	1.606(4)	O12-B4	1.464(7)
V1-O2	1.981(3)	O12-B5	1.331(7)
V1-O4	1.918(3)	O13-B5	1.384(7)
V1-O16 <sup>2</sup>	1.929(3)	O14-B5	1.350(7)
V1-O7 <sup>3</sup>	1.981(3)	O14-B6	1.456(7)
V2-V3	3.0155(8)	O15-B6	1.450(7)
V2-O3	1.610(4)	O15-B7	1.350(6)
V2-O4	1.928(3)	O16-B6	1.474(6)
V2-O10	1.947(3)	O17-B6	1.447(6)
V2-O13 <sup>2</sup>	1.983(3)	O7-B7	1.384(11)
V2-O16 <sup>2</sup>	1.948(3)	N1-C1	1.492(13)
V3-O10	1.923(3)	N2-C2	1.68(2)
V3-O10 <sup>2</sup>	1.923(3)	N2-C3	1.479(16)
V3-O11	1.609(5)	N3-C4 <sup>1</sup>	1.527(17)
V3-O13 <sup>2</sup>	1.963(3)	N3-C4	1.527(17)
V3-O13	1.963(3)	C1-C2	1.58(2)
O2-B1	1.391(10)	C3-C3 <sup>1</sup>	1.09(3)
O4-B2	1.484(7)	C3-C4	1.68(2)
O5-B1	1.354(6)	C4-C4 <sup>1</sup>	1.28(5)
O5-B2	1.462(7)	N4-C5	1.506(10)
O19-B2	1.479(7)	N5-C6	1.505(10)
O19-B3	1.344(7)	N5-C6 <sup>4</sup>	1.505(10)

O20-B3	1.376(7)	O6-O6 <sup>4</sup>	1.39(4)
O8-B3	1.363(7)	C5-C6	1.494(10)
O8-B4	1.489(7)	O22-O22 <sup>3</sup>	1.26(6)
O9-B2	1.464(8)		

**Table S2.** Bond angles [deg] for **VB**

Atom-Atom-Atom	Angle/°	Atom-Atom-Atom	Angle/°
V2-V1-V1 <sup>1</sup>	143.80(2)	V1-O4-V2	98.96(15)
O1-V1-V1 <sup>1</sup>	105.35(15)	B2-O4-V1	132.8(3)
O1-V1-V2	110.85(15)	B2-O4-V2	127.8(3)
O1-V1-O2	108.5(2)	B1-O5-B2	128.0(5)
O1-V1-O4	110.8(2)	B3-O19-B2	119.7(4)
O1-V1-O16 <sup>2</sup>	107.39(18)	B3-O8-B4	119.7(4)
O1-V1-O7 <sup>3</sup>	109.8(2)	B4-O9-B2	115.6(4)
O2-V1-V1 <sup>1</sup>	39.42(11)	V3-O10-V2	102.36(15)
O2-V1-V2	123.38(14)	B4-O10-V2	125.8(3)
O2-V1-O7 <sup>3</sup>	76.92(15)	B4-O10-V3	131.8(3)
O4-V1-V1 <sup>1</sup>	123.67(11)	B5-O12-B4	130.5(4)
O4-V1-V2	40.65(10)	V3-O13-V2 <sup>2</sup>	99.67(14)
O4-V1-O2	87.82(16)	B5-O13-V2 <sup>2</sup>	129.4(3)
O4-V1-O16 <sup>2</sup>	81.48(14)	B5-O13-V3	128.1(3)
O4-V1-O7 <sup>3</sup>	139.35(18)	B5-O14-B6	129.7(4)
O16 <sup>2</sup> -V1-V1 <sup>1</sup>	126.24(10)	B7-O15-B6	133.1(5)
O16 <sup>2</sup> -V1-V2	41.28(9)	V1 <sup>2</sup> -O16-V2 <sup>2</sup>	97.92(14)
O16 <sup>2</sup> -V1-O2	144.06(19)	B6-O16-V1 <sup>2</sup>	130.1(3)
O16 <sup>2</sup> -V1-O7 <sup>3</sup>	89.18(15)	B6-O16-V2 <sup>2</sup>	131.7(3)
O7 <sup>3</sup> -V1-V1 <sup>1</sup>	39.43(11)	V1 <sup>2</sup> -O7-V1 <sup>3</sup>	101.1(2)
O7 <sup>3</sup> -V1-V2	123.04(13)	B7-O7-V1 <sup>2</sup>	128.24(15)
V1-V2-V3	143.19(4)	B7-O7-V1 <sup>3</sup>	128.24(15)
O3-V2-V1	110.06(15)	C3-N2-C2	129.1(12)
O3-V2-V3	106.75(15)	C4 <sup>1</sup> -N3-C4	49.6(19)
O3-V2-O4	108.18(19)	N1-C1-C2	110.8(9)
O3-V2-O10	109.51(18)	C1-C2-N2	119.6(11)
O3-V2-O13 <sup>2</sup>	108.99(19)	N2-C3-C4	97.0(12)
O3-V2-O16 <sup>2</sup>	108.97(18)	C3 <sup>1</sup> -C3-N2	169.7(7)
O4-V2-V1	40.39(10)	C3 <sup>1</sup> -C3-C4	93.3(9)
O4-V2-V3	125.48(11)	N3-C4-C3	93.9(12)
O4-V2-O10	90.22(15)	C4 <sup>1</sup> -C4-N3	65.2(10)
O4-V2-O13 <sup>2</sup>	142.82(16)	C4 <sup>1</sup> -C4-C3	86.7(9)
O4-V2-O16 <sup>2</sup>	80.75(13)	O5-B1-O2	119.8(4)
O10-V2-V1	124.14(11)	O5 <sup>1</sup> -B1-O2	119.8(4)
O10-V2-V3	38.53(10)	O5 <sup>1</sup> -B1-O5	120.3(7)

