Summary

Magnesium and calcium batteries are considered promising post-Li battery technologies due to the low redox potentials of Mg and Ca metals (–2.37 V and –2.87 V *vs*. SHE, respectively), high gravimetric and volumetric densities of metal anodes, and natural abundance of Mg and Ca elements, which is an important factor in terms of price and sustainability. The main challenge is the development of suitable electrolytes that enable reversible plating/stripping of Mg/Ca with high Coulombic efficiency in a wide potential window, while also being compatible with the metal anode and cathode materials.

The focus of the thesis is to explore fluorinated alkoxyborate and alkoxyaluminate electrolytes for Mg and Ca batteries. Starting with Mg alkoxyborates, we prepare a series of electrolytes with different degrees of fluorination and investigate the correlations between anion structure and the physicochemical and electrochemical properties of the electrolytes. Enhanced electrochemical performance is observed when boron is replaced by aluminium.

In the second part, MgAlhfip as a state-of-the-art electrolyte comes under the spotlight. We investigate the influence of the MgAlhfip synthesis procedure on electrolyte purity and its electrochemical performance. To improve the purity of underperforming electrolytes, we introduce two approaches: an additional step in the synthesis procedure and the use of scavenging additives.

The potential of MgAlhfip electrolyte for practical applications is examined in terms of its tolerance towards water contamination. To mimic conditions in practical applications, we study the effects of additives under aggravated cycling conditions.

In the last part, we introduce a novel CaAlhfip electrolyte for Ca batteries. We benchmark its physicochemical and electrochemical properties with a model CaBhfip electrolyte. Additionally, we expose the decomposition of solvent on the Ca metal anode as a key issue of Ca electrolytes.

Keywords: Mg batteries, synthesis, fluorinated alkoxyborate electrolytes, fluorinated alkoxyaluminate electrolytes, Ca batteries