STRUCTURAL
CHEMISTRY

Volume 74 (2018)

Supporting information for article:

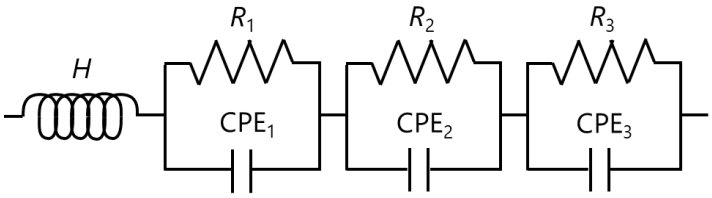
Effect of the ammonium ion on proton conduction in porous ionic crystals based on Keggin-type silicododecatungstate

Satoru Miyazawa, Reina Hosono, Ryota Osuga, Junko Nomura Kondo and Sayaka Uchida

S1. Experimental details

Thermogravimetry (TG) was measured with a Thermo Plus 2 thermogravimetric analyzer (Rigaku Corporation) with α-Al_2O_3 as a reference under a dry N_2 flow (100 mL min^{-1}) and in the temperature range of rt−773 K (Figure S1). IR spectra were measured in the range of 400–4000 cm^{-1} with KBr pellets and a JASCO FT/IR 4100 spectrometer (JASCO) equipped with a TGS detector at a resolution of 4 cm^{-1} (Figure S2).

The following electrical equivalent circuit was used to fit the impedance spectra. CPE_1, CPE_2, and CPE_3 are the constant phase elements, which is often used for imperfect capacitors. *H* represents the effects of the external circuit. R_1, R_2, and R_3 are the bulk, grain boundary, and electrode interface resistances, respectively.



S2. Computing details

Data collection: CrystalClear (Rigaku Inc., 2007), cell refinement: CrystalClear (Rigaku Inc., 2007), data reduction: CrystalClear (Rigaku Inc., 2007), structure solution: SHELXL97 (Sheldrick, 2008), structure refinement: SHELXL-2014/8, molecular graphics: Crystal Structure 4.2 (Rigaku, 2015), software used to prepare materials for publication: Crystal Structure 4.2 (Rigaku, 2015).

Table S1 Experimental details

Crystal data	
C54H80Cr6N8O69SiW12	$F(000)$ = 8232
4491.50	D = 2.853 Mg m-3
Monoclinic, C2/c	Mo$K\alpha$ radiation, λ = 0.71073 Å
a = 32.303(4) Å	Cell parameters from 15010 reflections
b = 25.546(3) Å	θ = 1.04-25.36°
c = 13.561(2) Å	μ = 13.867 mm^{-1}
β = 110.885(6)°	T = 93 K
V = 10456(2)	Block, brown
Z = 4	0.20 × 0.20 × 0.20 mm
Data Collection	
Rigaku Saturn	7855 reflections with $I > 2\sigma(I)$
Graphite monochromator	θ_{max} = 25.344, θ_{min} = 1.044
ω / 2θ scans	h = –38 → +38
35536 measured reflections	k = –30 → +28
9475 independent reflections	l = –14 → +16
Refinement	
Refinement on F^2	9437 reflections
Least-squares matrix: full	662 parameters
$R[F^2 > 2\sigma(F^2)]$ = 0.0577	no restraint
$wR(F^2)$ = 0.1750	Hydrogen atoms were not included
S = 1.111	

Table S2 Fractional atomic coordinates and isotropic or equivalent displacement parameters (Å^2)

	x	*y*	*z*	U_{iso}* / U_{eq}
W1	0.08250(2)	0.22238(2)	0.45055(4)	0.02356(16)
W2	0.08301(2)	0.36076(2)	0.45091(4)	0.02339(16)
W3	-0.00012(2)	0.29122(2)	0.50938(4)	0.02474(16)
W4	0.08314(2)	0.36002(2)	0.19168(5)	0.02883(17)
W5	0	0.15333(3)	0.25	0.0382(2)
W6	0	0.42973(3)	0.25	0.0330(2)
W7	0.08256(2)	0.22207(3)	0.19064(6)	0.0415(2)
Cr1	0.34683(6)	0.52309(7)	0.52795(15)	0.0187(4)
Cr2	0.24018(6)	0.55105(8)	0.46334(15)	0.0209(4)
Cr3	0.27545(6)	0.43017(8)	0.52804(16)	0.0216(4)
Si1	0	0.29143(15)	0.25	0.0102(9)*
O1	0.0330(5)	0.3426(6)	0.2808(12)	0.015(3)*
O2	0.0294(5)	0.2917(6)	0.3661(13)	0.017(3)*
O3	0.0314(5)	0.2395(6)	0.2824(12)	0.016(3)*
O4	0.0290(5)	0.2929(6)	0.1733(13)	0.016(3)*
O5	0.0435(4)	0.2405(4)	0.5211(11)	0.050(3)
O6	0.0432(4)	0.3430(4)	0.5201(12)	0.056(3)
O7	0.0436(6)	0.1669(7)	0.3857(13)	0.100(7)
O8	0.1048(6)	0.2185(7)	0.3406(12)	0.097(7)
O9	0.1215(3)	0.1885(4)	0.1630(8)	0.038(2)
O10	0.1046(4)	0.2919(4)	0.4768(11)	0.047(3)
O11	0.0429(5)	0.4156(7)	0.3831(9)	0.100(7)
O12	0.0423(6)	0.4141(7)	0.1891(11)	0.096(7)
O13	0.1054(4)	0.2930(5)	0.2051(14)	0.093(7)
O14	0	0.4958(5)	0.25	0.039(4)

