**Memristive response of a new class of hydrated vanadium oxide intercalation compounds**

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**Supporting Information**

**Table S1. Refinement parameters.** Refinement parameters derived from Rietveld refinement of powder X-ray diffraction data acquired for δ-[Ni(H2O)4]0.2475V2O5•0.147H2O**.** The diffraction data, refined pattern, and residuals are depicted in Figure 1. Rietveld refinement of the collected pattern was performed using the GSAS/EXPGUI software suite.1 The refined structures were rendered using the VESTA software suite.2

|  |  |
| --- | --- |
| **Lattice parameters and refinement statistics** | |
| Space Group | *C2/m* |
| *a* | 11.75306(6) Å |
| *b* | 3.64801(2) Å |
| *c* | 10.37164(9) Å |
| *β* | 95.087(1) Å |
| *V* | 442.935(5) Å3 |
| χ2 | 4.241 |
| ­Rwp | 12.52% |
| Rw | 0.0920 |

**Table S2. Atom positions and occupancies.** Values of atom positions, thermal parameters, and occupancies, as determined from Rietveld refinement of powder X-ray diffraction data acquired for δ-[Ni(H2O)4]*x*V2O5 depicted in Figure 1.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Atom Label** | ***X*** | ***Y*** | ***z*** | **U*iso*\*102** | **Occupancy** |
| Ni | 0.00(0) | 0.00(0) | 0.50(0) | 0.50(7) | 0.495(2) |
| V(1) | 0.94692(7) | 0.00(0) | 0.14244(11) | 0.58(3) | 1.000(0) |
| V(2) | 0.24086(8) | 0.00(0) | 0.13860(11) | 0.39(3) | 1.000(0) |
| O(1) | 0.90946(23) | 0.50(0) | 0.11983(35) | 0.07(9) | 1.000(0) |
| O(2) | 0.09015(26) | 0.00(0) | 0.0940(4) | 0.03(8) | 1.000(0) |
| O(3) | 0.26481(24) | 0.50(0) | 0.1042(4) | 0.08(9) | 1.000(0) |
| O(4) | 0.97053(27) | 0.00(0) | 0.2965(4) | 1.6(1) | 1.000(0) |
| O(5) | 0.24031(25) | 0.00(0) | 0.2980(4) | 1.0(1) | 1.000(0) |
| Ow | 0.38417(34) | 1.104(1) | 0.5177(5) | 5.3(3) | 0.569(4) |
| \*Ow = oxygen from water ligand | | | | | |

**Table S3. Bond lengths and angles for unique polyhedra.** Bond distances and angles for the three unique polyhedra within the δ-[Ni(H2O)4]*x*V2O5 depicted in Figure 1 structure.

