Li₂MnSiO₄ : High storage capacity cathode material for Li ion battery applications

Vishwanathan Ramar and Palani Balaya National University of Singapore

Department of Mechanical Engineering EA-05-50, 10 Kent Ridge Crescent, Singapore-117576 (E-mail: <u>vishwanathan.ramar@nus.edu.sg</u>)

Increasing demands in green and sustainable energy production and its storage have stimulated significant effort in developing safer and cheaper Li-ion batteries (LIBs) with high specific energy and power. LIBs have been considered to be one of the most impending energy storage systems in powering electric vehicles to promote the low emission transport and power-grid applications, however they demand development of lightweight and long life batteries¹.². Increasing the specific capacity of cathode material is considered as a potential approach in increasing the energy density and to lower the weight and volume of the batteries³. ⁴. To that end, there has been a focus for successful development of alternative candidates to phospho-olivine LiFePO₄. Lithium metal silicates, especially Li₂MnSiO₄ has attracted attention as materials of next generation cathode material owing to their high specific capacity. Li2MnSiO4 has the theoretical probability to facilitate extraction / insertion of two moles of lithium ions per formulae unit resulting in \sim 333 mAh.g $\frac{5-7}{2}$. Additionally, lithium metal silicates also have rich polymorphism owing to a range of crystal chemistry. Moreover, these materials are relatively cheap, abundant and safe which makes them more sustainable. However, isolation of different polymorphs, achieving full capacity, good cycle life and rate performance have been challenging until now owing to structure instability caused by J-T effect and amorphization of the material during subsequent cycling^{$\underline{8}, \underline{9}$}.

In this investigation, we study the role of temperature in isolating different polymorphs and their electrochemical kinetics. Here, we present three different polymorphs (Pmn2₁, Pmnb and P2₁/n) isolated by controlling the temperature and hence synthesis conditions. We are able to extract more than one lithium (close to theoretical) successfully from all the above crystal structures. These material exhibits high storage capacity of ~263 mAh.g⁻¹ at 0.1C and impressive rate performance up to 9C at room temperature.

References

1. M. Armand; J. M. Tarascon, *Nature* **2008**, 451, (7179), 652-657.

- 2. B. Kang; G. Ceder, Nature 2009, 458, (7235), 190-193.
- 3. P. G. Bruce; B. Scrosati; J.-M. Tarascon, *Angewandte Chemie International Edition* **2008**, 47, (16), 2930-2946.
- 4. A. Magasinski; P. Dixon; B. Hertzberg; A. Kvit; J. Ayala; G. Yushin, *Nat Mater* **2010**, *9*, (4), 353-358.
- 5. A. Nytén; A. Abouimrane; M. Armand; T. Gustafsson; J. O. Thomas, *Electrochemistry Communications* **2005**, 7, (2), 156-160.
- 6. R. Dominko; M. Bele; M. Gaberšček; A. Meden; M. Remškar; J. Jamnik, *Electrochemistry Communications* **2006**, 8, (2), 217-222.
- 7. M. Kuezma; S. Devaraj; P. Balaya, *Journal of Materials Chemistry* **2012**, 22, (39), 21279-21284.

8. R. Dominko, *Journal of Power Sources* **2008**, 184, (2), 462-468.

9. Y.-X. Li; Z.-L. Gong; Y. Yang, *Journal of Power Sources* **2007**, 174, (2), 528-532.