**Table 1S.** The bond lengths and bond angles of H4L

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angle** | o | **Bond length** | Ǻ |
| H(50)-O(28)-C(26) | 108.03 | O(28)-H(50) | 0.95 |
| H(49)-O(27)-C(17) | 108.23 | O(27)-H(49) | 0.95 |
| O(28)-C(26)-C(25) | 122.35 | C(26)-O(28) | 1.37 |
| O(28)-C(26)-C(15) | 117.22 | C(17)-O(27) | 1.37 |
| N(8)-C(3)-C(5) | 125.86 | C(17)-C(18) | 1.40 |
| N(8)-C(3)-C(1) | 117.39 | C(12)-O(14) | 1.22 |
| N(7)-C(4)-C(6) | 125.72 | C(11)-O(16) | 1.21 |
| N(7)-C(4)-C(2) | 116.92 | N(10)-H(40) | 1.01 |
| O(27)-C(17)-C(18) | 121.65 | N(10)-C(12) | 1.45 |
| O(27)-C(17)-C(13) | 117.36 | N(9)-H(39) | 1.00 |
| O(14)-C(12)-C(13) | 123.08 | N(9)-C(11) | 1.45 |
| O(14)-C(12)-N(10) | 121.20 | N(8)-N(9) | 1.40 |
| C(13)-C(12)-N(10) | 115.57 | N(7)-N(10) | 1.39 |
| O(16)-C(11)-C(15) | 124.81 | C(4)-N(7) | 1.30 |
| O(16)-C(11)-N(9) | 121.48 | C(3)-N(8) | 1.30 |
| C(15)-C(11)-N(9) | 113.43 |  |  |
| H(40)-N(10)-C(12) | 112.54 |  |  |
| H(40)-N(10)-N(7) | 113.42 |  |  |
| C(12)-N(10)-N(7) | 117.21 |  |  |
| H(39)-N(9)-C(11) | 112.49 |  |  |
| H(39)-N(9)-N(8) | 111.13 |  |  |
| C(11)-N(9)-N(8) | 117.22 |  |  |
| N(9)-N(8)-C(3) | 120.59 |  |  |
| N(10)-N(7)-C(4) | 121.80 |  |  |

**Table 2S . Bond angles and lengths of [Cr2(H2L)(OAc)2(OH)2].2H2O**

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angle** | 0 | **Bond length** | Ǻ |
| Cr(30)-O(21)-C(20) | 115.58 | C(37)-C(40) | 1.45 |
| Cr(30)-N(18)-N(19) | 116.58 | C(37)-O(39) | 1.36 |
| C(37)-O(39)-Cr(29) | 126.11 | C(37)-O(38) | 1.28 |
| C(37)-O(38)-Cr(30) | 132.76 | C(33)-O(36) | 1.33 |
| C(33)-O(36)-Cr(30) | 109.89 | C(33)-O(35) | 1.38 |
| C(33)-O(35)-Cr(29) | 72.26 | Cr(30)-O(38) | 1.93 |
| Cr(30)-N(18)-C(15) | 125.80 | Cr(30)-O(36) | 1.91 |
| O(39)-Cr(29)-O(7) | 112.25 | Cr(30)-O(32) | 4.49 |
| O(39)-Cr(29)-N(4) | 134.40 | Cr(30)-O(31) | 1.86 |
| O(35)-Cr(29)-O(31) | 44.90 | Cr(29)-O(39) | 1.84 |
| O(35)-Cr(29)-O(32) | 117.51 | Cr(29)-O(35) | 4.50 |
| O(35)-Cr(29)-O(7) | 65.38 | Cr(29)-O(31) | 1.77 |
| O(35)-Cr(29)-N(4) | 38.62 | C(6)-O(7) | 1.37 |
| O(31)-Cr(29)-O(32) | 109.20 | N(5)-C(6) | 1.37 |
| O(31)-Cr(29)-O(7) | 107.82 | N(4)-Cr(29) | 4.77 |
| O(31)-Cr(29)-N(4) | 83.49 | N(4)-N(5) | 1.27 |
| O(32)-Cr(29)-O(7) | 118.22 | O(21)-Cr(30) | 1.91 |
| O(32)-Cr(29)-N(4) | 112.88 | Cr(29)-O(32) | 1.61 |
| O(7)-Cr(29)-N(4) | 29.91 | O(28)-H(42) | 0.95 |
| Cr(29)-N(4)-N(5) | 11.37 | C(27)-O(28) | 1.36 |
| Cr(29)-N(4)-C(1) | 114.83 | C(26)-C(27) | 1.41 |
| Cr(29)-O(7)-C(6) | 144.79 | C(20)-O(21) | 1.33 |
| Cr(30)-O(32)-Cr(29) | 37.16 | N(19)-C(20) | 1.35 |
| Cr(30)-O(31)-Cr(29) | 135.07 | N(18)-Cr(30) | 1.92 |
| O(38)-Cr(30)-O(36) | 159.63 | N(18)-N(19) | 1.37 |
| O(38)-Cr(30)-O(32) | 73.28 | C(15)-N(18) | 1.43 |
| O(38)-Cr(30)-O(31) | 89.02 | O(14)-H(41) | 0.95 |
| O(38)-Cr(30)-O(21) | 67.89 | C(13)-O(14) | 1.35 |
| O(38)-Cr(30)-N(18) | 108.75 | O(7)-Cr(29) | 1.73 |
| O(36)-Cr(30)-O(32) | 86.58 | C(1)-N(4) | 1.43 |
| O(36)-Cr(30)-O(31) | 71.14 |  |  |
| O(36)-Cr(30)-O(21) | 123.53 |  |  |
| O(36)-Cr(30)-N(18) | 90.70 |  |  |
| O(32)-Cr(30)-O(31) | 16.20 |  |  |
| O(32)-Cr(30)-O(21) | 122.88 |  |  |
| O(32)-Cr(30)-N(18) | 154.21 |  |  |
| O(31)-Cr(30)-O(21) | 135.59 |  |  |
| O(31)-Cr(30)-N(18) | 144.89 |  |  |
| O(21)-Cr(30)-N(18) | 79.52 |  |  |
| O(39)-Cr(29)-O(35) | 126.70 |  |  |
| O(39)-Cr(29)-O(31) | 97.71 |  |  |
| O(39)-Cr(29)-O(32) | 109.64 |  |  |

