

## **Supporting Information**

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## Preferential Formation of Homochiral Helical Sandwich-Shaped Architectures via Metal-Mediated Assembly of Tris(imidazoline) Ligands with a Set of d<sup>3</sup>-d<sup>10</sup> Transition-Metal Ions

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## I. Crystal structure of the coordination polymer of CdCl<sub>2</sub>·2DMF



*Figure 1.* Crystal structure of the coordination polymer of CdCl<sub>2</sub>·2DMF. C (gray), N (blue), Cl (green), O (red), and Cd (pink). All hydrogen atoms have been omitted for clarity.





*Figure 2.* <sup>1</sup>H and <sup>13</sup>C NMR spectra of (S,S,S)-4 in CDCl<sub>3</sub>



*Figure 3.* <sup>1</sup>H and <sup>13</sup>C NMR spectra of (R,R,R)-4 in CDCl<sub>3</sub>

III. Copies of <sup>1</sup>H and <sup>13</sup>C NMR spectra of the sandwich-shaped (*M*)-M<sub>3</sub>(L<sub>S</sub>)<sub>2</sub> complexes



*Figure 4.* <sup>1</sup>H and <sup>13</sup>C NMR spectra for (M)-Ag<sub>3</sub>(L<sub>5</sub>)<sub>2</sub>(NO<sub>3</sub>)<sub>3</sub> complex in CD<sub>2</sub>Cl<sub>2</sub> at 293K



*Figure 5.* <sup>1</sup>H and <sup>13</sup>C NMR spectra for (M)-Pd<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>2</sub>Cl<sub>2</sub> at 293 K



*Figure 6.* <sup>1</sup>H and <sup>13</sup>C NMR spectra for (M)-Cu<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>3</sub>OD at 293 K



*Figure 7.* <sup>1</sup>H and <sup>13</sup>C NMR spectra for (M)-Cd<sub>3</sub>(L<sub>*S*</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>3</sub>OD at 293 K



*Figure 8.* <sup>1</sup>H and <sup>13</sup>C NMR spectra for (*M*)-Zn<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>3</sub>OD at 293 K



*Figure 9.* <sup>1</sup>H and <sup>13</sup>C NMR spectra for (M)-Co<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>3</sub>OD at 293 K



*Figure 10*. <sup>1</sup>H and <sup>13</sup>C NMR spectra for (*M*)-Mn<sub>3</sub>(L<sub>5</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>3</sub>OD at 293 K



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*Figure 11*. <sup>1</sup>H and <sup>13</sup>C NMR spectra for (*M*)-Ni<sub>3</sub>(L<sub>*s*</sub>)<sub>2</sub>Cl<sub>6</sub> complex in CD<sub>3</sub>OD at 293 K



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IV. Copies of ESI-TOF mass spectra of the sandwich-shaped (*M*)-M<sub>3</sub>(L<sub>S</sub>)<sub>2</sub> complexes

*Figure 14.* The ESI-TOF mass spectrum of (M)-Ag<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>(NO<sub>3</sub>)<sub>3</sub>



Figure 15. The ESI-TOF mass spectrum of (M)-Pd<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub>



*Figure 16.* The ESI-TOF mass spectrum of (*M*)-Cu<sub>3</sub>(L<sub>s</sub>)<sub>2</sub>Cl<sub>6</sub>



*Figure 17.* The ESI-TOF mass spectrum of (*M*)-Cd<sub>3</sub>(L<sub>s</sub>)<sub>2</sub>Cl<sub>6</sub>



*Figure 18.* The ESI-TOF mass spectrum of (M)-Zn<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub>



*Figure 19.* The ESI-TOF mass spectrum of (*M*)-Co<sub>3</sub>(L<sub>s</sub>)<sub>2</sub>Cl<sub>6</sub>



Figure 20. The ESI-TOF mass spectrum of (M)-Mn<sub>3</sub>(L<sub>s</sub>)<sub>2</sub>Cl<sub>6</sub>



*Figure 21.* The ESI-TOF mass spectrum of (M)-Ni<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>6</sub>







*Figure 23.* The ESI-TOF mass spectrum of (M)-Fe<sub>3</sub>(L<sub>S</sub>)<sub>2</sub>Cl<sub>9</sub>