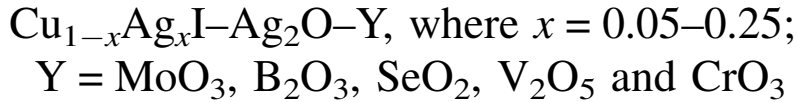


All-solid-state cells based on solid electrolyte systems



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Abstract

All-solid-state cells of the configuration (–)Ag + SE//SE//I₂-phenothiazine + C(+) using the best conducting compositions of the solid electrolyte systems, namely, Cu_{1–x}Ag_xI–Ag₂O–Y where x = 0.05, 0.1, 0.15, 0.2 and 0.25, Y = MoO₃, B₂O₃, SeO₂, V₂O₅ and CrO₃, as the electrolytes were fabricated. Discharge, polarization and power characteristics of these cells were also evaluated. The open circuit voltage values of these cells were in the range 620–635 mV. The stability of these cells has been indicated by the constancy of their OCV over a period of 6 months. The polarization and discharge studies on these cells have shown that typical cells based on the electrolytes with Y = B₂O₃, SeO₂ and V₂O₅ would possess discharge capacities of 12.84, 3.76 and 5.05 mA h and specific energy of 6.55, 1.81 and 2.77 W h kg^{–1}, respectively. The solid electrolytes have good electrochemical stability and compatibility with the Ag/Phenothiazine–I₂ electrode couple thus offering their suitability of application in microwatt power sources.

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1. Introduction

During the last quarter of the 20th century, many researchers have shown an increased interest in the development of new battery materials having appreciable energy conversion characteristics without pollution. The importance of developing new solid electrolyte materials for a variety of applications has also been realized in recent years [1]. Owing to several advantages associated with all-solid-state batteries, in safety and storage aspects, such as long shelf-life, lack of corrosion, spill-proof, etc., over those batteries having fluid electrolytes, the search for appropriate solid electrolyte materials and solving certain inherent interfacial problems connected with electrode/electrolyte interfaces appear to remain an important task of battery research [2–7]. The development of new solid substances for both electrodes and electrolytes is expected to create opportunities for new types of electrical energy generation and storage systems, which may themselves, in turn, revolutionize many industrial areas.

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Following the development of the first solid state cell based on Ag^+ ionic conductor with the configuration of $\text{Ag}/\text{Ag}_3\text{SI}/\text{C} + \text{I}_2$, many silver batteries were developed with various electrolytes and modified electrodes [8–12], but the discharge capacities and specific energies are less and still to be improved. The capacity and specific energy are mainly dependent on the conductivity of the electrolyte material used. Search for the better cathode materials which replace iodine in the silver anode batteries is also continuing worldwide [10,13–15]. Silver-ion conducting superionic materials in the systems $\text{Cu}_{1-x}\text{Ag}_x\text{I}-\text{Ag}_2\text{O}-\text{Y}$ where $x = 0.05, 0.1, 0.15, 0.2$ and 0.25 , $\text{Y} = \text{MoO}_3, \text{B}_2\text{O}_3, \text{SeO}_2, \text{V}_2\text{O}_5$ and CrO_3 , having electrical conductivities in the order of 10^{-2} to 10^{-3} S cm^{-1} at 295 K, in the solid state, were synthesized and characterized in our laboratory [16–20]. This paper deals with the discharge, polarization and power characteristics of the all-solid-state cells fabricated using the above-prepared superionic solids as electrolytes.

