**Table S1 Fractional Atomic Coordinates, Isotropic Thermal Parameters ($Å^2$) for $NiB_{12}O_{14}(OH)_{10}$**

| Atom | x | y | z | U(iso) |
|---|---|---|---|---|
| Ni1 | 0.5000 | 0.0000 | 0.5000 | 0.00732(18) |
| B1 | 0.5859(3) | 0.0161(5) | 0.2069(4) | 0.0071(8) |
| B2 | 0.3537(4) | 0.2945(5) | 0.4391(5) | 0.0072(8) |
| B3 | 0.2191(4) | 0.0367(4) | 0.3569(5) | 0.0099(9) |
| B4 | 0.8524(4) | 0.4018(5) | 0.4003(5) | 0.0161(10) |
| B5 | 0.4794(3) | 0.2504(5) | 0.2463(5) | 0.0083(9) |
| B6 | 0.9718(4) | 0.0997(5) | 0.7080(5) | 0.0148(9) |
| O1 | 0.3179(2) | -0.0374(3) | 0.4690(3) | 0.0123(6) |
| O2 | 0.2248(2) | 0.0029(3) | 0.1928(3) | 0.0121(5) |
| O3 | 0.5829(2) | 0.1735(3) | 0.2212(3) | 0.0084(5) |
| O4 | 0.8946(2) | 0.0034(3) | 0.6097(3) | 0.0108(5) |
| O5 | 0.4499(2) | 0.2118(3) | 0.3970(3) | 0.0066(5) |
| O6 | 0.2363(2) | 0.2109(3) | 0.3802(3) | 0.0089(5) |
| O7 | 0.6612(2) | -0.0476(3) | 0.1221(3) | 0.0093(5) |
| O8 | 0.3706(2) | 0.1995(3) | 0.1148(3) | 0.0093(6) |
| O9 | 0.5119(2) | -0.0793(3) | 0.2735(3) | 0.0074(5) |
| O10 | 1.0773(3) | 0.1421(4) | 0.6734(3) | 0.0199(7) |
| O11 | 0.9506(3) | 0.1518(3) | 0.8499(3) | 0.0229(7) |
| O12 | 0.8348(4) | 0.3518(5) | 0.5416(4) | 0.0329(10) |
| H1 | 0.184(4) | 0.264(6) | 0.316(6) | 0.034(15) |
| H2 | 1.083(5) | 0.107(6) | 0.597(6) | 0.035(16) |
| H3 | 0.805(5) | 0.389(7) | 0.568(7) | 0.03(2) |
| H4 | 0.323(5) | 0.141(6) | 0.132(6) | 0.034(16) |
| H5 | 0.289(5) | -0.126(7) | 0.480(6) | 0.053(18) |

**Table S2 Anisotropic displacement parameters ($Å^2$) for $NiB_{12}O_{14}(OH)_{10}$**

| | U11 | U22 | U33 | U23 | U13 | U12 |
|---|---|---|---|---|---|---|
| Ni1 | 0.0082(3) | 0.0070(3) | 0.0072(3) | 0.0004(3) | 0.0025(2) | 0.0011(3) |
| B1 | 0.0066(17) | 0.010(2) | 0.0043(16) | 0.0001(17) | 0.0006(14) | -0.0004(17) |
| B2 | 0.009(2) | 0.0057(18) | 0.0069(18) | -0.0007(15) | 0.0027(15) | 0.0006(15) |
| B3 | 0.011(2) | 0.009(2) | 0.0096(19) | 0.0004(15) | 0.0022(16) | -0.0020(15) |
| B4 | 0.017(2) | 0.016(2) | 0.015(2) | 0.0032(18) | 0.0042(19) | 0.0065(18) |
| B5 | 0.009(3) | 0.0078(18) | 0.0079(18) | -0.0008(15) | 0.0017(18) | -0.0012(17) |
| B6 | 0.011(2) | 0.018(2) | 0.014(2) | -0.0005(18) | 0.0025(17) | -0.0057(17) |
| O1 | 0.0099(14) | 0.0112(15) | 0.0151(14) | 0.0023(10) | 0.0009(11) | -0.0017(10) |
| O2 | 0.0119(12) | 0.0148(12) | 0.0108(12) | -0.0012(12) | 0.0052(10) | -0.0033(12) |
| O3 | 0.0075(13) | 0.0068(12) | 0.0121(13) | 0.0005(10) | 0.0045(10) | 0.0007(10) |
| O4 | 0.0085(12) | 0.0145(12) | 0.0104(11) | -0.0019(12) | 0.0040(9) | -0.0034(12) |
| O5 | 0.0081(13) | 0.0079(12) | 0.0041(11) | 0.0001(10) | 0.0024(10) | 0.0006(10) |
| O6 | 0.0070(13) | 0.0096(12) | 0.0089(13) | 0.0015(11) | -0.0010(10) | -0.0002(10) |
| O7 | 0.0103(13) | 0.0074(12) | 0.0118(12) | -0.0019(10) | 0.0059(10) | -0.0013(9) |
| O8 | 0.0092(13) | 0.0126(13) | 0.0069(13) | 0.0011(10) | 0.0032(10) | -0.0040(11) |
| O9 | 0.0079(13) | 0.0052(12) | 0.0096(12) | -0.0012(10) | 0.0033(10) | 0.0000(10) |
| O10 | 0.0154(16) | 0.0316(17) | 0.0142(15) | -0.0083(14) | 0.0063(12) | -0.0126(13) |
| O11 | 0.0253(17) | 0.0297(17) | 0.0160(14) | -0.0095(13) | 0.0097(13) | -0.0191(13) |
| O12 | 0.046(2) | 0.035(2) | 0.0246(19) | 0.0160(16) | 0.0233(18) | 0.0290(19) |