**Table 3S. Bond angles and lengths of [Mn2(H2L)(OH)2].H2O**

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angles** | **o** | **Bond length** | **Ǻ** |
| Mn(32)-O(33)-Mn(31) | 48.77 | Mn(32)-O(33) | 1.56 |
| C(21)-N(20)-N(19) | 111.34 | Mn(32)-O(34) | 3.50 |
| Mn(32)-N(19)-N(20) | 115.03 | Mn(31)-O(34) | 1.54 |
| Mn(32)-N(19)-C(16) | 119.77 | Mn(31)-O(33) | 3.34 |
| N(20)-N(19)-C(16) | 123.03 | O(29)-H(30) | 0.95 |
| O(34)-Mn(32)-O(33) | 128.13 | N(4)-Mn(31) | 1.90 |
| O(34)-Mn(32)-O(22) | 109.51 | N(4)-N(5) | 1.32 |
| O(34)-Mn(32)-N(19) | 80.00 | C(28)-O(29) | 1.35 |
| O(33)-Mn(32)-O(22) | 121.18 | O(7)-Mn(31) | 2.13 |
| O(33)-Mn(32)-N(19) | 115.32 | C(6)-C(8) | 1.47 |
| O(22)-Mn(32)-N(19) | 82.31 | C(6)-O(7) | 1.26 |
| C(23)-C(21)-N(20) | 119.08 | N(5)-C(6) | 1.43 |
| O(22)-C(21)-N(20) | 119.04 |  |  |
| Mn(32)-O(22)-C(21) | 108.37 |  |  |
| C(23)-C(21)-O(22) | 121.87 |  |  |
| O(34)-Mn(31)-O(33) | 139.85 |  |  |
| O(34)-Mn(31)-O(7) | 117.62 |  |  |
| O(34)-Mn(31)-N(4) | 118.50 |  |  |
| O(33)-Mn(31)-O(7) | 56.36 |  |  |
| O(33)-Mn(31)-N(4) | 99.97 |  |  |
| O(7)-Mn(31)-N(4) | 78.89 |  |  |
| H(30)-O(29)-C(28) | 107.77 |  |  |
| O(29)-C(28)-C(27) | 124.15 |  |  |
| O(29)-C(28)-C(23) | 119.47 |  |  |
| Mn(32)-O(34)-Mn(31) | 42.97 |  |  |
| H(15)-O(14)-C(13) | 107.59 |  |  |
| O(14)-C(13)-C(12) | 125.00 |  |  |
| O(14)-C(13)-C(8) | 118.70 |  |  |
| N(19)-C(16)-C(18) | 127.37 |  |  |
| N(19)-C(16)-C(17) | 112.62 |  |  |
| Mn(31)-O(7)-C(6) | 105.30 |  |  |
| C(8)-C(6)-O(7) | 123.73 |  |  |
| C(8)-C(6)-N(5) | 119.15 |  |  |
| O(7)-C(6)-N(5) | 117.09 |  |  |
| C(6)-N(5)-N(4) | 112.12 |  |  |
| Mn(31)-N(4)-N(5) | 117.29 |  |  |
| Mn(31)-N(4)-C(1) | 103.38 |  |  |
| N(5)-N(4)-C(1) | 124.62 |  |  |
| N(4)-C(1)-C(2) | 100.16 |  |  |
| N(4)-C(1)-C(3) | 130.40 |  |  |