2. Experimental

2.1. Materials and fabrication of cells

In our earlier work involving preparation and characterization of new solid electrolytes in the systems $\text{Cu}_{1-x}\text{Ag}_x\text{I}-\text{Ag}_2\text{O}-\text{Y}$ where $x = 0.05, 0.1, 0.15, 0.2$ and 0.25 , $\text{Y} = \text{MoO}_3, \text{B}_2\text{O}_3, \text{SeO}_2, \text{V}_2\text{O}_5$ and CrO_3 , the best conducting compositions were identified to be $30\text{Cu}_{0.75}\text{Ag}_{0.25}\text{I}-35\text{Ag}_2\text{O}-35\text{MoO}_3$ (System I), $40\text{Cu}_{0.95}\text{Ag}_{0.05}\text{I}-45\text{Ag}_2\text{O}-15\text{B}_2\text{O}_3$ (System II), $(40\text{Cu}_{0.75}\text{Ag}_{0.25}\text{I}-30\text{Ag}_2\text{O}-30\text{SeO}_2$ (System III), $40\text{Cu}_{0.75}\text{Ag}_{0.25}\text{I}-40\text{Ag}_2\text{O}-20\text{V}_2\text{O}_5$ (System IV), and $35\text{Cu}_{0.95}\text{Ag}_{0.05}\text{I}-32.5\text{Ag}_2\text{O}-32.5\text{CrO}_3$ (System V), exhibiting electrical conductivities of 1.7×10^{-3} [16], 2.1×10^{-3} [17], 1.4×10^{-3} [18], 1.3×10^{-2} [19] and 1.1×10^{-3} [20] S cm^{-1} , respectively, at 295 K. In the earlier work, it was also found that the silver ionic transport numbers (t_{Ag^+}) of all these materials are greater than 0.95 which confirmed the superionic nature of these material compositions [16–20]. These compositions were used as solid electrolytes for the fabrication of silver anode all-solid-state cells. Cells containing the above compositions of Systems I, II, III, IV, and V as the electrolytic media were designated as Cells A, B, C, D, and E, respectively.

The anode materials were prepared by mixing pure silver powder (the anode element) and the corresponding solid electrolyte specimen in the weight ratio 2:1. The solid electrolyte was added to silver powder to improve the interfacial contacts. Since pure iodine may sublime and lead to self-discharge of the cell, a charge-transfer complex (CTC) namely iodine-phenothiazine CTC (I_2 , phenothiazine in the weight ratio of 3:2) was chosen as the cathode material. Interestingly, I_2 -CTCs are found to offer promising cathodes for solid-state batteries [13,21,22]. A small quantity of graphite powder was also incorporated in the cathode compartment in order to reduce the cathode polarization effects which generally result in loss of power output.

Circular discs of 1 cm diameter and about 2 mm thickness were prepared by pressing the anode, solid electrolyte (SE) and cathode layers together. Anode material (0.25 g) was first poured into a die and small pressure was applied to form a layer. The electrolyte material (0.5 g) was added and a small pressure was applied. Finally, the cathode material (0.25 g) was poured over the electrolyte layer in the die and the three layers were pressed at 4 tonnes cm^{-2} , to obtain a pellet of a solid-state cell having the configuration of $(-)\text{Ag} + \text{SE}/\text{SE}/\text{C} + \text{I}_2$ -phenothiazine CTC (+), and placed in between two silver foils. The anode face was directly attached to the foil while the cathode face was held via graphite disc, acting as a current collector. The entire cell assembly was mounted between two thin ebonite plates through screw arrangements and electrode leads taken out for measurements. The cell was covered with epoxy resin in order to prevent sublimation of iodine.

2.2. Evaluation of cell parameters

Identical configurations of Cells A, B, C, D, and E were employed for the detailed discharge and polarization studies. The open circuit voltage (OCV) of the cells were measured at room temperature as a function of time without load ($R_L = \infty$), using a high input impedance multi-meter, in order to evaluate their stability and shelf life. Short circuit current (I_{SC}) of the cells were also measured with an EZ multi-meter, model DM-531T. For obtaining the discharge characteristics of individual cells, both the cell voltage and current were monitored as a function of time when a finite constant resistive load was used. The discharge capacity was obtained by integrating the current for the entire discharge time (using Microcal Origin program). All the cells were discharged until the cell voltage reached 70% of the initial voltage, which was chosen as the cut-off value. The energy density was calculated by multiplying the cell

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