**Table S3 Selected Bond Lengths (Å) and Angles (°) for $NiB_{12}O_{14}(OH)_{10}$**

| | | | |
|---|---|---|---|
| Ni1-O5 | 2.044(2) | O3-B1-O7 | 118.6(3) |
| Ni1-O1 | 2.076(3) | O3-B1-O9 | 121.0(3) |
| Ni1-O9 | 2.104(2) | O7-B1-O9 | 120.4(3) |
| B1-O3 | 1.353(5) | O5-B2-O7 | 113.9(3) |
| B1-O7 | 1.366(5) | O5-B2-O8 | 110.1(3) |
| B1-O9 | 1.392(5) | O7-B2-O8 | 108.6(3) |
| B2-O5 | 1.426(5) | O5-B2-O6 | 111.4(3) |
| B2-O7 | 1.447(5) | O7-B2-O6 | 107.0(3) |
| B2-O8 | 1.488(5) | O8-B2-O6 | 105.4(3) |
| B2-O6 | 1.513(5) | O4-B3-O2 | 112.5(3) |
| B3-O4 | 1.441(5) | O4-B3-O1 | 111.7(3) |
| B3-O2 | 1.462(5) | O2-B3-O1 | 111.2(3) |
| B3-O1 | 1.464(5) | O4-B3-O6 | 107.8(3) |
| B3-O6 | 1.510(4) | O2-B3-O6 | 107.0(3) |
| B4-O12 | 1.350(6) | O1-B3-O6 | 106.2(3) |
| B4-O2 | 1.366(5) | O12-B4-O2 | 121.7(4) |
| B4-O11 | 1.375(5) | O12-B4-O11 | 118.5(4) |
| B5-O3 | 1.417(4) | O2-B4-O11 | 119.8(4) |
| B5-O5 | 1.454(5) | O3-B5-O5 | 113.6(3) |
| B5-O9 | 1.472(5) | O3-B5-O9 | 111.1(3) |
| B5-O8 | 1.556(5) | O5-B5-O9 | 111.4(3) |
| B6-O10 | 1.361(5) | O3-B5-O8 | 109.0(3) |
| B6-O4 | 1.362(5) | O5-B5-O8 | 106.4(3) |
| B6-O11 | 1.377(5) | O9-B5-O8 | 104.8(3) |
| | | O10-B6-O4 | 121.0(4) |
| | | O10-B6-O11 | 116.8(4) |
| | | O4-B6-O11 | 122.1(4) |