O15	0.1210(3)	0.3943(6)	0.1594(8)	0.057(4)
O16	0.1068(6)	0.3658(7)	0.3421(10)	0.092(6)
O17	0.0419(5)	0.2425(5)	0.0495(13)	0.095(6)
O18	0	0.0887(5)	0.25	0.060(5)
O19	0.1219(3)	0.3939(4)	0.5478(9)	0.047(3)
O20	0.0435(5)	0.1677(7)	0.1940(11)	0.098(7)
O21	0.1219(4)	0.1906(5)	0.5453(12)	0.069(4)
O22	0.0437(4)	0.3434(4)	0.0507(12)	0.082(5)
O23	0.0004(5)	0.2928(5)	0.6340(10)	0.066(4)
O24	0.2877(3)	0.5013(3)	0.5074(6)	0.0173(17)
O25	0.2705(3)	0.5978(4)	0.5828(7)	0.033(2)
O26	0.3425(3)	0.5884(4)	0.6030(7)	0.029(2)
O27	0.2634(3)	0.5910(3)	0.3706(7)	0.026(2)
O28	0.3310(3)	0.5608(4)	0.3935(6)	0.0261(19)
O29	0.3292(3)	0.4201(4)	0.6547(7)	0.031(2)
O30	0.3729(3)	0.4901(4)	0.6696(7)	0.029(2)
O31	0.3052(3)	0.4019(3)	0.4360(7)	0.030(2)
O32	0.3597(3)	0.4611(3)	0.4576(7)	0.028(2)
O33	0.2062(3)	0.5206(4)	0.5480(7)	0.033(2)
O34	0.2432(3)	0.4487(4)	0.6222(7)	0.032(2)
O35	0.2039(3)	0.5089(4)	0.3421(7)	0.031(2)
O36	0.2193(3)	0.4274(4)	0.4046(7)	0.030(2)
O100	-0.0246(11)	0.3907(14)	0.716(3)	0.100(11)*
O101	0.0005(11)	0.4441(13)	-0.053(3)	0.091(10)*
O102	0.0857(12)	0.5487(14)	0.193(3)	0.098(11)*
N1	0.4126(3)	0.5440(4)	0.5507(8)	0.024(2)
N2	0.2612(4)	0.3526(5)	0.5604(9)	0.031(3)

N3	0.1891(4)	0.6077(4)	0.4155(8)	0.024(2)
N4	0.1116(10)	0.5104(11)	0.385(2)	0.135(10)*
C1	0.3111(5)	0.6074(6)	0.6232(10)	0.030(3)
C2	0.2976(4)	0.5894(5)	0.3479(10)	0.023(3)
C3	0.3613(4)	0.4485(6)	0.6989(10)	0.030(3)
C4	0.3386(4)	0.4198(5)	0.4200(10)	0.026(3)
C5	0.2142(5)	0.4830(6)	0.6106(11)	0.034(3)
C6	0.1992(5)	0.4607(6)	0.3377(11)	0.032(3)
C7	0.4226(4)	0.5923(5)	0.5198(9)	0.026(3)
C8	0.4656(4)	0.6057(5)	0.5336(10)	0.028(3)
C9	0.5009(4)	0.5710(5)	0.5820(11)	0.030(3)
C10	0.4901(4)	0.5227(5)	0.6123(10)	0.026(3)
C11	0.4465(5)	0.5116(5)	0.5967(12)	0.031(3)
C12	0.5477(4)	0.5855(6)	0.6015(13)	0.040(4)
C13	0.5635(5)	0.6284(7)	0.6905(14)	0.049(4)
C14	0.2946(5)	0.3172(5)	0.5903(11)	0.030(3)
C15	0.2888(4)	0.2687(5)	0.6338(13)	0.037(4)
C16	0.2480(5)	0.2569(6)	0.6420(11)	0.034(3)
C17	0.2140(5)	0.2925(6)	0.6017(13)	0.038(4)
C18	0.2211(4)	0.3398(5)	0.5617(11)	0.030(3)
C19	0.2426(7)	0.2070(6)	0.6967(16)	0.059(6)
C20	0.2542(13)	0.2216(15)	0.818(3)	0.139(13)*
C21	0.1725(4)	0.6287(6)	0.4852(12)	0.034(3)
C22	0.1432(5)	0.6701(6)	0.4588(12)	0.041(4)
C23	0.1286(5)	0.6916(6)	0.3589(12)	0.036(3)
C24	0.1449(5)	0.6678(6)	0.2844(12)	0.041(4)
C25	0.1742(5)	0.6273(6)	0.3163(10)	0.035(3)

