High entropy multication rock salt oxides for lithium ion batteries

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High entropy oxides (HEOs) are solid state inorganic compounds in which entropy, rather than enthalpy, plays a dominant role in stabilizing a single-phase structure at high temperatures. This work has been motivated in part by prior studies of multicomponent alloys in which four or more cations occupy the same crystallographic site in equal proportions, known as high entropy alloys (HEAs), which have superior mechanical properties and high radiation tolerances due in part to high configurational entropy.[1] In the case of HEOs, the first example is the rock salt (Mg0.2Ni0.2Co0.2Cu0.2Zn0.2)O, which has generated a great deal of interest in this class of materials.[2,3] We have recently demonstrated that HEOs prepared by mechanochemical synthesis can be prepared in pure form, and may be useful for catalysis.[4,5] It has also been shown that HEOs are of interest for high ionic conductivity and electrochemical energy storage.[6,7] In this study, we have examined the electrochemical cycling of new high entropy rock salt phases versus lithium, and found an effect of composition on the cycling performance. Samples were prepared through high energy milling of starting binary oxides, which proceed to decompose and then reform pure compounds at high temperature. We have confirmed that entropy plays a role in this transformation for these rock salt HEOs, and that the precise composition has an impact on the temperature and kinetics of pure phase formation; investigations of the synthesis and subsequent decomposition have been conducted in our laboratory using high temperature in-situ X-ray diffraction on a Panalytical diffractometer equipped with an XRK900 stage. STEM/EDS studies on quenched ex-situ samples will be presented that show how elemental segregation occurs as a function of temperature. The results of this study will be highly impactful for the growing community of researchers investigating the design and synthesis of the new class of materials, the high entropy oxides.

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