

DEVELOPMENT OF SRI LANKAN VEIN GRAPHITE AND MODIFICATION OF $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3})\text{O}_2$ AS ELECTRODE MATERIALS FOR LITHIUM-ION RECHARGEABLE BATTERIES

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This study was based on developing low-cost and performance enhanced electrode materials for the rechargeable lithium-ion batteries (LIBs). It was carried out by developing Sri Lankan natural vein graphite and $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ based transition metal oxides. Enhancement of purity and surface structure of vein graphite are essential before it is used for anode electrode. A cost effective acid leaching method developed in this study, showed the successful removal of undesirable sulfide and carbonate impurities at very low acid concentration and at low temperature. Mild oxidation and lithium carbonate coating methods were investigated to improve the surface structure of the purified graphite. Chemical oxidation modified the graphite surface by formation of oxidizing species and elimination of structural imperfections. The coating of Li_2CO_3 in graphite surface didn't affect the crystal structure. The anodes fabricated with purified vein graphite shows promising discharge capacity but with poor cycle performance. However, the cycle performance of purified graphite was enhanced due to both surface modification methods. The developed Sri Lankan vein graphite electrode in this study will be a promising anode due to its better electrochemical performance without expensive conductive additive.

Under the cathode development, phase pure $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ and its derivatives were synthesized by glycine nitrate combustion process while keeping the glycine: nitrate ratio as 0.6. All these synthesized powder compositions resulted sub-micron size secondary particles formed by aggregating nano-scale primary particles. Electrical conductivity of all synthesized materials showed semiconducting behavior and improved conductivity in Na, Mg, Ba and Cu-Mg substituted compositions. The $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ showed the highest initial discharge capacity with significantly high rate capability. Overall, the study shows the improvement of electrochemical performance depending on the nature and level of substituting element, and its effect on cation mixing, enhancement of structural stability and electrical conductivity. Among the synthesized materials, the $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-0.04}\text{Na}_{0.04})\text{O}_2$ showed an initial discharge capacity as that of $\text{Li}(\text{Ni}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3})\text{O}_2$ with an improved cycle performance. Cu-Mg co-substituted, $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-(x+y)}\text{Cu}_x\text{Mg}_y)\text{O}_2$ ($x+y = 0.11$) and Ba substituted $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-0.04}\text{Ba}_{0.04})\text{O}_2$ electrodes also showed potentiality of them specially for high voltage LIB applications. The test LIB cells assembled with developed anode and cathode materials showed their potential for practical LIB with promising electrochemical performance.