C26	0.0973(6)	0.7382(7)	0.3252(15)	0.054(5)
C27	0.1151(12)	0.7828(9)	0.399(2)	0.109(11)

Table S3 Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
W1	0.0170(3)	0.0282(3)	0.0223(3)	0.0032(2)	0.0032(2)	0.0051(2)
W2	0.0180(3)	0.0304(3)	0.0167(3)	-0.00719(19)	0.0000(2)	0.0043(2)
W3	0.0253(3)	0.0291(3)	0.0165(3)	0.00589(19)	0.0034(2)	0.0026(2)
W4	0.0205(3)	0.0255(3)	0.0461(4)	0.0013(2)	0.0187(3)	-0.0057(2)
W5	0.0455(5)	0.0107(4)	0.0682(6)	0	0.0323(5)	0
W6	0.0635(6)	0.0106(4)	0.0284(4)	0	0.0204(4)	0
W7	0.0173(3)	0.0444(4)	0.0612(5)	-0.0357(3)	0.0121(3)	-0.0005(2)
Cr1	0.0154(9)	0.0199(10)	0.0192(10)	0.0021(7)	0.0042(7)	0.0006(7)
Cr2	0.0188(10)	0.0258(11)	0.0179(10)	0.0005(8)	0.0062(8)	0.0047(8)
Cr3	0.0150(9)	0.0221(10)	0.0273(11)	0.0072(8)	0.0068(8)	0.0019(8)
O5	0.043(6)	0.021(5)	0.102(10)	-0.007(6)	0.046(7)	-0.001(5)
O6	0.037(6)	0.022(6)	0.128(11)	-0.006(6)	0.051(7)	-0.001(5)
O7	0.106(13)	0.132(15)	0.097(12)	-0.082(11)	0.080(11)	-0.082(12)
O8	0.105(12)	0.135(14)	0.080(10)	-0.079(10)	0.069(10)	-0.103(11)
O9	0.024(5)	0.050(6)	0.035(6)	-0.009(5)	0.006(4)	0.013(5)
O10	0.041(6)	0.033(6)	0.088(9)	-0.003(5)	0.050(7)	0.005(4)
O11	0.119(12)	0.161(16)	0.036(7)	0.035(8)	0.048(8)	0.122(12)
O12	0.127(13)	0.129(14)	0.059(8)	0.056(9)	0.067(9)	0.103(12)
O13	0.038(7)	0.047(8)	0.122(13)	-0.018(7)	-0.060(8)	0.008(6)
O14	0.059(10)	0.010(6)	0.043(9)	0	0.015(7)	0
O15	0.031(6)	0.107(10)	0.032(6)	-0.005(6)	0.009(5)	-0.041(6)
O16	0.116(13)	0.137(14)	0.044(8)	0.041(8)	0.053(8)	0.102(11)
O17	0.067(9)	0.029(7)	0.125(13)	-0.030(7)	-0.045(9)	-0.018(6)
O18	0.086(13)	0.014(7)	0.053(10)	0	-0.010(9)	0
O19	0.030(5)	0.046(7)	0.064(7)	-0.043(6)	0.016(5)	-0.018(5)