|  |  |  |
| --- | --- | --- |
| Polyhedron | Vector | Bond length (Å) |
| NiO6 | Ni\_O(4) | 2.108(4) |
| Ni\_O(4) | 2.108(4) |
| Ni\_O(w) | 2.006(4) |
| Ni\_O(w) | 2.006(4) |
| Ni\_O(w) | 2.006(4) |
| Ni\_O(w) | 2.006(4) |
| V(1)O6 | V(1)\_O(1) | 1.8861(8) |
| V(1)\_O(1) | 1.8861(8) |
| V(1)\_O(2) | 1.7989(31) |
| V(1)\_O(2) | 2.452(4) |
| V(1)\_O(3) | 2.1420(30) |
| V(1)\_O(4) | 1.597(4) |
| V(2)O6 | V(2)\_O(1) | 2.0081(28) |
| V(2)\_O(2) | 1.7905(3) |
| V(2)\_O(3) | 1.8845(9) |
| V(2)\_O(3) | 1.8845(9) |
| V(2)\_O(3) | 2.513(4) |
| V(2)\_O(5) | 1.654(4) |
| V-V distances | V(1)\_V(2) | 3.0293(10) |
| V(2)\_V(2) | 3.4276(19) |
| V(1)\_V(2) | 3.4584(12) |
| V(1)\_V(2) | 3.4966(16) |
| V(1)\_V(1) | 3.64801(2) |
| V(2)\_V(2) | 3.64801(2) |
| Polyhedron | Bonds | Angles |
| NiO6 | O(4)\_Ni\_O(4) | 180.000(0) |
| O(4)\_Ni\_O(w) | 92.25(15) |
| O(4)\_Ni\_O(w) | 87.75(15) |
| V(1)O6 | O(1)\_V(1)\_O(1) | 150.50(18) |
| O(1)\_V(1)\_O(2) | 100.33(9) |
| O(1)\_V(1)\_O(3) | 76.03(8) |
| O(1)\_V(1)\_O(4) | 98.18(12) |
| O(1)\_V(1)\_O(2) | 100.33(9) |
| O(1)\_V(1)\_O(3) | 76.03(8) |
| O(1)\_V(1)\_O(4) | 98.18\*12) |
| O(2)\_V(1)\_O(3) | 153.22(16) |
| O(2)\_V(1)\_O(4) | 101.27(18) |
| O(3)\_V(1)\_O(4) | 105.51(16) |
| V(2)O6 | O(1)\_V(2)\_O(2) | 159.55(16) |
| O(1)\_V(2)\_O(3) | 79.42(9) |
| O(1)\_V(2)\_O(3) | 79.42(9) |
| O(1)\_V(2)\_O(5) | 100.85(17) |
| O(2)\_V(2)\_O(3) | 96.43(9) |
| O(2)\_V(2)\_O(3) | 96.43(9) |
| O(2)\_V(2)\_O(5) | 99.60(17) |
| O(3)\_V(2)\_O(3) | 150.88(12) |
| O(3)\_V(2)\_O(5) | 101.73(12) |
| O(3)\_V(2)\_O(5) | 101.73(12) |

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**FIG. S1 Energy dispersive X-ray spectroscopy for the δ-[M(H2O)4]xV2O5 ternary bronzes.** EDX spectra suggesting stoichiometries of δ-[Ni(H2O)4]0.267V2O5 (black), δ-[Co(H2O)4]0.270V2O5 (red), and δ-[Zn(H2O)4]0.168V2O5 (blue).



**FIG. S2 Device geometries.** Optical microscopy images of individual-nanowire devices fabricated for (a) δ-[Ni(H2O)4]0.25V2O5; (b) δ-[Co(H2O)4]0.25V2O5; and (c) dehydrated δ-Ni0.25V2O5.



**FIG. S3 Single-nanowire transport properties of δ-[Zn(H2O)4]0.168V2O5.** (a) Resistance *versus* temperature curves and (b) current *versus* voltage curves as a function of temperature. The Zn analog that has a somewhat reduced occupancy as a result of larger cation size and further shows relatively diminished hysteretic behavior, which can be attributed to a lower concentration of protons created by hydrolysis.

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**Figure S4. Current *vs*. voltage curves for δ-[Co(H2O)4]*x*V2O5 showing relaxation with continued cycling.** IV curves demonstrate relaxation of the hysteresis loop to a minimum width after 10 sweeps along the same voltage range (0 🡪 2V). Similar behavior is observed when cycling in a negative voltage window (0 🡪 -2V). Despite relaxation of the hysteresis loop, the pristine (uncycled) resistance states (as observed in cycles 1 and 11) can be recovered by changing the polarity of the applied bias. The inset to the figure shows the excellent reproducibility of the hysteresis across cycles 7—10.



**Figure S5. Energy dispersive X-ray spectroscopy of the mixed quaternary bronzes.** Two EDS spectra collected for intermediate compositions: Co1-*x*Zn*x* (blue) and Ni1-xZn*x* (black).

**References**

1. B.H. Toby.: EXPGUI, a graphical user interface for GSAS. *J. Appl. Crystallogr.* **34**, 210 (2001).

2. K. Momma, and F. Izumi.: VESTA 3 for three-dimensional visualization of crystal, volumetric and morphology data. *J. Appl. Crystallogr.* **44**, 1272 (2011).