O10-V2-O13 <sup>2</sup>	77.00(14)	O5-B2-O4	112.1(4)
O10-V2-O16 <sup>2</sup>	141.40(14)	O5-B2-O19	108.1(5)
O13 <sup>2</sup> -V2-V1	123.07(10)	O5-B2-O9	109.5(5)
O13 <sup>2</sup> -V2-V3	39.92(9)	O19-B2-O4	108.0(5)
O16 <sup>2</sup> -V2-V1	40.80(9)	O9-B2-O4	109.0(4)
O16 <sup>2</sup> -V2-V3	124.15(10)	O9-B2-O19	110.1(4)
O16 <sup>2</sup> -V2-O13 <sup>2</sup>	87.83(14)	O19-B3-O20	117.6(5)
V2-V3-V2 <sup>2</sup>	145.90(5)	O19-B3-O8	123.7(5)
O10-V3-V2 <sup>2</sup>	124.39(10)	O8-B3-O20	118.6(5)
O10-V3-V2	39.11(10)	O9-B4-O8	109.2(4)
O10 <sup>2</sup> -V3-V2	124.39(10)	O9-B4-O10	110.6(4)
O10 <sup>2</sup> -V3-V2 <sup>2</sup>	39.11(10)	O9-B4-O12	109.2(5)
O10-V3-O10 <sup>2</sup>	137.8(2)	O10-B4-O8	108.8(5)
O10 <sup>2</sup> -V3-O13	78.04(13)	O12-B4-O8	106.8(4)
O10 <sup>2</sup> -V3-O13 <sup>2</sup>	89.29(14)	O12-B4-O10	112.1(4)
O10-V3-O13 <sup>2</sup>	78.04(13)	O12-B5-O13	122.0(5)
O10-V3-O13	89.29(14)	O12-B5-O14	117.4(5)
O11-V3-V2 <sup>2</sup>	107.05(2)	O14-B5-O13	120.6(5)
O11-V3-V2	107.05(2)	O14-B6-O16	113.0(4)
O11-V3-O10	111.11(10)	O15-B6-O14	106.4(5)
O11-V3-O10 <sup>2</sup>	111.11(10)	O15-B6-O16	110.5(4)
O11-V3-O13	107.75(11)	O17-B6-O14	107.7(4)
O11-V3-O13 <sup>2</sup>	107.75(11)	O17-B6-O15	109.9(5)
O13 <sup>2</sup> -V3-V2	40.41(10)	O17-B6-O16	109.2(4)
O13 <sup>2</sup> -V3-V2 <sup>2</sup>	125.64(11)	O15-B7-O15 <sup>1</sup>	118.1(8)
O13-V3-V2 <sup>2</sup>	40.41(10)	O15-B7-O7	120.9(4)
O13-V3-V2	125.64(11)	O15 <sup>1</sup> -B7-O7	120.9(4)
O13 <sup>2</sup> -V3-O13	144.5(2)	C6-N5-C6 <sup>4</sup>	164(3)
V1 <sup>1</sup> -O2-V1	101.2(2)	C6-C5-N4	112.7(11)
B1-O2-V1 <sup>1</sup>	129.20(12)	C5-C6-N5	111.9(16)
B1-O2-V1	129.20(12)		

