

# Supporting Information

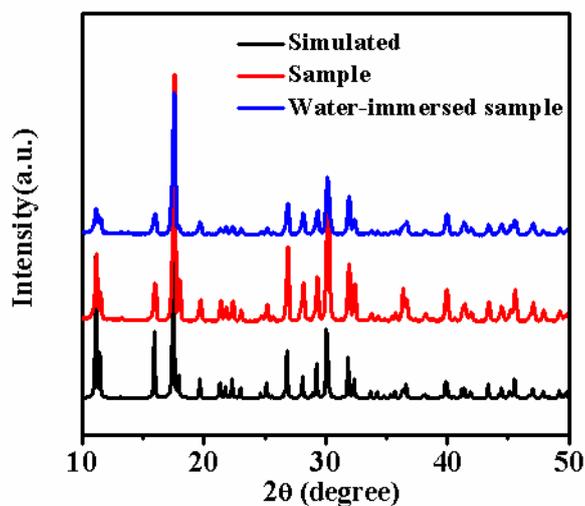
## Proton conduction in organically templated 3D open-framework vanadium-nickel pyrophosphate

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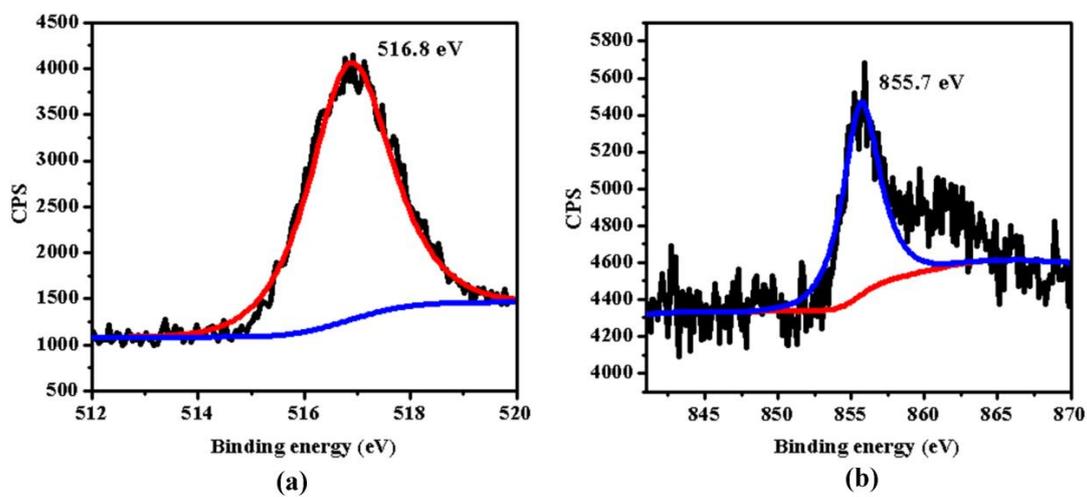
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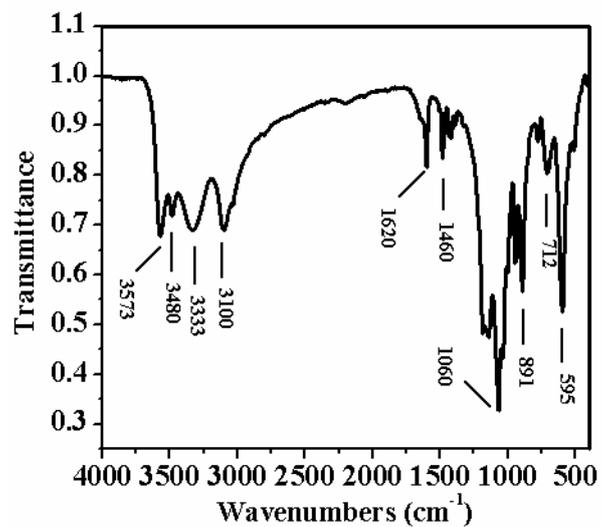
## S1. Supporting Figures



