

Abstract

Mg batteries are one of the possible candidates for a new generation of batteries, due to non-dendritic Mg deposition, high gravimetric (2206 mAh/g) and volumetric (3832 mAh/cm³) capacity of Mg metal. Unfortunately, Mg metal is plagued by passivation in many electrolyte solutions, so special electrolytes are required to achieve reversible Mg deposition and stripping. In addition, the development of Mg batteries is hindered by precarious insertion of Mg ions into inorganic cathode hosts; because of this we decided to employ organic materials as cathodes in Mg batteries.

Because of the electrophilic nature of the majority of organic materials, we were obliged to use non-nucleophilic Mg electrolytes with high oxidative stability. Initial tests were performed with anthraquinone as a model compound. Like the case of Li organic batteries, dissolution of active materials into the electrolyte was identified as a major issue, causing rapid capacity fading. This issue was solved by grafting anthraquinone and hydroquinone onto graphene nanoribbons. Grafted composites showed improved capacity retention and were used without any added carbon black or binder, but still the overall capacity of the composites was relatively low. Organic polymers are an efficient approach to avoid dissolution of active materials, so we used poly(anthraquinonyl sulfide) as an active material to achieve relatively good capacities with long-term cycling stability. Use of synthesized Mg nanopowder anode instead of Mg foil led to higher initial capacities, close to theoretical ones, without any long-term improvement. Increase of the battery voltage was achieved by moving from anthraquinone to benzoquinone electrochemical moiety inside the polymer.

In the last part of the thesis, we explored the electrochemical activity of anthraquinone in a beaker cell in different electrolytes. Significant changes in electrochemical mechanism and reduction potential were observed when moving from supporting electrolyte to Mg electrolyte. In addition, the effect of water addition on electrochemical behaviour was also explored. The mechanism of electrochemical activity was explored through use of infrared spectroscopy. Reduction of carbonyl bonds was confirmed as a mechanism of electrochemical activity in the case of both anthraquinone and poly(anthraquinonyl sulfide).

Keywords: Mg batteries, organic cathode materials, non-nucleophilic electrolytes, polymers, electrochemical reaction mechanism