Symmetry transformations used to generate equivalent atoms: <sup>1</sup>+X,1-Y,+Z; <sup>2</sup>1-X,+Y,1-Z; <sup>3</sup>1-X,1-Y,1-Z; <sup>4</sup>2-X,+Y,2-Z

**Table S3.** Proton Conductivity ( $\sigma$ ) and power density (P) values of several reported nafion-based composite membranes modified with crystalline porous materials under comparable testing conditions<sup>a</sup>

Membrane	Test temperature (°C)	Proton conductivity			Conditions	Fuel cell performance		Conditions	Ref.
		$\sigma_{\text{pure Nafion}}$ (S cm <sup>-1</sup> )	$\sigma_{\text{composite-membrane}}$ (S cm <sup>-1</sup> )	$\sigma_{\text{composite-membrane}}/\sigma_{\text{pure Nafion}}$		$P_{\text{pure Nafion}}$ (mW cm <sup>-2</sup> )	$P_{\text{composite-membrane}}$ (mW cm <sup>-2</sup> )		
POMs Nafion-(NKFs@PWA)* C <sub>52</sub> S <sub>4</sub> H <sub>0.5</sub> PW <sub>12</sub> O <sub>40</sub> /	< 60	6.8 × 10 <sup>-2</sup>	6.1 × 10 <sup>-2</sup>	0.90	25 °C	-	-	-	1
		3.6 × 10 <sup>-2</sup>	5.3 × 10 <sup>-2</sup>	1.47	25 °C,	266	351	80 °C,	2