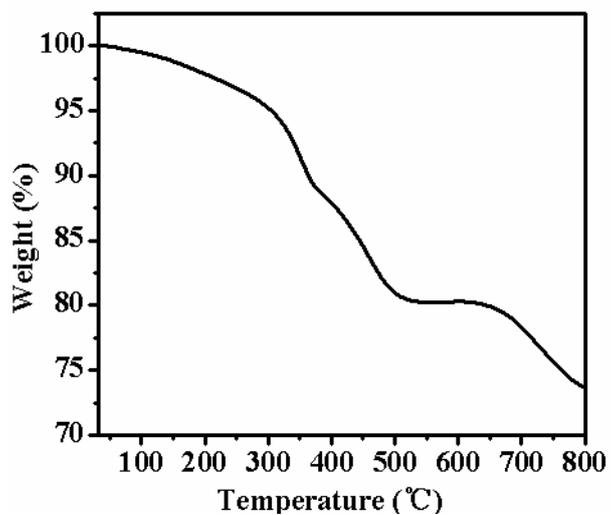
**Fig. S1** Single crystal simulated- (red), power- (black) and water immersed- (blue) power-XRD patterns of **1**.



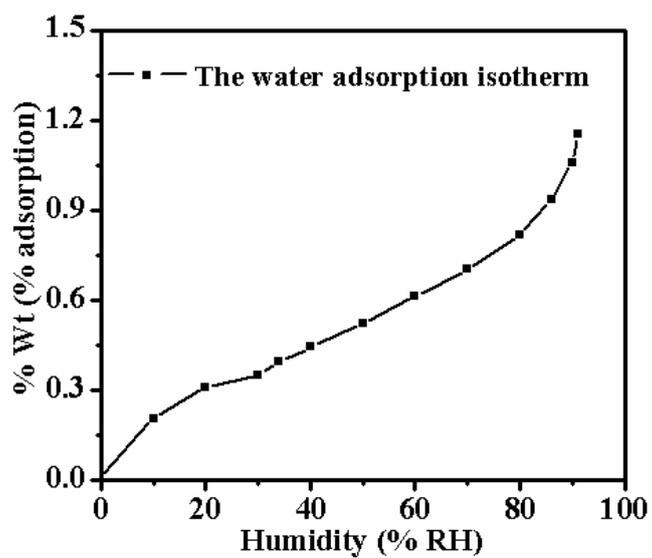
**Fig. S2** XPS spectra of V  $2P_{3/2}$  (a) and Ni  $2P_{3/2}$  (b).



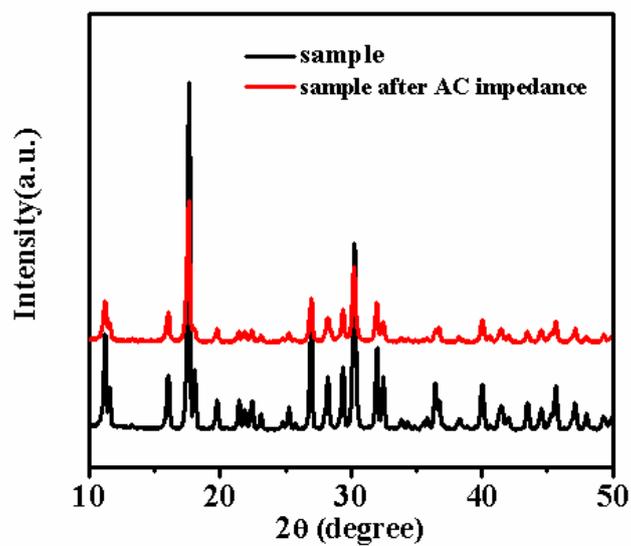
**Fig. S3** FT-IR spectrum of **1**.



