



ORIGINAL ARTICLE

Ion transport properties of magnesium bromide/dimethyl sulfoxide non-aqueous liquid electrolyte



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ABSTRACT

Nonaqueous liquid electrolyte system based dimethyl sulfoxide DMSO and magnesium bromide (MgBr_2) is synthesized via 'Solvent-in-Salt' method for the application in magnesium battery. Optimized composition of $\text{MgBr}_2/\text{DMSO}$ electrolyte exhibits high ionic conductivity of 10^{-2} S/cm at ambient temperature. This study discusses different concentrations from 0 to 5.4 M of magnesium salt, representing low, intermediate and high concentrations of magnesium salt which are examined in frequency dependence conductivity studies. The temperature dependent conductivity measurements have also been carried out to compute activation energy (E_a) by least square linear fitting of Arrhenius plot: $\log \sigma - 1/T$. The transport number of Mg^{2+} ion determined by means of a combination of d.c. and a.c. techniques is ~ 0.7 . A prototype cell was constructed using nonaqueous liquid electrolyte with Mg anode and graphite cathode. The Mg/graphite cell shows promising cycling.

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Introduction

Due to gasoline high cost and probability of oil depletion, demands for renewable energy sources increased. Rechargeable batteries are the most important sources of renewable energy especially batteries to run electric cars. But the dream of the spread of the electric car is still far because we need battery systems suitable for load leveling applications. They have to be large, safe, and made of cheap and abundant compo-

nents. Most importantly, it is mandatory that they have a very prolonged lifecycle [1,2].

The lithium (Li) battery used as a power source because of its high specific power and high energy density. However, high demand for Li battery tends to make increase in Li price due to geographically limitedness in the earth crust [1]. As an alternative to lithium, magnesium has been foregrounded. Magnesium batteries have recently attracted great interest due to their high energy density and environmentally friendly components, coupled with magnesium's low cost ($\sim \$2700/\text{ton}$ for Mg compared to $\$64,000/\text{ton}$ for Li) and abundance in the earth's crust ($\sim 13.9\%$ Mg compared to $\sim 0.0007\%$ of Li) [3–5].

The kinetically sluggish Mg intercalation and diffusion in cathode materials and the incompatibility between anode and electrolyte due to the high polarizing ability of the Mg^{2+} cation are the major obstacles that prohibit Mg batteries from

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commercialization [6–8]. Thus, the great challenge in the commercialization of Mg batteries is the development of an electrolyte which is stable in contact with the electrode materials, does not form a blocking layer, low cost and has a wide electrochemical window [9].

The development of magnesium electrolytes is considered the most important challenge for the commercial application of rechargeable magnesium (Mg) batteries because electrolyte properties govern battery performance and determine the class of cathodes utilized [10]. Aurbach et al. [8] developed a prototype Mg cell using Mo_6S_8 Chevrel phase cathode, $\text{Mg}(\text{AlCl}_2\text{-EtBu})_2$ /tetrahydrofuran electrolyte with a little fade of capacity. However, the development of rechargeable Mg batteries still hindered by using electrolyte suffers from the use of very volatile solvents such as THF, and organohaloaluminate electrolytes with highly corrosive nature, high cost, high air sensitivity, and low anodic stability which limit the choice of cathodes [11,12].

Magnesium bromide (MgBr_2) is the fast conducting salt in a number of crystalline and amorphous materials. Furthermore, Dimethyl sulfoxide (DMSO) is an organosulfur compound with Melting point 19°C and Boiling point 189°C and the formula $(\text{CH}_3)_2\text{SO}$. This colorless liquid is an important polar aprotic solvent that dissolves both polar and nonpolar compounds and is miscible in a wide range of organic solvents as well as water. Recently, Peng et al. [13] report that a Li- O_2 cell composed of a DMSO-based electrolyte and a NPG electrode can sustain reversible cycling, retaining 95% of its capacity after 100 cycles and having >99% purity of Li_2O_2 formation at the cathode, even on the 100th cycle, and its complete oxidation on charge. Although, to date, there is no works on DMSO-based electrolyte for magnesium batteries have been done. This prompted us to try the test dimethyl sulfoxide/ MgBr_2 as an electrolyte for magnesium battery.

Upon the above considerations, a key obstacle to obtain the better performance, new electrolyte system for rechargeable magnesium batteries should be developed. Therefore, in this article, new classes of non-aqueous liquid ‘Solvent-in-Salt’ electrolytes, $\text{MgBr}_2/\text{DMSO}$ are discussed. The electrolytes were characterized by using XRD and impedance spectroscopy. Generally, this work mainly focuses on studies of ionic conductivity and Mg^{2+} ion transference with the aim of gaining understanding in ionic conduction of magnesium in this electrolyte system. With non-aqueous electrolyte optimum composition, Mg/Graphite cell is assembled, and its cycling performances will be briefly examined to evaluate the applicability of the electrolyte to solid-state magnesium batteries.