	Nafion				92% RH			35% RH	
	Cs <sub>2.5</sub> H <sub>0.5</sub> PMo <sub>12</sub> O <sub>40</sub> /Nafion	$3.6 \times 10^{-2}$	$5.8 \times 10^{-2}$	1.61	25 °C, 92% RH	266	420	80 °C, 35% RH	2
	Nafion-(SN@APTES-PWA)*	$6.7 \times 10^{-2}$	$8.1 \times 10^{-2}$	1.21	25 °C, 100% RH	-	-	-	3
	VB-7/Nafion	$3.82 \times 10^{-5}$	$1.09 \times 10^{-4}$	2.85	70 °C, 100% RH	772	898	80 °C, 100% RH	This work
	GSiW <sub>11</sub> -Nafion	0.189	0.226	1.19	80 °C, 100% RH	287	372	80 °C, 98% RH	4
	Nafion-10%HPW(30)Zr*	$8.23 \times 10^{-3}$	$6.0 \times 10^{-3}$	0.73	120 °C, 25% RH	-	-	-	5
	Nafion-10%HPW(45)Zr*	$8.23 \times 10^{-3}$	$6.8 \times 10^{-3}$	0.83	120 °C, 25% RH	-	-	-	5
	Nafion/PTA*	$1.0 \times 10^{-2}$	$1.5 \times 10^{-2}$	1.5	120 °C, 35% RH	-	-	-	6
	Nafion /PTA*	$1.7 \times 10^{-2}$	$1.6 \times 10^{-2}$	0.94	120 °C, 35% RH	-	-	-	7
	Nafion /PTA/SiO <sub>2</sub> *	$1.3 \times 10^{-2}$	$1.5 \times 10^{-2}$	1.15	120 °C, 35% RH	-	-	-	8
	Nafion/ FBC-21/PW	$5.55 \times 10^{-2}$	$6.8 \times 10^{-2}$	1.23	-	-	-	-	9
	[Cd(H <sub>2</sub> L)(M bpy)] <sub>n</sub> /Nafion	$3.82 \times 10^{-7}$	$4.94 \times 10^{-7}$	1.29	30 °C, 100% RH	-	-	-	10
	[Ni(L) <sub>0.5</sub> (M bpy)(H <sub>2</sub> O) <sub>2</sub> ] <sub>2</sub> /Nafion	$3.82 \times 10^{-7}$	$4.04 \times 10^{-7}$	1.06	30 °C, 100% RH	-	-	-	10
	UiO-66-NH <sub>2</sub> /Nafion-0.6	$4.7 \times 10^{-2}$	$7.7 \times 10^{-2}$	1.64	40 °C	-	-	-	11
	UiO-66-SO <sub>3</sub> H/Nafion-0.6	$4.7 \times 10^{-2}$	$1.04 \times 10^{-1}$	2.21	40 °C	-	-	-	11
	UiO-66-NH <sub>2</sub> + UiO-66-SO <sub>3</sub> H/Nafion-0.6	$4.7 \times 10^{-2}$	$1.34 \times 10^{-1}$	2.85	40 °C	-	-	-	11
	{[Mn <sub>3</sub> (μ <sub>4</sub> -L) <sub>2</sub> (H <sub>2</sub> O) <sub>7</sub> ]·4H <sub>2</sub> O} <sub>n</sub> /Nafion	$4.04 \times 10^{-7}$	$5.97 \times 10^{-7}$	1.48	30 °C, 100% RH	-	-	-	12
	{[Mn <sub>3</sub> (μ <sub>5</sub> -L) <sub>2</sub> (H <sub>2</sub> O) <sub>6</sub> ]·4H <sub>2</sub> O} <sub>n</sub> /Nafion	$4.04 \times 10^{-7}$	$5.53 \times 10^{-7}$	1.37	30 °C, 100% RH	-	-	-	12
	{[Mn <sub>3</sub> (μ <sub>7</sub> -L) <sub>2</sub> (H <sub>2</sub> O) <sub>2</sub> ] <sub>n</sub> /Nafion	$4.04 \times 10^{-7}$	$4.90 \times 10^{-7}$	1.21	30 °C, 100% RH	-	-	-	12
	Zn-MOF/Nafion	$3.89 \times 10^{-3}$	$7.29 \times 10^{-3}$	1.87	80 °C, 58% RH	-	-	-	13
	Nafion/phytic@MIL-101	$2.17 \times 10^{-2}$	$6.08 \times 10^{-2}$	2.8	80 °C, 58% RH	-	-	-	14
MOFs	[Ni(H <sub>2</sub> L)(bpyBr) <sub>2</sub> ] <sub>2</sub> ·2H <sub>2</sub> O/Nafion	$3.82 \times 10^{-7}$	$8.89 \times 10^{-7}$	2.32	60 °C, 100% RH	-	-	-	15
	[Ni <sub>2</sub> (H <sub>2</sub> L) <sub>2</sub> (bpyBr) <sub>2</sub> (H <sub>2</sub> O) <sub>3</sub> ] <sub>n</sub> /Nafion	$3.82 \times 10^{-7}$	$7.43 \times 10^{-7}$	1.94	60 °C, 100% RH	-	-	-	15
	Tb-MOF/Nafion-5	$8.48 \times 10^{-3}$	$1.53 \times 10^{-2}$	1.81	80 °C, 100% RH	-	-	-	16
	Nafion/ZIF-8@PCMs	0.165	0.24	1.45	80 °C, 100% RH	-	-	-	17
	{[Cd(L) <sub>1/2</sub> (H <sub>2</sub> O) <sub>2</sub> ]·H <sub>2</sub> O} <sub>n</sub> /Nafion	$5.96 \times 10^{-7}$	$7.36 \times 10^{-7}$	1.23	60 °C, 100% RH	-	-	-	18
	{[Ni(H <sub>2</sub> L)(bpyBr) <sub>2</sub> (H <sub>2</sub> O) <sub>2</sub> ]·H <sub>2</sub> O} <sub>n</sub> /Nafion	$5.96 \times 10^{-7}$	$6.58 \times 10^{-7}$	1.10	60 °C, 100% RH	-	-	-	18
	{[Ni(H <sub>2</sub> L)(bipy)(H <sub>2</sub> O)]·H <sub>2</sub> O} <sub>n</sub> /Nafion	$5.96 \times 10^{-7}$	$7.39 \times 10^{-7}$	1.24	60 °C, 100% RH	-	-	-	18
	[Co(H <sub>2</sub> L)(bipy)(H <sub>2</sub> O) <sub>2</sub> ] <sub>n</sub> /Nafion	$5.96 \times 10^{-7}$	$8.16 \times 10^{-7}$	1.37	60 °C, 100% RH	-	-	-	18
	{[Co(L) <sub>1/2</sub> (H <sub>2</sub> O) <sub>2</sub> ]·H <sub>2</sub> O} <sub>n</sub> /Nafion	$5.96 \times 10^{-7}$	$1.039 \times 10^{-6}$	1.74	60 °C, 100% RH	-	-	-	19
	{Mn(L) <sub>1/2</sub> (H <sub>2</sub> O) <sub>2</sub> ]·	$5.96 \times 10^{-7}$	$7.36 \times 10^{-7}$	1.23	60 °C,	-	-	-	19