**Fig. S4** TGA curve of **1**.



**Fig. S5** The water adsorption isotherm of **1** at room temperature.



**Fig. S6** The XRD patterns of the sample (black) and the pellet used impedance measurement (red).

## S2. Supporting Tables

**Table S1.** Selected bonds length (Å) and angles (°) for **1**.

V(1)-O(5)	1.617(4)	Ni(1)-O(3)	2.032(2)
V(1)-O(2)	1.996(2)	Ni(1)-O(3)#1	2.032(2)
V(1)-O(2)#4	1.996(2)	Ni(1)-O(3)#2	2.032(2)
V(1)-O(4)#5	2.014(2)	Ni(1)-O(3)#3	2.032(2)
V(1)-O(4)#6	2.014(2)	Ni(1)-O(7)#1	2.077(3)
V(1)-O(6)	2.298(4)	Ni(1)-O(7)	2.077(3)
O(4)-V(1)#7	2.014(2)	P(1)-O(4)	1.527(2)
P(1)-O(2)	1.521(2)	P(1)-O(1)	1.6254(17)
P(1)-O(3)	1.499(2)	O(1)-P(1)#4	1.6254(17)
O(5)-V(1)-O(2)	100.65(12)	O(3)#1-Ni(1)-O(3)	180.0
O(5)-V(1)-O(2)#4	100.65(12)	O(3)#-Ni(1)-O(3)#2	93.45(13)
O(2)-V(1)-O(2)#4	89.02(13)	O(3) -Ni(1)-O(3)#2	86.55(13)
O(5)-V(1)-O(4)#5	97.80(12)	O(3)#1-Ni(1)-O(3)#3	86.55(13)
O(2)-V(1)-O(4)#5	161.51(10)	O(3)-Ni(1)-O(3)#3	93.45(13)
O(2)#4-V(1)-O(4)#5	86.54(9)	O(3)#2-Ni(1)-O(3)#3	180.0
O(5)-V(1)-O(4)#6	97.80(12)	O(3)#1-Ni(1)-O(7)#1	93.08(9)
O(2)-V(1)-O(4)#6	86.54(9)	O(3)-Ni(1)-O(7)#1	86.92(9)
O(2)#4-V(1)-O(4)#6	161.51(10)	O(3)#2-Ni(1)-O(7)#1	86.92(9)
O(4)#5-V(1)-O(4)#6	92.03(13)	O(3)#3-Ni(1)-O(7)#1	93.08(9)
O(5)-V(1)-O(6)	179.13(17)	O(3)#1-Ni(1)-O(7)	86.92(9)
O(2)-V(1)-O(6)	79.96(10)	O(3)-Ni(1)-O(7)	93.08(9)
O(2)#2-V(1)-O(6)	79.96(10)	O(3)#2-Ni(1)-O(7)	93.08(9)
O(4)#4-V(1)-O(6)	81.60(10)	O(3)#2-Ni(1)-O(7)	86.92(9)
O(4)#6-V(1)-O(6)	81.60(10)	O(7)#1-Ni(1)-O(7)	180.0
O(3)-P(1)-O(2)	113.06(13)	O(3)-P(1)-O(1)	105.39(14)

O(3)-P(1)-O(4)	113.17(13)	O(2)-P(1)-O(1)	105.96(13)
O(2)-P(1)-O(4)	112.36 (13)	O(4)-P(1)-O(1)	106.11(14)
P(1)#4-O(1)-P(1)	121.38(19)	P(1)-O(2)-V(1)	131.54(13)
P(1)-O(3)-Ni(1)	136.03(14)	P(1)-O(4)-V(1)#7	131.59(14)

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Symmetry transformations used to generate equivalent atoms:

#1 $-x+1, -y+1, -z$	#2 $x, y, -z$	#3 $-x+1, -y+1, z$	#4 $x, y, -z+1$
#5 $x+1/2, -y+3/2, -z+1$	#6 $x+1/2, -y+3/2, z$	#7 $x-1/2, -y+3/2, z$	#8 $-x, -y+1, z$
#9 $-x, -y+1, -z$			

**Table S2. Hydrogen bonds for 1 [(Å) and (°)]**

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
<b>O(6)-H(6A)...O(1)#10</b>	0.99(2)	1.88(2)	2.28(8)	178(6)
<b>O(6)-H(6B)...O(5)#7</b>	0.98(2)	2.06(2)	3.022(6)	166(6)
<b>O(7)-H(7)...O(4)#2</b>	0.98(2)	1.89(3)	2.809(3)	155(5)
<b>N(1)-H(1)...O(3)#11</b>	0.99(2)	2.15(3)	2.940(5)	135(3)
<b>N(1)-H(1)...O(3)#12</b>	0.99(2)	2.15(3)	2.940(5)	135(3)
<b>N(1)-H(1)...O(7)#9</b>	0.99(2)	2.28(8)	2.981(6)	126(7)

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Symmetry transformations used to generate equivalent atoms:

#1 $-x+1, -y+1, -z$	#2 $x, y, -z$	#3 $-x+1, -y+1, z$	#4 $x, y, -z+1$
#5 $x+1/2, -y+3/2, -z+1$	#6 $x+1/2, -y+3/2, z$	#7 $x-1/2, -y+3/2, z$	#8 $-x, -y+1, z$
#9 $-x, -y+1, -z$	#10 $-x+3/2, y+1/2, -z+1$	#11 $x-1, y, -z$	#12 $x-1, y, z$