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I hereby certify that the work embodied in this thesis is the result of original research done by me except where otherwise stated in this thesis. The thesis work has not been submitted for a degree or professional qualification to any other university or institution. I declare that this thesis is written by myself and is free of plagiarism and of sufficient grammatical clarity to be examined. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.



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Chapter 4 is published as D.T. Murphy, S. Schmid, J.R. Hester, P.E.R. Blanchard, and W. Miiller. Coordination site disorder in spinel-type LiMnTiO4. *Inorganic Chemistry* **54**, 4636-4643 (2015). DOI: 10.1021/ic502747p.

The contributions of the co-authors are as follows:

* A/Prof Schmid provided the initial project direction and edited the manuscript drafts.
* I prepared the manuscript drafts. The manuscript was revised by Dr Hester and Dr. Blanchard.
* I co-designed the study with A/Prof Siegbert Schmid and performed all the laboratory work at the School of Materials Science and Engineering and the Singapore Synchrotron Light Source. I also analyzed the data.
* All microscopy, including sample preparation, was conducted by me in the Facility for Analysis, Characterization, Testing and Simulation.
* Dr James Hester assisted in the collection of the neutron powder diffraction data.
* Dr Peter Blanchard assisted in the interpretation of the X-ray absorption spectroscopy data and carried out the spectral interpretation.
* Dr Wojciech Miiller assisted in the collection and provide guidance in the interpretation of the magnetic measurement data.

Chapter 5 is published as H. V Doan, B. Yao, Y. Fang, A. Sartbaeva, U. Hintermair, V. P Ting, Controlled Formation of Hierarchical Metal-Organic Frameworks using CO2 Expanded Solvent Systems. In press, *ACS Sustainable Chemistry & Engineering* (2017). DOI: 10.1021/acssuschemeng.7b01429.

The contributions of the co-authors are as follows:

* Prof Ting suggested the materials area and edited the manuscript drafts.
* I wrote the drafts of the manuscript. The manuscript was revised together with Dr. Sartbaeva and Dr. Yao.
* I performed all the materials synthesis, collected X-ray diffraction patterns and visible light spectra, carried transmission electron microscopy, and conducted data evaluation.
* Dr. Y. Fang conducted the Rietveld analysis of the powder X-ray diffraction data and single crystal structure determinations.
* Dr U. Hintermair conducted the molecular dynamics simulations.
* Ms. A. Sartbaeva prepared the samples for electron microscopy.

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