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J Material Sci Eng 2018, Volume 7
DOI: 10.4172/2169-0022-C11-123

7th International Conference on

Smart Materials and Structures

July 02-03, 2018 | Vienna, Austria

Pairing transition metal difluoride cathodes with silicon-based anodes for high energy density lithium ion batteries

Nasr Bensalah and **Fadi Z Kamand**
Qatar University, Qatar

Lithium (Li)-ion batteries have proven to be vital in meeting the critical challenges of integrating renewable energy sources into a smart electrical grid as well as replacing internal combustion car engines with environmentally friendly electrical motors. To be cost-competitive, such applications require significantly reduced cost and increased energy density of the Li-ion cells beyond state of the art. The energy density of such cells depends on the volumetric capacity of their electrodes. Silicon (Si) - based anodes and fluoride (F) - based cathodes demonstrate great potential for the revolutionary enhancements in the energy storage of Li-ion cells. Unfortunately, these materials also suffer from several shortcomings, such as high electrical resistivity, low Li diffusivity and significant volume change during the battery operation, which limit their stability and power characteristics. By rationally nano-engineering the building blocks for these electrodes, we will overcome these limitations while delivering over 80 % of their theoretical capacity. We report herein the successful methodology to produce uniform size-controlled mixed metal fluorides nanocomposites. The independent and precise control over the cathode composition and morphology allowed us to detect that the initial cathode charge-transfer resistance determines the rate of capacity fading with cycling. Systematic electrochemical measurements in combination with post-mortem analyses led to the conclusion that the cathode stability strongly depends on the ability to prevent formation and growth of a resistive cathode solid electrolyte interphase (CEI), which, in turn, strongly depends on the metal composition. Metal difluoride cathodes were paired with CNT/Si nanocomposites anodes and were tested in full cells. Our findings will help to establish a way to developing Si-based anodes//MFs-based cathodes for commercial LIBs.