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Controllable Formation of Lithium Carbonate Surface Phase during Synthesis of Nickel-rich $LiNi_{0.9}Mn_{0.1}O_2$ in Air and Its Protection Role in Electrochemical Reaction

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Abstract

It has been well recognized that Li_2CO_3 is inevitably formed at the surface of nickel rich layered compounds (NRLC) upon storage in air which is usually considered to be detrimental to the performance. Actually, its formation may have been triggered during synthesis of NRLC. However, little is known of its formation during synthesis of NRLC in air and its interaction behaviors with electrolyte during electrochemical process. Herein, we successfully tailor Li_2CO_3 surface phase simply by controlling calcination time during synthesis of structurally well ordered $LiNi_{0.9}Mn_{0.1}O_2$ particles with great similarity in structure, morphology, size and crystallinity in air using a facile sol-gel method. The thickness of Li_2CO_3 layer is for the first time observed to decrease with increased calcination time, accompanied by the reduced Ni^{2+} at surface. More oxygen vacancies caused by more Ni^{2+} owing to the difficulty of oxidizing Ni^{2+} to Ni^{3+} at the surface are proposed here to be responsible for the formation of more Li_2CO_3 for shorter calcination times. Li_2CO_3 surface phase is evidenced to favor not