**Table 4S . Bond angles and lengths of [Co2(H2L)(OAc)2].H2O**

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angle** | **o** | **Bond length** | **Ǻ** |
| H(15)-O(14)-C(13) | 107.68 | C(33)-O(35) | 1.30 |
| O(14)-C(13)-C(12) | 124.63 | C(33)-O(34) | 1.29 |
| O(14)-C(13)-C(8) | 119.11 | Co(32)-O(39) | 2.01 |
| N(19)-C(16)-C(18) | 123.25 | Co(32)-O(34) | 1.91 |
| N(19)-C(16)-C(17) | 117.23 | Co(31)-O(38) | 2.02 |
| C(37)-O(39)-Co(32) | 82.78 | Co(31)-O(35) | 1.90 |
| C(37)-O(38)-Co(31) | 82.99 | O(29)-H(30) | 0.95 |
| C(40)-C(37)-O(39) | 126.10 | C(28)-O(29) | 1.36 |
| C(40)-C(37)-O(38) | 124.73 | C(21)-O(22) | 1.30 |
| O(39)-C(37)-O(38) | 109.1 | N(20)-C(21) | 1.38 |
| Co(32)-N(19)-N(20) | 105.90 | N(19)-Co(32) | 1.90 |
| Co(32)-N(19)-C(16) | 137.83 | N(19)-N(20) | 1.34 |
| N(20)-N(19)-C(16) | 116.24 | C(1)-N(4) | 1.45 |
| N(4)-C(1)-C(3) | 123.82 | C(6)-O(7) | 1.31 |
| N(4)-C(1)-C(2) | 117.61 | N(5)-C(6) | 1.38 |
| C(21)-N(20)-N(19) | 113.74 | N(4)-Co(31) | 1.91 |
| C(33)-O(35)-Co(31) | 131.77 | N(4)-N(5) | 1.33 |
| C(33)-O(34)-Co(32) | 132.16 | C(37)-O(39) | 1.32 |
| C(36)-C(33)-O(35) | 113.69 | C(37)-O(38) | 1.33 |
| C(36)-C(33)-O(34) | 114.33 | O(7)-Co(31) | 1.89 |
| O(35)-C(33)-O(34) | 131.98 | C(16)-N(19) | 1.47 |
| O(34)-Co(32)-O(22) | 84.30 | C(13)-O(14) | 1.35 |
| O(34)-Co(32)-N(19) | 166.43 |  |  |
| O(22)-Co(32)-N(19) | 86.11 |  |  |
| O(38)-Co(31)-O(35) | 90.21 |  |  |
| O(38)-Co(31)-O(7) | 172.21 |  |  |
| O(38)-Co(31)-N(4) | 100.71 |  |  |
| O(35)-Co(31)-O(7) | 82.24 |  |  |
| O(35)-Co(31)-N(4) | 166.21 |  |  |
| O(7)-Co(31)-N(4) | 86.49 |  |  |
| H(30)-O(29)-C(28) | 107.65 |  |  |
| O(29)-C(28)-C(27) | 126.38 |  |  |
| O(29)-C(28)-C(23) | 117.49 |  |  |
| Co(31)-O(7)-C(6) | 104.85 |  |  |
| C(8)-C(6)-O(7) | 120.94 |  |  |
| C(8)-C(6)-N(5) | 119.16 |  |  |
| O(7)-C(6)-N(5) | 119.81 |  |  |
| C(6)-N(5)-N(4) | 115.26 |  |  |
| Co(31)-N(4)-N(5) | 106.84 |  |  |
| Co(31)-N(4)-C(1) | 137.46 |  |  |
| N(5)-N(4)-C(1) | 115.68 |  |  |
| H(45)-C(3)-H(44) | 109.52 |  |  |
| Co(32)-O(22)-C(21) | 101.21 |  |  |
| C(23)-C(21)-O(22) | 119.22 |  |  |
| C(23)-C(21)-N(20) | 119.95 |  |  |
| O(22)-C(21)-N(20) | 120.79 |  |  |

