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Multivalent metal (Mg, Ca, Al) anode batteries as sustainable high-energy alternative to Li-ion

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Abstract

Impending energy transfer from fossil-based energy resources towards renewables multiplied demand for battery energy storage due to requirements connected with electromobility and stationary energy storage. High-demand and limited availability of materials used in contemporary Li-ion batteries have led to several raw materials becoming listed as critical (graphite, lithium, cobalt, nickel, silicon). Hence, several alternative battery technologies (Na-ion, multivalent, sulfur batteries) are being considered as alternatives or supplements to Li-ion.

High gravimetric and volumetric capacity of multivalent metals coupled with their abundance offers an interesting alternative. However, their practical development seemed distant due to limited performance of multivalent electrolytes and suitable cathodes. Multivalent metal anodes are incompatible with conventional Li-ion electrolytes since passive layers on multivalent metals typically block transport of multivalent ions. On the cathode side, high charge density of multivalent ions leads to slow solid-state diffusion, difficult desolvation and inclination towards conversion reaction instead of insertion in conventional inorganic hosts.

The rapid development of multivalent electrolytes in recent years has opened a path towards the exploration of new classes of cathodes, such as organic materials. Organic active materials offer a possibility to circumvent many of the limitations encountered in inorganic hosts due to their soft structure. Additionally, they could be produced from bio-derived feedstock and at lower synthesis temperatures leading to significantly reduced environmental footprint of battery production. In our work, we employ carbonyl-based organic compounds, which show good electrochemical reversibility and long-term cyclability in Mg, Ca and Al batteries. The electrochemical mechanism of organic cathodes is investigated with a complement of *operando* and *ex situ* characterization techniques (IR, EM, XPS) revealing remaining challenges of multivalent battery research.