ABSTRACT

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It is now clear that we are entering in an era where energy storage technologies, other than the Li-ion based, will become prominent. This will mitigate the cost and geopolitical issues related to Li-ion-based storage technologies. Amongst the storage technologies, Al-ion technologies are the newest players, which have the capacity to bring a paradigm shift in the way we talk about the cost, maintenance, availability, disposal, and recycling strategies of batteries. The widely abundant aluminium reserves will make the processing and delivery of such storage systems locally possible. This makes them ideal candidates for large-scale usage. Currently, the main challenge is to identify viable electrode materials and their coupling with an appropriate electrolyte to achieve high performing aluminium-ion batteries (AIBs).

Although Al^{3+} exhibit a small radius, their large hydrated radius impedes insertion/ extraction into/from the host structure. Further, the strong electrostatic interactions of Al-ions with the host structure and higher charge density makes their insertion/extraction more difficult. This leads to sluggish kinetics, high over-potential, and eventual collapse of the host structure. Therefore, desired materials that would facilitate efficient intercalationdeintercalation should have wider channels. More recently, a new class of materials has been reported, which have much more open network like structure. These are zeolite imidazole based framework (ZIF) structures. In this thesis, the intercalation chemistry of Al-ion in ZIF 67 crystal structure is reported. It is shown that Al^{3+} preferentially intercalates into the crystal structure of ZIF 67 in AlCl₃ electrolyte. The Al^{3+} storage capacity of ZIF 67 is estimated to be ~ 288 mAh g⁻¹, with excellent cycling stability. To induce a quantum jump in the specific capacity value, a strategy based on the use of redox additive modified electrolytes is proposed. The fruitfulness of the strategy is established by using K₃Fe(CN)₆ redox additives in AlCl₃ electrolyte. The discharge capacity jumps from ~ 288 mAh g⁻¹ to ~ 1452 mAh g⁻¹ when the pristine AlCl₃ electrolyte is modified with optimized concentration of K₃Fe(CN)₆.

Incorporating conducting polymer into active electrode materials is another approach to increase the specific capacity of the material. Hence, the work also proposes the use of polypyrrole (PPy) decorated ZIF 67 composite as a high performing electrode for aqueous AIBs. Performance improvement of >50% is shown to prove the correctness of the hypothesis.