**Table 5S . Bond angles and lengths of [Ni(H4L)Cl2].2H2O**

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angle** | **o** | **Bond length** | **o** |
| Cl(33)-Ni(31)-N(19) | 25.67 | C(21)-O(22) | 1.23 |
| Cl(33)-Ni(31)-O(7) | 93.81 | N(20)-C(21) | 2.41 |
| Cl(33)-Ni(31)-N(4) | 95.02 | N(19)-Ni(31) | 1.83 |
| Cl(32)-Ni(31)-O(22) | 32.83 | N(19)-N(20) | 1.35 |
| Cl(32)-Ni(31)-N(19) | 147.17 | C(16)-N(19) | 1.38 |
| Cl(32)-Ni(31)-O(7) | 90.00 | Ni(31)-Cl(33) | 2.14 |
| Cl(32)-Ni(31)-N(4) | 90.00 | Ni(31)-Cl(32) | 2.14 |
| O(22)-Ni(31)-N(19) | 114.33 | O(29)-H(30) | 0.97 |
| O(22)-Ni(31)-O(7) | 109.50 | C(28)-O(29) | 1.36 |
| O(22)-Ni(31)-N(4) | 109.50 | C(6)-O(7) | 2.20 |
| N(19)-Ni(31)-O(7) | 109.50 | N(5)-C(6) | 1.26 |
| N(19)-Ni(31)-N(4) | 109.50 | N(4)-Ni(31) | 1.82 |
| O(7)-Ni(31)-N(4) | 104.00 | N(4)-N(5) | 1.35 |
| H(30)-O(29)-C(28) | 120.00 | O(14)-H(15) | 0.97 |
| O(29)-C(28)-C(27) | 120.00 | C(13)-O(14) | 1.36 |
| O(29)-C(28)-C(23) | 120.00 | O(22)-Ni(31) | 1.79 |
| N(19)-C(16)-C(18) | 120.00 | O(7)-Ni(31) | 1.79 |
| N(19)-C(16)-C(17) | 120.00 | C(1)-N(4) | 1.38 |
| C(23)-C(21)-N(20) | 125.31 |  |  |
| H(15)-O(14)-C(13) | 120.00 |  |  |
| O(14)-C(13)-C(12) | 120.00 |  |  |
| O(14)-C(13)-C(8) | 120.00 |  |  |
| Ni(31)-O(7)-C(6) | 82.96 |  |  |
| C(8)-C(6)-O(7) | 131.61 |  |  |
| C(8)-C(6)-N(5) | 131.61 |  |  |
| O(7)-C(6)-N(5) | 96.79 |  |  |
| C(6)-N(5)-N(4) | 104.00 |  |  |
| Ni(31)-N(4)-N(5) | 104.00 |  |  |
| Ni(31)-N(4)-C(1) | 109.50 |  |  |
| O(22)-C(21)-N(20) | 109.38 |  |  |
| C(21)-N(20)-N(19) | 97.94 |  |  |
| Ni(31)-N(19)-N(20) | 104.00 |  |  |
| Ni(31)-O(22)-C(21) | 104.00 |  |  |
| C(23)-C(21)-O(22) | 125.31 |  |  |
| N(4)-C(1)-C(3) | 120.00 |  |  |
| N(4)-C(1)-C(2) | 120.00 |  |  |
| Ni(31)-N(19)-C(16) | 109.50 |  |  |
| N(20)-N(19)-C(16) | 109.50 |  |  |
| Cl(33)-Ni(31)-O(22) | 139.98 |  |  |

