Abstract

Ion conducting polymers and ceramics are the two widely studied solid electrolytes for four decades. However, each of them has its advantages and disadvantages. On one hand, despite high grain interior conductivity, ceramic ionic conductors are not applicable to devices due to large grain boundary impedance. On the other hand, solid polymer electrolytes are flexible, establish smooth contacts at the interface, but due to inadequate ionic conductivity not found suitable for the devices. With an approach that their combination, a hybrid electrolyte may give desired solid electrolytes, the present work is planned. Thus two approaches viz. 'polymer in ceramic' and 'ceramic in polymer' have been used to developed novel Na^+ and Li^+ ion-based composites. Thesis, therefore deals with the synthesis and thorough characterization of Li^+ and Na^+ ion conducting hybrid polymer-NASICON nanocomposites. These were synthesized by solution casting and one-pot synthesis routes. The electrical, structural, and thermal properties of the composites are rigorously studied for various compositions. Further, the thesis explores the conductivity-structure correlation using sophisticated techniques viz. synchrotron-based XAS and XPS and proposes a generalized ion transport mechanism for these composites. Finally, it also explores possibility of use of these composites in all-solid-state supercapacitors. The first two chapters deal with the literature survey, gaps, statement of problems, objectives, and experimental techniques.

The systematic electrical, thermal and structural analysis on Na^+ and Li^+ ion conducting polymer-NASICON electrolytes are discussed in the subsequent two chapters (3 and 4). The composites are found to be highly conducting approaching ~ 10^{-4} $\Omega^{-1}cm^{-1}$ at 40°C with appreciable thermal cycling stability up to 100°C. Chapter 5 discusses the results for structural investigations describing coupling/decoupling of cations with ether oxygen of PEO. A nice correlation between structure and electrical transport could be established to define a generalized ion transport mechanism.

The potential candidature of these composites in all-solid-state electric double layer and pseudo supercapacitors is finally explored in the current thesis (Chapter 6). The cells exhibit a maximum energy storage capacity of ~ 102 F-g^{-1} per electrode at a current density of 1 A-g^{-1} with appreciable cycling. The composites developed in the present work are found to be potential candidates for electrolyte/electrode applications. The last chapter concludes the work and proposes future scope.