	H <sub>2</sub> O <sub>n</sub> /Nafion					100% RH				
	{[Zn(L) <sub>1/2</sub> (H <sub>2</sub> O) <sub>2</sub> ·H <sub>2</sub> O] <sub>n</sub> /Nafion	5.96 × 10 <sup>-7</sup>	6.86 × 10 <sup>-7</sup>	1.15		60 °C, 100% RH	-	-	-	19
	Pb-MOF/Nafion-7	8.48 × 10 <sup>-3</sup>	1.82 × 10 <sup>-2</sup>	2.14		80 °C, 100% RH	-	-	-	20
	MIL-101-SO <sub>3</sub> H/Nafion	> 80	0.15	0.19	1.27	105 °C, 100% RH	-	-	-	21
	Nafion/Zr-MOF-808	Unkno wn	2.29 × 10 <sup>-3</sup>	2.96 × 10 <sup>-3</sup>	1.29	35% RH	-	-	-	22
	Nafion/P-COF	< 60	0.098	0.141	1.44	25 °C, 100% RH	525	724	80 °C, 50% RH	23
	Nafion/Z-COF		0.22	0.14	1.571	80 °C, 100% RH	-	-	-	24
COFs	fibrous COF(TfBD)@Nafion/GO	60 ~ 80	0.30	0.13	2.31	80 °C, 95% RH	-	-	-	25
	(CN <sub>3</sub> H <sub>6</sub> ) <sub>2</sub> (C <sub>10</sub> O <sub>8</sub> H <sub>4</sub> )/Nafion		1.33 × 10 <sup>-1</sup>	9.62 × 10 <sup>-2</sup>	1.38	65 °C, 98% RH	-	-	-	26
	(CN <sub>4</sub> H <sub>7</sub> )(C <sub>10</sub> O <sub>8</sub> H <sub>5</sub> )/Nafion		1.92 × 10 <sup>-1</sup>	9.62 × 10 <sup>-2</sup>	1.996	65 °C, 98% RH	-	-	-	26
	(CN <sub>5</sub> H <sub>8</sub> )(C <sub>10</sub> O <sub>8</sub> H <sub>5</sub> )/Nafion		1.98 × 10 <sup>-1</sup>	9.62 × 10 <sup>-2</sup>	2.06	65 °C, 98% RH	-	-	-	26
HOFs	9%-iHOF-8/Nafion	> 80	1.6 × 10 <sup>-1</sup>	6.2 × 10 <sup>-2</sup>	2.58	100 °C, 98% RH	73.5	50	-	27
	7.5%-iHOF-13/Nafion		1.7 × 10 <sup>-1</sup>	6.2 × 10 <sup>-2</sup>	2.74	100 °C, 98% RH	104.7	50.4	-	28

<sup>a</sup> Terms marked with an asterisk (\*) in the sample description are abbreviations. The corresponding full forms are listed below: PWA (Phosphotungstic acid), HPW (Phosphotungstic acid), PTA (supported HPA).

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