O20	0.087(11)	0.159(16)	0.065(9)	-0.078(10)	0.047(8)	-0.101(11)
O21	0.037(7)	0.063(8)	0.105(11)	0.071(8)	0.022(7)	0.027(6)
O22	0.048(7)	0.017(6)	0.118(12)	-0.012(6)	-0.047(8)	0.004(5)
O23	0.108(12)	0.057(9)	0.056(8)	0.021(6)	0.058(9)	0.036(7)
O24	0.014(4)	0.019(4)	0.016(4)	0.001(3)	0.003(3)	0.002(3)
O25	0.025(5)	0.038(6)	0.033(5)	-0.016(4)	0.008(4)	0.003(4)
O26	0.023(5)	0.033(5)	0.027(5)	-0.007(4)	0.005(4)	-0.001(4)
O27	0.029(5)	0.024(5)	0.031(5)	0.009(4)	0.018(4)	0.007(4)
O28	0.019(4)	0.033(5)	0.021(5)	0.007(4)	0.002(4)	0.003(4)
O29	0.020(5)	0.035(5)	0.034(5)	0.012(4)	0.007(4)	0.002(4)
O30	0.022(5)	0.040(6)	0.020(5)	0.010(4)	0.000(4)	-0.006(4)
O31	0.031(5)	0.016(5)	0.043(6)	-0.004(4)	0.015(4)	-0.005(4)
O32	0.016(4)	0.028(5)	0.041(5)	-0.006(4)	0.012(4)	-0.002(4)
O33	0.026(5)	0.041(6)	0.038(6)	0.014(4)	0.020(4)	0.019(4)
O34	0.028(5)	0.029(5)	0.040(6)	0.009(4)	0.015(4)	0.007(4)
O35	0.029(5)	0.028(5)	0.031(5)	-0.003(4)	0.007(4)	0.008(4)
O36	0.025(5)	0.021(5)	0.041(6)	0.003(4)	0.009(4)	-0.001(4)
N1	0.022(5)	0.020(5)	0.029(6)	-0.005(4)	0.008(5)	-0.003(4)
N2	0.021(6)	0.034(7)	0.032(6)	-0.001(5)	0.003(5)	-0.006(5)
N3	0.030(6)	0.024(6)	0.021(6)	-0.003(4)	0.011(5)	0.008(5)
C1	0.031(7)	0.034(8)	0.023(7)	-0.001(5)	0.009(6)	0.001(6)
C2	0.026(6)	0.018(6)	0.025(7)	0.000(5)	0.008(5)	0.009(5)
C3	0.016(6)	0.049(9)	0.023(7)	0.006(6)	0.003(5)	-0.004(6)
C4	0.015(6)	0.023(7)	0.037(8)	0.007(5)	0.006(5)	-0.006(5)
C5	0.029(7)	0.043(9)	0.035(8)	0.007(7)	0.017(6)	0.005(7)
C6	0.032(7)	0.036(9)	0.028(7)	-0.009(6)	0.009(6)	0.002(6)
C7	0.022(6)	0.036(8)	0.014(6)	0.005(5)	-0.003(5)	-0.004(5)

C8	0.023(6)	0.031(7)	0.028(7)	0.006(6)	0.009(5)	0.001(6)
C9	0.014(6)	0.031(7)	0.041(8)	0.003(6)	0.007(5)	-0.006(5)
C10	0.017(6)	0.028(7)	0.032(7)	0.004(5)	0.006(5)	0.001(5)
C11	0.034(8)	0.018(7)	0.044(9)	-0.001(6)	0.016(6)	0.005(6)
C12	0.015(6)	0.036(8)	0.066(11)	0.006(7)	0.011(7)	-0.005(6)
C13	0.038(9)	0.054(11)	0.053(10)	-0.013(8)	0.011(8)	-0.016(8)
C14	0.029(7)	0.024(7)	0.033(8)	0.008(6)	0.006(6)	0.007(6)
C15	0.021(7)	0.018(7)	0.061(10)	0.005(6)	0.000(6)	-0.006(5)
C16	0.029(7)	0.033(8)	0.037(8)	0.012(6)	0.010(6)	-0.016(6)
C17	0.020(7)	0.044(10)	0.049(10)	0.017(7)	0.011(7)	-0.005(6)
C18	0.022(7)	0.029(7)	0.038(8)	-0.002(6)	0.009(6)	-0.014(6)
C19	0.062(12)	0.035(10)	0.064(12)	0.020(8)	0.003(10)	-0.025(8)
C21	0.023(7)	0.037(8)	0.041(8)	-0.005(6)	0.009(6)	0.010(6)
C22	0.046(9)	0.035(8)	0.046(9)	-0.007(7)	0.022(7)	0.018(7)
C23	0.027(7)	0.034(8)	0.050(10)	0.008(7)	0.016(7)	0.008(6)
C24	0.039(9)	0.046(9)	0.032(8)	-0.001(7)	0.007(7)	0.004(7)
C25	0.035(8)	0.040(8)	0.017(7)	-0.004(6)	-0.007(6)	0.016(6)
C26	0.038(9)	0.040(10)	0.084(13)	0.001(9)	0.022(9)	0.022(8)
C27	0.17(3)	0.048(13)	0.080(18)	-0.011(12)	0.013(19)	0.018(16)

