

Effect of single-walled carbon nanotube sub-carbon additives and graphene oxide coating for enhancing the 5 V $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ cathode material performance in lithium-ion batteries

Yi-De Tsai ^{a,b}, Jeng-Ywan Shih ^b, Ying-Jeng James Li ^{a,b}, Tai-Feng Hung ^a, Li-Fan Hsu ^a, Sayee Kannan Ramaraj ^c, Rajan Jose ^d, Chelladurai Karuppiyah ^{a*}, and Chun-Chen Yang ^{a,b,e*}

^a Battery Research Center of Green Energy, Ming Chi University of Technology, New Taipei City, 24301, Taiwan

^b Department of Chemical Engineering, Ming Chi University of Technology, New Taipei City, 24301, Taiwan

^c PG and Research Department of Chemistry, Thiagarajar College, Tamil Nadu, Madurai, 625009, India

^d Nanostructured Renewable Energy Materials Laboratory, Faculty of Industrial Sciences and Technology, University Malaysia Pahang, Kuantan, 26300, Malaysia

^e Department of Chemical and Materials Engineering, Chang Gung University, Kwei-shan, Taoyuan, 333, Taiwan

ABSTRACT

High-voltage spinel $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (LNMO) is a promising cathode material for next-generation lithium-ion batteries (LIBs), but its poor cycle performance has impeded its commercialization. In this study, we developed highly stable LNMO cathode materials having an octahedral morphology through a solid-state high-energy ball-mill-cum-spray-drying method. We also developed a novel strategy for modifying this cathode material with two kinds of carbon materials, thereby improving the electrochemical cycling performance. Introducing single-walled carbon nanotubes (SWCNTs) as a sub-carbon conductive additive during the slurry preparation process improved the conductivity of electrons between the particles of the cathode material. The LNMO electrode modified with the SWCNT sub-carbon additives exhibited an average Coulombic efficiency of 99.4% after 500 cycles at 1C, compared with 98.9% for the pristine LNMO-based electrode. Furthermore, we used a wet-chemical method to coat graphene oxide (GO) onto the post-sintered LNMO cathode material to act as a protective layer, preventing corrosion induced by HF in the electrolyte. The capacity retention of the GO-coated LNMO electrode after 500 cycles at 1C (91.8%) was higher than that of the pristine LNMO (52.5%). The corresponding dual-modification strategy, combining the SWCNTs and GO, provided LNMO cathode materials exhibiting superior rate performance and cyclability, with an average Coulombic efficiency of 99.3% and capacity retention of 92.9% after 500 cycles at 1C. Thus, the LNMO cathode materials prepared in this study possessed excellent electrochemical properties favoring their marketability, applicability, and competitiveness for application in high-voltage LIBs.

KEYWORDS

Carbon nanotubes; High-voltage cathode material; $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$; Solid-state reaction; Spray-dry method

ACKNOWLEDGEMENTS

Financial support from the Ministry of Science and Technology, Taiwan (Project No: MOST 108-2221-E-131 -022-MY3) is gratefully acknowledged.