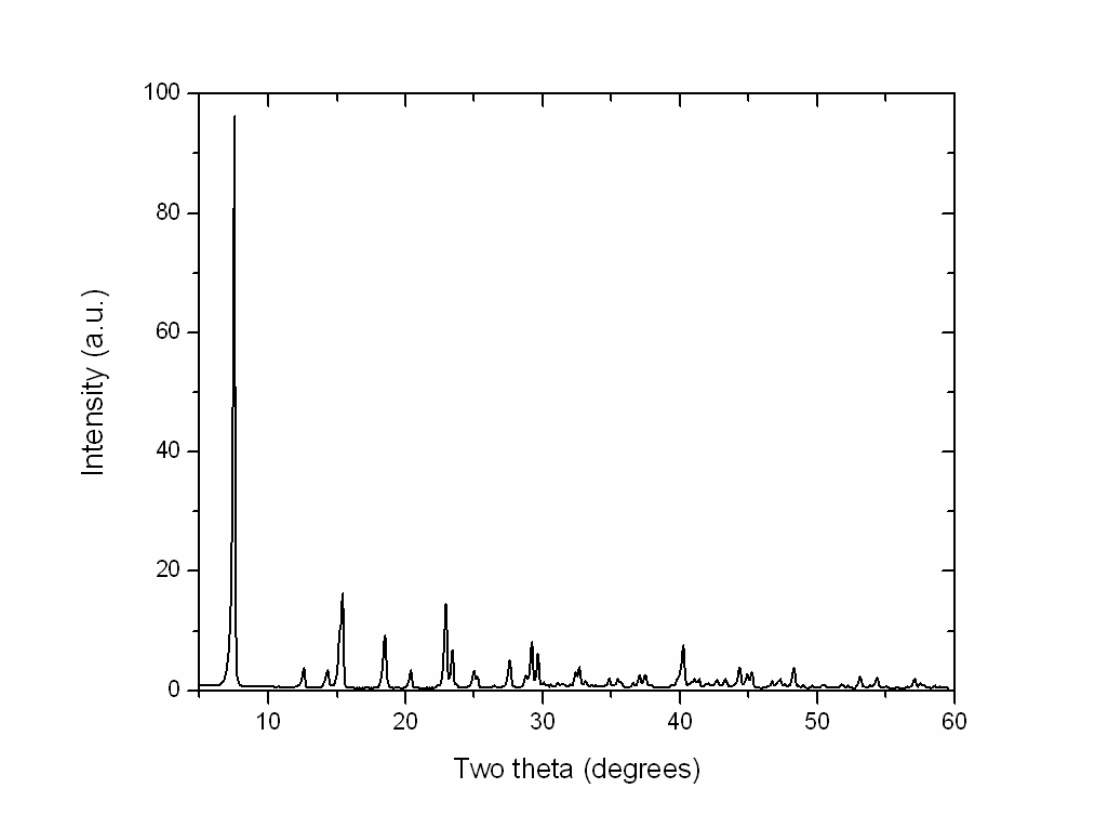


Figure S1. Powder X-ray diffraction pattern of $NiB_{12}O_{14}(OH)_{10}$.

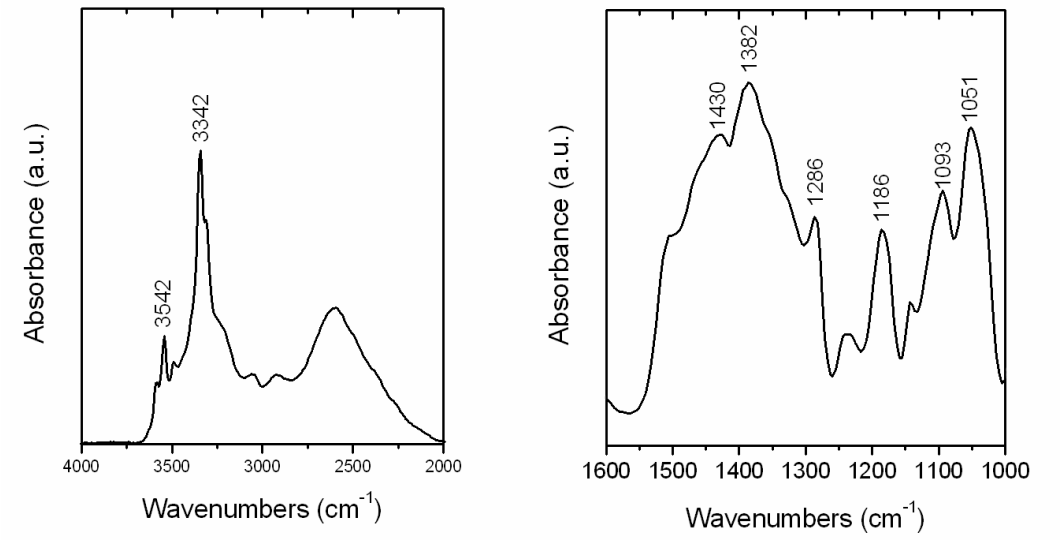


Figure S2. IR spectrum of $NiB_{12}O_{14}(OH)_{10}$. The bands at 3342 and 3542 $cm^{-1}$ correspond to the hydroxyl groups in the structure; the bands at 1430 $cm^{-1}$, 1382 $cm^{-1}$ and 1286 $cm^{-1}$ belong to the B-O antisymmetric stretch in $[BO_3]$; the bands at 1186 $cm^{-1}$, 1093 $cm^{-1}$ and 1051 $cm^{-1}$ belong to the B-O antisymmetric stretch of $[BO_4]$.

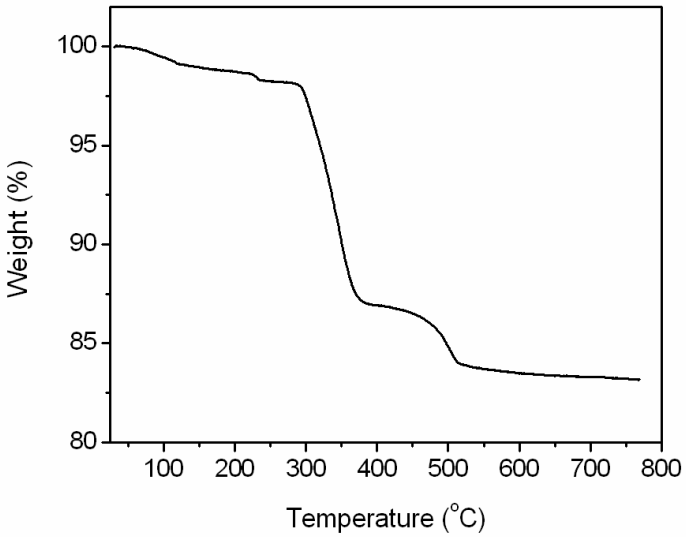


Figure S3. TGA measurement exhibits a total weight-loss of about 16.8 wt% below 800°C, which originates from the dehydration of the hydroxyl groups in $NiB_{12}O_{14}(OH)_{10}$. The content of protons in the structure determined by the weight-loss value is consistent with the structural calculation result.