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Title : CHARACTERIZATION OF ZnO/MnO₂-DOPED Li₂CO₃-LiI BINARY SOLID ELECTROLYTE FOR LITHIUM ION PRIMARY CELL

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The continuity in all-solid-state lithium ion battery development is very important considering the growing demand for a reliable and safety improved energy storage device. In this work, a solid state electrolyte composed of Li₂CO₃ and LiI was successfully prepared using mechanical milling method followed by low sintering process. The optimized sintering condition of the solid electrolyte was at 60°C for 24 hours. From the measurement, the xLi₂CO₃-yLiI (x = 80, y = 20) solid electrolyte displayed the highest electrical conductivity of 1.03x10⁻³ Scm⁻¹ at room temperature. The dissociation of LiI has provided supplemental charge carriers for ions transport in the solid, which much higher than that of the pure Li₂CO₃ sample. A noteworthy increase of ionic conductivity up to 1.75x10⁻³ Scm⁻¹ was observed at room temperature with the presence of ZnO filler. The enhancement in electrical conductivity could be due to the interaction between ionic species and Lewis acid-base group on the filler surface and hence, improve the dissociation rate of ions in solid. Incorporation of 9 wt.% MnO₂ has successfully amplified the overall conductivity of the sample (3.87x10⁻¹ Scm⁻¹) by increasing the available charge carrier concentration and newly established percolation network for the migration of ions. The interactions between ions and filler's surface groups were also responsible for conductivity enhancement in solid by providing coordination sites for ions transport process. FTIR has revealed the complexation between binary electrolytes and oxide fillers. This could be ascribed to the shift of metal-oxygen absorption band in the samples. Additionally, the increase in

absorption intensity of metal-iodine (Li-I) peak signified the increment of charge carrier concentration as a function of fillers. The analysis by XRD technique has confirmed the structural transformation in the solid based on the formation of new peaks and also peak splitting phenomena. The ceramic filler addition has resulted in structural modification of solid in order to accommodate more ions. Subsequently, the increase in defect formations was expected and higher migration rates of ions via Frenkel and Schottky defect were possible. The temperature-dependent analysis demonstrates that the Li₂CO₃-LiI, Li₂CO₃-LiI-ZnO and Li₂CO₃-LiI-MnO₂ samples follow Arrhenius behavior. This was explained by the linear function of conductivity towards temperature suggesting that the systems were thermally activated. Meanwhile, the activation energy was found to be inversely proportional to the total conductivity of the samples. The discharge characteristic of the lithium battery has showed a discharge capacity of 165 µAh with stable discharge plateau of 4 hours at discharge rate of 10 µA. The incorporation of ZnO filler has prolonged the stability of discharge plateau up to 5 hours with discharge capacity of 234 µAh. The total discharge period was extended from 16 to 23 hours with the presence of ZnO filler. Meanwhile, a total discharge period of 25 hours with capacity of 255 µAh signified that the incorporation of MnO₂ filler has significantly improved the electrochemical stability of the cell. Additions of ceramic fillers have effectively enhanced the stability of electrochemical cell while providing an additional coordination sites for migration of ions in solid.