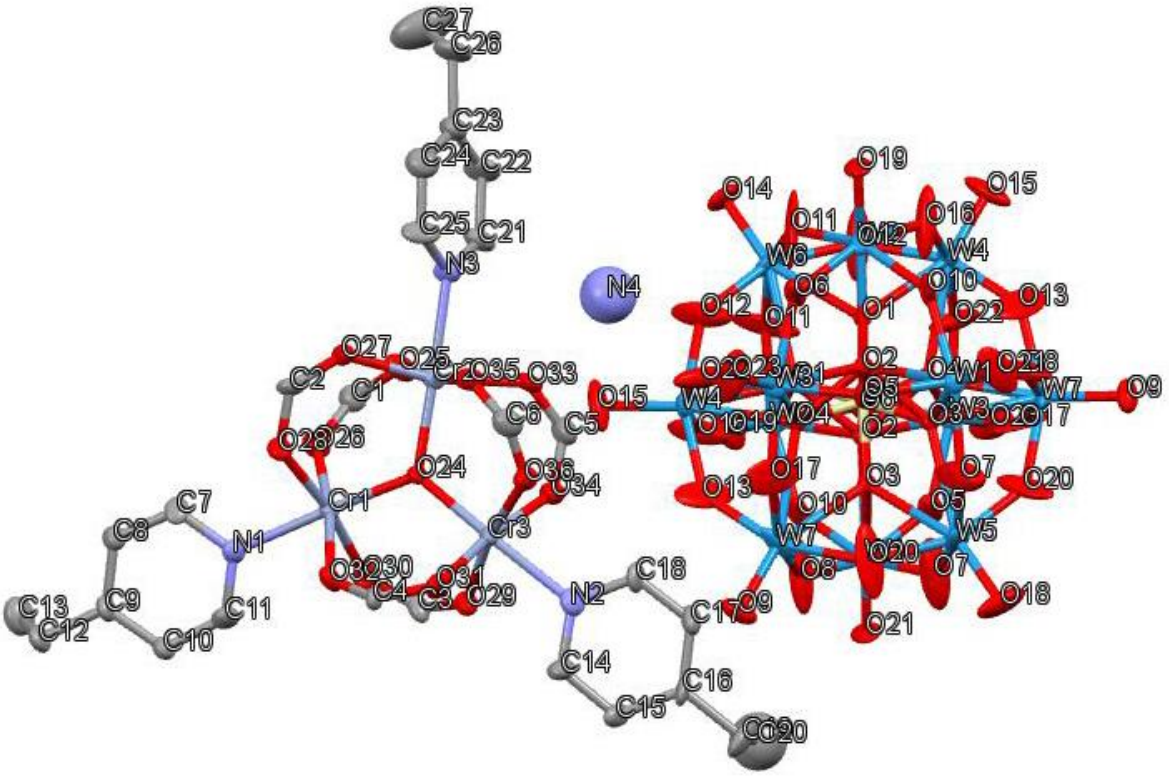
Table S4 Selected geometric parameters (Å, °)

W1-O21	1.663(10)	O21-W1-O8	100.6(8)	O9-W7-O8	100.9(7)
W1-O8	1.875(13)	O21-W1-O7	101.1(8)	O20-W7-O8	86.5(6)
W1-O7	1.891(13)	O8-W1-O7	88.3(5)	O9-W7-O13	102.3(7)
W1-O5	1.891(10)	O21-W1-O5	101.2(6)	O20-W7-O13	156.6(8)
W1-O10	1.900(10)	O8-W1-O5	158.3(8)	O8-W7-O13	87.2(7)
W2-O19	1.687(9)	O7-W1-O5	88.1(5)	O20-W7-O17	89.9(7)
W2-O10	1.878(10)	O21-W1-O10	100.3(7)	O8-W7-O17	156.8(8)
W2-O16	1.896(12)	O8-W1-O10	88.4(5)	O13-W7-O17	87.0(5)
W2-O6	1.898(11)	O7-W1-O10	158.6(8)	O24-Cr1-O28	95.3(3)
W2-O11	1.908(12)	O5-W1-O10	87.1(4)	O24-Cr1-O32	93.9(3)
W3-O23	1.685(12)	O23-W3-O17	101.5(8)	O28-Cr1-O32	88.5(4)
W3-O17	1.799(14)	O23-W3-O5	101.1(6)	O24-Cr1-O26	94.1(4)
W3-O5	1.879(10)	O17-W3-O5	89.0(6)	O28-Cr1-O26	91.2(4)
W3-O6	1.893(10)	O23-W3-O6	99.4(7)	O32-Cr1-O26	172.0(4)
W3-O22	1.899(11)	O17-W3-O6	159.1(8)	O24-Cr1-O30	94.3(4)
W4-O15	1.685(10)	O5-W3-O6	87.9(5)	O28-Cr1-O30	170.3(4)
W4-O13	1.841(12)	O23-W3-O22	98.9(7)	O32-Cr1-O30	92.1(4)
W4-O12	1.903(12)	O17-W3-O22	88.3(5)	O26-Cr1-O30	86.8(4)
W4-O16	1.911(13)	O5-W3-O22	160.0(7)	O24-Cr1-N1	177.8(4)
W4-O22	1.929(12)	O6-W3-O22	87.5(5)	O28-Cr1-N1	85.8(4)
W5-O18	1.652(14)	O15-W4-O13	102.4(8)	O32-Cr1-N1	84.1(4)
W5-O20	1.856(13)	O15-W4-O12	100.4(7)	O26-Cr1-N1	87.9(4)
W5-O7	1.908(16)	O13-W4-O12	157.1(8)	O30-Cr1-N1	84.7(4)
W6-O14	1.687(12)	O15-W4-O16	100.7(7)	O24-Cr2-O27	95.7(4)
W6-O12	1.874(12)	O13-W4-O16	88.4(7)	O24-Cr2-O35	94.0(4)
W6-O11	1.877(13)	O12-W4-O16	88.6(6)	O27-Cr2-O35	90.0(4)

