

Synthesis of $\text{Li}_2\text{FeSiO}_4$ cathode materials using inexpensive and abundant materials by conventional methods

T. Mokkelbost¹, S. M. Hanetho¹, A. Fossdal¹, E. Sheridan¹, H. Zhou², F. Vullum-Bruer²

SINTEF Materials and Chemistry¹, Norwegian University of Science and Technology², 7465 TRONDHEIM, NORWAY

tommy.mokkelbost@sintef.no

Electrical energy may be efficiently stored in electrochemical storage units such as batteries and capacitors for a wide range of applications. Several different concepts of batteries exist, but in recent years, Li ion batteries have led to enhanced performance in a number of applications e.g. hybrid and electrical vehicles.

In the Li ion battery, the active electrode materials typically consist of graphite as the anode and a metal oxide as the cathode where both have the ability to intercalate Li ions. There is considerable interest in increasing the capacity of the battery by several means including replacing graphite with silicon(1) by different methods(2) and/or by changing the cathode materials. A commonly used cathode material in commercial batteries is lithium nickel manganese cobalt oxide. Both the nickel and cobalt pose problems environmentally and are relatively expensive.

Thus, there is a considerable focus on replacing the expensive cathode materials while retaining, or perhaps increasing, the performance and stability of lithium ion batteries. The introduction of LiFePO_4 has partly solved this problem, by removing the expensive, toxic and less abundant metals. However, due to the lack of electronic conductivity the introduction of carbon and the importance of nanosized particles results in more expensive processing routes.

The orthosilicates $\text{Li}_2(\text{Fe, Mn, Co})\text{SiO}_4$ are a new type of cathode material with possible additional benefits compared to LiFePO_4 such as two available Li ions per formula unit for charge transport and better stability due to the SiO_4 group(3). However, the orthosilicates have even lower conductivity than LiFePO_4 and there are challenges related to several different crystallographic polymorphs. These issues need to be resolved, preferably using inexpensive processing routes and precursors. Here we present possible ways to accomplish this by using raw materials readily available in Norway combined with conventional processing routes. The cathode materials are compared with similar materials synthesis by PVA assisted gel formation process. (4, 5)

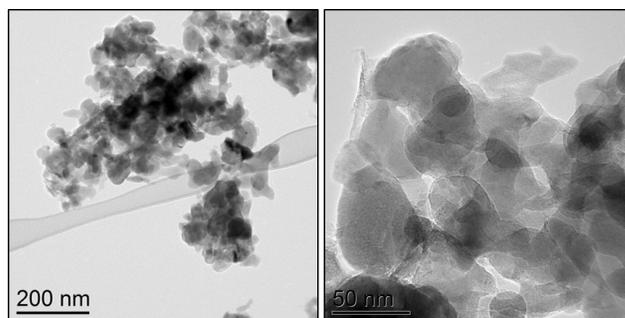
Experimental

$\text{Li}_2\text{FeSiO}_4/\text{C}$ composites were prepared by mixing Li_2CO_3 , iron source, silica source and carbon source. The different sources were

- Fe_3O_4 submicron powders and Fe-oxalate
- Fused silica, colloidal silica and silica waste from Si production
- Sucrose, glucose and pitch.

The powder mixtures were pressed uniaxially to enhance the reaction and formation of the active $\text{Li}_2\text{FeSiO}_4$ phase. The pellets were heat treated in an argon atmosphere at 700 °C for 10 hours with a hold at 350 °C for 5 hours.

The $\text{Li}_2\text{FeSiO}_4/\text{C}$ composites were examined for phase purity (XRD) and morphology (SEM and TEM). The electrochemical performance of the CR2016 coin cell battery assemblies consisting of the above mentioned $\text{Li}_2\text{FeSiO}_4$ cathode materials, a Li foil anode, Celgard 2320 separator and 1.0M LiPF_6 in 3:7 EC:DEC electrolyte was studied. The assembly and composition of the components was similar to previous work by Zhou et al. (4) for comparison. Charge/discharge behavior was studied by galvanostatic cycling to evaluate the charge capacity and stability while the insertion and extraction reactions were studied by cyclic voltammetry. The electrochemical characteristics of the different batteries are discussed and related to the synthesis techniques and different precursors of the cathodes.



Transmission electron micrographs of mainly $\text{Li}_2\text{FeSiO}_4$ synthesized using Li_2CO_3 , Fe_2O_3 and SiO_2

Acknowledgement

Financial support is gratefully acknowledged from the Research Council of Norway RENERGI program, grant number 216469/E20. Per Erik Vullum (SINTEF) is acknowledged for TEM imaging.

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