**Table 6S. Bond angles and lengths of [Ni(H2L)].3H2O**

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angle** | **o** | **Bond length** | **Ao** |
| O(16)-C(11)-N(9) | 118.56 | N(10)-C(12) | 1.37 |
| C(15)-C(11)-N(9) | 123.57 | N(9)-C(11) | 1.37 |
| C(12)-N(10)-N(7) | 116.79 | N(8)-Ni(27) | 1.86 |
| C(11)-N(9)-N(8) | 116.80 | N(8)-N(9) | 1.34 |
| Ni(27)-N(8)-N(9) | 106.57 | N(7)-Ni(27) | 1.86 |
| Ni(27)-N(8)-C(3) | 140.50 | N(7)-N(10) | 1.34 |
| N(9)-N(8)-C(3) | 112.93 | C(4)-N(7) | 1.46 |
| Ni(27)-N(7)-N(10) | 106.58 | O(16)-Ni(27) | 1.81 |
| Ni(27)-N(7)-C(4) | 140.43 | C(15)-C(26) | 1.39 |
| N(10)-N(7)-C(4) | 112.99 | C(15)-C(22) | 1.46 |
| O(16)-Ni(27)-O(14) | 76.76 | O(14)-Ni(27) | 1.81 |
| O(16)-Ni(27)-N(8) | 89.85 | C(12)-O(14) | 1.31 |
| O(16)-Ni(27)-N(7) | 166.59 | C(12)-C(13) | 1.45 |
| O(14)-Ni(27)-N(8) | 166.61 | C(11)-O(16) | 1.31 |
| O(14)-Ni(27)-N(7) | 89.83 | C(3)-N(8) | 1.45 |
| N(8)-Ni(27)-N(7) | 103.56 |  |  |
| N(7)-C(4)-C(6) | 126.50 |  |  |
| N(7)-C(4)-C(2) | 113.05 |  |  |
| N(8)-C(3)-C(5) | 126.47 |  |  |
| N(8)-C(3)-C(1) | 113.02 |  |  |
| O(14)-C(12)-C(13) | 117.66 |  |  |
| O(14)-C(12)-N(10) | 118.53 |  |  |
| C(13)-C(12)-N(10) | 123.82 |  |  |
| O(16)-C(11)-C(15) | 117.87 |  |  |
| Ni(27)-O(16)-C(11) | 108.22 |  |  |
| Ni(27)-O(14)-C(12) | 108.27 |  |  |

**Table 7S . Bond lengths and angles of [Zn(H2L)]**

|  |  |  |  |
| --- | --- | --- | --- |
| **Bond angles** | **0** | **Bond length** | **A0** |
| Zn(29)-N(8)-N(9) | 115.10 | O(27)-H(50) | 0.97 |
| Zn(29)-N(8)-C(3) | 117.76 | O(28)-H(51) | 0.97 |
| N(9)-N(8)-C(3) | 127.07 | C(17)-O(27) | 1.21 |
| N(8)-C(3)-C(5) | 130.68 | C(17)-C(18) | 1.46 |
| N(8)-C(3)-C(1) | 103.86 | O(16)-Zn(29) | 2.04 |
| C(11)-N(9)-N(8) | 113.37 | C(11)-O(16) | 1.30 |
| Zn(29)-O(16)-C(11) | 111.14 | C(12)-O(14) | 1.34 |
| Zn(29)-O(14)-C(12) | 53.23 | C(3)-N(8) | 1.43 |
| Zn(29)-N(7)-N(10) | 121.60 | O(14)-Zn(29) | 4.86 |
| Zn(29)-N(7)-C(4) | 111.52 | N(10)-H(41) | 1.05 |
| N(10)-N(7)-C(4) | 124.82 | N(9)-H(40) | 1.05 |
| O(16)-Zn(29)-O(14) | 127.34 | N(10)-C(12) | 1.31 |
| O(16)-C(11)-N(9) | 121.77 | N(9)-C(11) | 1.40 |
| C(15)-C(11)-N(9) | 118.64 | N(8)-Zn(29) | 2.06 |
| O(14)-C(12)-C(13) | 32.76 | N(8)-N(9) | 1.30 |
| O(14)-C(12)-N(10) | 115.52 | N(7)-Zn(29) | 1.97 |
| C(13)-C(12)-N(10) | 148.09 | N(7)-N(10) | 1.32 |
| H(40)-N(9)-N(8) | 123.32 | C(4)-N(7) | 1.47 |
| O(16)-Zn(29)-N(8) | 78.61 | C(26)-O(28) | 1.21 |
| O(16)-Zn(29)-N(7) | 143.95 |  |  |
| O(14)-Zn(29)-N(8) | 146.83 |  |  |
| O(14)-Zn(29)-N(7) | 17.61 |  |  |
| N(8)-Zn(29)-N(7) | 135.90 |  |  |
| H(40)-N(9)-C(11) | 123.32 |  |  |