W7-O9	1.671(9)	O15-W4-O22	98.1(6)	O24-Cr2-O25	92.1(4)
W7-O20	1.889(13)	O13-W4-O22	89.4(5)	O27-Cr2-O25	91.1(4)
W7-O8	1.902(16)	O12-W4-O22	86.1(6)	O35-Cr2-O25	173.6(4)
W7-O13	1.938(13)	O16-W4-O22	161.1(8)	O24-Cr2-O33	96.0(3)
W7-O17	1.967(14)	O18-W5-O20	101.4(6)	O27-Cr2-O33	168.3(4)
Cr1-O24	1.913(8)	O20-W5-O20	157.3(12)	O35-Cr2-O33	88.9(4)
Cr1-O28	1.963(8)	O20-W5-O7	87.3(6)	O25-Cr2-O33	88.8(4)
Cr1-O32	1.968(9)	O20-W5-O7	88.6(7)	O24-Cr2-N3	178.4(4)
Cr1-O26	1.985(9)	O18-W5-O7	100.4(6)	O27-Cr2-N3	83.3(4)
Cr1-O30	1.989(8)	O20-W5-O7	88.6(7)	O35-Cr2-N3	87.2(4)
Cr1-N1	2.105(10)	O20-W5- O7	87.3(6)	O25-Cr2-N3	86.7(4)
Cr2-O24	1.917(8)	O7-W5-O7	159.1(12)	O33-Cr2-N3	85.1(4)
Cr2-O27	1.964(8)	O18-W5-O3	156.6(4)	O24-Cr3-O31	94.8(4)
Cr2-O35	1.966(9)	O14-W6-O12	102.3(5)	O24-Cr3-O34	92.9(4)
Cr2-O25	1.970(9)	O12-W6-O12	155.5(11)	O31-Cr3-O34	172.2(4)
Cr2-O33	2.008(9)	O14-W6-O11	101.1(6)	O24-Cr3-O29	95.0(4)
Cr2-N3	2.116(10)	O12-W6-O11	86.5(7)	O31-Cr3-O29	91.5(4)
Cr3-O24	1.901(8)	O12-W6-O11	88.8(6)	O34-Cr3-O29	88.4(4)
Cr3-O31	1.964(9)	O14-W6-O11	101.1(6)	O24-Cr3-O36	94.9(4)
Cr3-O34	1.973(9)	O12-W6-O11	88.8(6)	O31-Cr3-O36	87.6(4)
Cr3-O29	1.975(9)	O12-W6-O11	86.5(7)	O34-Cr3-O36	91.1(4)
Cr3-O36	1.983(9)	O11-W6-O11	157.8(12)	O29-Cr3-O36	170.1(4)
Cr3-N2	2.114(12)	O9-W7-O20	101.0(7)	O24-Cr3-N2	176.2(4)

Symmetry codes: (1) x, y, z, (2) -x, y, -z+1/2, (3) x+1/2, y+1/2, z, (4) -x+1/2, y+1/2, -z+1/2 (5) -x, -y, -z, (6) x, -y, z-1/2, (7) -x+1/2, -y+1/2, -z, (8)'x+1/2, -y+1/2, z-1/2