Experimental

All tested electrolytes consisted of a magnesium bromide (492 ppm of moisture, Sigma) with different concentrations, $x = 0.0, \dots, 0.55$ M in dimethyl sulfoxide (Loba Chemie). The resulting solutions were stirred for 2 h at room temperature.

The XRD patterns of MgBr_2 , $\text{MgBr}_2/\text{DMSO}$ precipitated solid product were taken using Rigaku diffractometer type RINT-Ultima IV/S. The diffraction system based with Cu tube anode with voltage 40 kV and current 40 mA.

Conductivity measurements of $\text{MgBr}_2/\text{DMSO}$ liquid electrolyte were performed using impedance method. Electrolyte

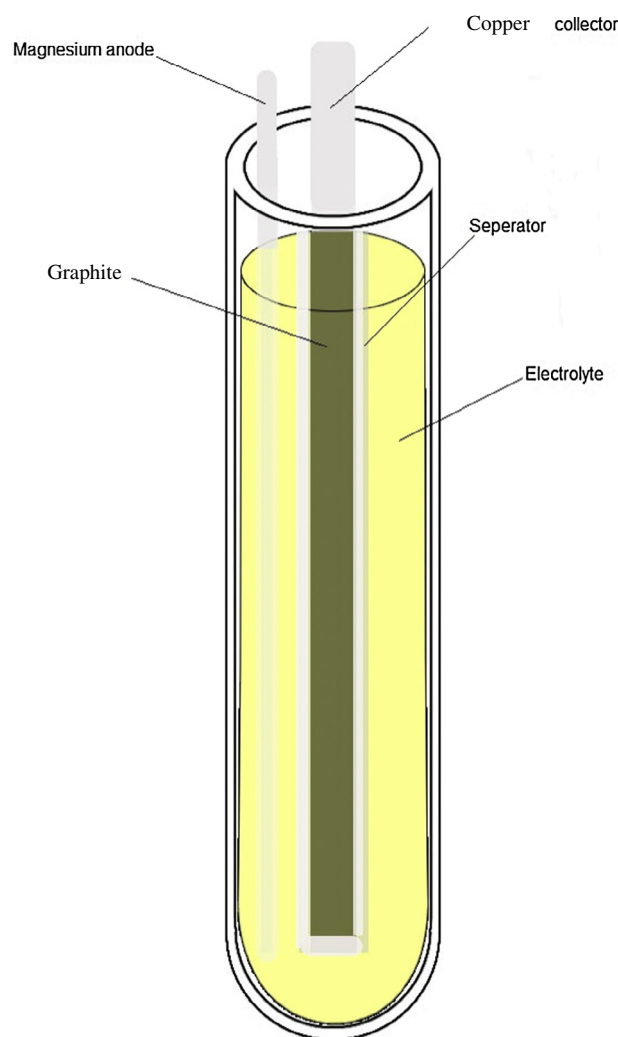


Fig. 1 Schematic design of the laboratory cell.

samples were put into a conductivity cell between two similar aluminum electrodes. The whole assembly was placed in a furnace monitored by a temperature controller. The rate of heating was adjusted to be 2 K min^{-1} . Impedance measurements were performed on Gwinstek LCR-8110G in the frequency ranging from 20 Hz to 10 MHz at different temperatures.

Slurry was obtained by mixing 1 g graphite, 0.1 g magnesium sulfate and 0.2 g PVA binder using magnetic stirrer hot plate (60°C) for 2 h. The cathode (reduced electrode) is prepared by coating pole of stainless steel by 0.1 g of this slurry in dry atmosphere at 100°C for two hours. Entek PE membrane separator has been used. The whole assembly was shown in Fig. 1. The tube cell was discharged at room temperature on a multi-channel battery test system (NEWARE BTS-TC35) with the charge/discharge time of 2/2 h and 10 min rest. The current density was $10\ \mu\text{A}/\text{cm}^2$.

Results and discussion

Plausible reaction model between MgBr_2 and DMSO is shown in Fig. 2a. In the hexahydrate ($\text{MgBr}_2 \cdot 6\text{H}_2\text{O}$) the magnesium cation is surrounded only by water molecules, forming distinct octahedral [14]. By dissolving $\text{MgBr}_2 \cdot 6\text{H}_2\text{O}$ in DMSO two

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