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Synthesis of Flexible Mn1-x-y(CexLay)O2-δ Ultrathin-Film Device for Highly-Stable Pseudocapacitance from end-of-life Ni-MH batteries

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Abstract : The present work details a three-stage strategy based on selective purification of rare earth oxide (REOs) isolated from end-of-life nickel-metal hydride (Ni-MH) batteries leading to high-yield fabrication of defect-rich Mn1-x-y(Ce_xLa_y)O2-δ film. In step one, major impurities (Fe and Al) were removed from a REE-rich solution. In step two, the resulting solution with trace content of Mn was further purified through electrodeposition which resulted in the synthesis of a non-stoichiometric $Mn_{-1-x-y}(Ce_xLa_{xy})$O2-δ ultra-thin film, with controllable thicknesses (5-650 nm) and transmittance (~29-100%)in which Ce4+/3+ and La3+ ions were dissolved in MnO2-x lattice. Due to percolation impacts on the optoelectronic properties of ultrathin films, a representative Mn1-x-y(CexLay)O2-δ film with 86% transmittance exhibited an outstanding areal capacitance of 3.4 mF•cm-2, mainly attributed to the intercalation/de-intercalation of anionic O2- charge carriers through the atomic tunnels of the stratified Mn1-x-y(CexLay)O2-δ crystallites. Furthermore, the Mn1-x-y(CexLay)O2-δ exhibited excellent capacitance retention of ~90% after 16,000 cycles. Such stability was shown to be associated with intervalence charge transfers occurring among interstitial Ce/La cations and Mn oxidation states within the Mn_{-1-x-y}(CexLay)O2-δ structure. The energy and power densities of the transparent flexible Mn_{-1-x-y}(CexLay)O2-δ full-cell pseudocapacitor device with a solid-state electrolyte was measured to be 0.088 µWh.cm-2 and 843 µW.cm-2, respectively. These values showed insignificant changes under vigorous twisting and bending to 45-180˚, confirming these materials are intriguing alternatives for size-sensitive energy storage devices. In step three, the remaining solution purified further, that led to the formation of REOs (La, Ce, and Nd) nanospheres with ~40-50 nm diameter.

Keywords : spent Ni-MH batteries, green energy, flexible pseudocapacitor, rare earth elements
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