Atomic numbering scheme



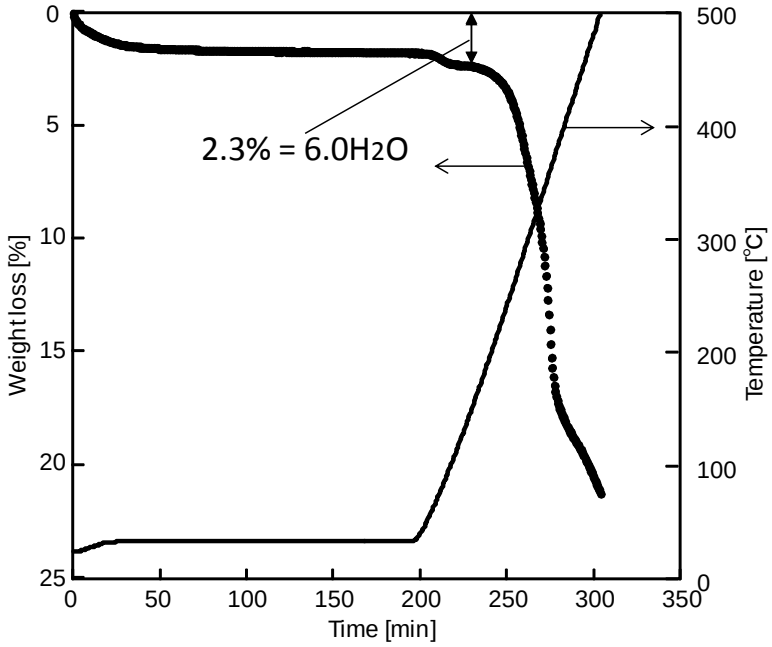


Figure S1 Thermogravimety of **I-NH$_4^+$**.

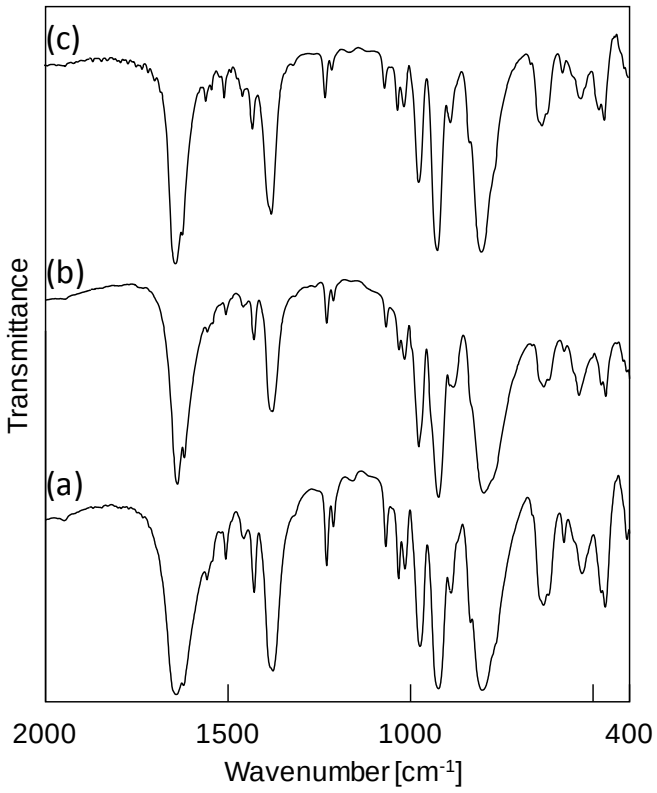


Figure S2 IR spectra of (a) **I-Li$^+$**, (b) **I-K$^+$**, and (c) **I-NH$_4^+$**.

