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Development of High Entropy Spinel Oxides Prepared *via* Ball Milling

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In this work, high entropy oxides with MAl_2O_4 spinel structure, i.e. $(Zn_{0.25}Cu_{0.25}Co_{0.25}Mg_{0.25})Al_2O_4$, were successfully synthesized *via* one-step mechanochemical method. Moreover, their lithiated derivates of the final composition: $Li_{0.5}(Zn_{0.125}Cu_{0.125}Co_{0.125}Mg_{0.125})_{0.5}Al_2O_{3.5}F_{0.5}$,$Li_{0.5}(Zn_{0.125}Cu_{0.125}Co_{0.125}Mg_{0.125})_{0.5}Al_2O_{3.5}Cl_{0.5}$ were prepared by mechanochemical reaction between oxide precursors and LiF and LiCl, respectively. The phase evolution as well as the structure of the prepared oxides were controlled by XRD. The morphology of the samples was studied by STEM and HR-TEM and supported by EDX elemental analyses. The STEM images showed nanocrystalline nature of as-prepared samples with average crystalline size in the range of 6 to 17 nm. The analyses of HR-TEM micrographs of samples showed excellent agreement of the average interplanar distances in comparison to the XRD analyses. The EDX analyses demonstrated a homogeneous distribution of the elements. The influence of the preparation method on the oxidation state of elements was investigated by XPS. The electrochemical properties were studied by cyclic voltammetry of Li insertion in the potential window of 0.01–3.0 V vs Li^+/Li at scan rate of 0.1 mV s^{-1}. The charge capacity of $(Zn_{0.25}Cu_{0.25}Co_{0.25}Mg_{0.25})Al_2O_4$ was found to be 54 mAh g^{-1}. The charge capacity of $Li_{0.5}(Zn_{0.125}Cu_{0.125}Co_{0.125}Mg_{0.125})_{0.5}Al_2O_{3.5}F_{0.5}$ was shown to be 71 mAh g^{-1} and for $Li_{0.5}(Zn_{0.125}Cu_{0.125}Co_{0.125}Mg_{0.125})_{0.5}Al_2O_{3.5}Cl_{0.5}$ – 52 mAh g^{-1}. According to the achieved results, the mechanochemical method provides the route towards development of novel high entropy oxides.

Keywords: spinel structure, high entropy oxide, nanomaterials, mechanosynthesis

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