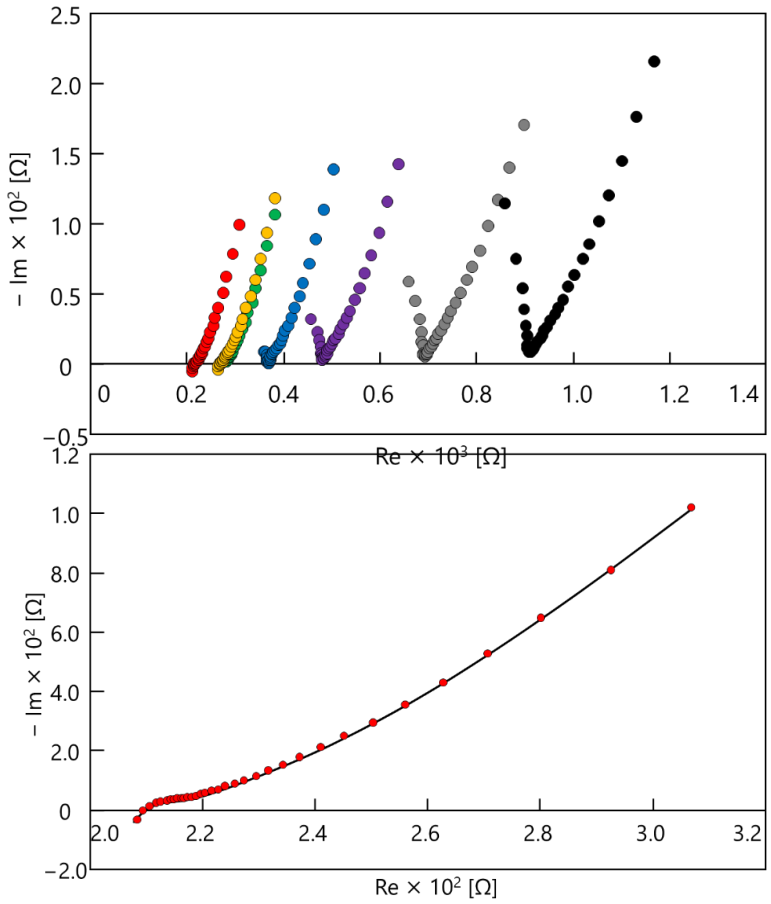


Figure S3 (Top) Nyquist plots of the impedance spectra of **I-NH$_4^+$** at 303–363 K and RH95%. Black, grey, purple, blue, green, yellow, and red plots show the data at 303, 313, 323, 333, 343, 353 and 363 K, respectively. (Bottom) Enlarged plots at 363 K with the fitting data (solid line).

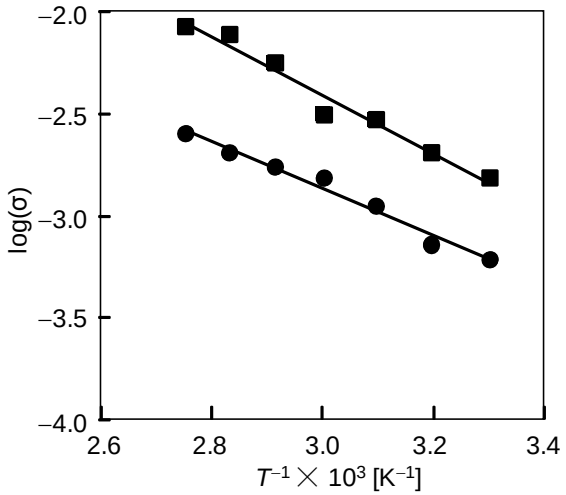


Figure S4 Arrhenius plot of the temperature dependent proton conductivities (303–363 K) of **I-Li$^+$** at RH95% (square). The data of **I-NH$_4^+$** (circle, Figure 3a) is shown as a reference.

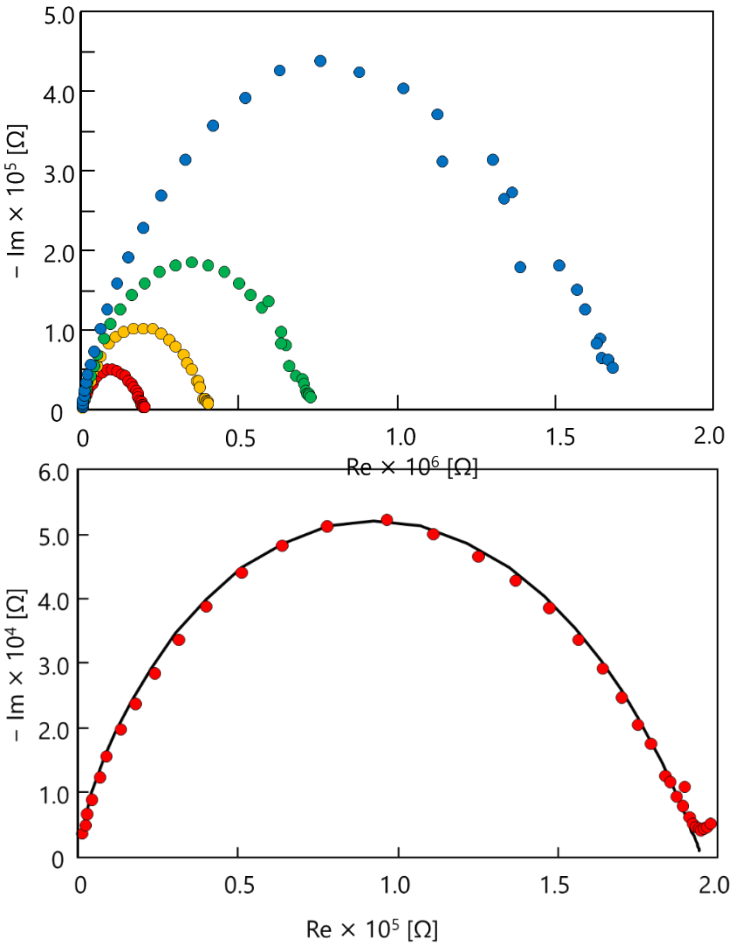


Figure S5 (Top) Nyquist plots of the impedance spectra of **I-NH$_4^+$** at 333–363 K and RH50%. Blue, green, yellow, and red plots show the data at 333, 343, 353 and 363 K, respectively. (Bottom) Enlarged plots at 363 K with the fitting data (solid line).