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Full Length Research Paper

Mixed electrolyte for an improved battery system

Don-Lawson Chioma

Department of Science Laboratory Technology, School of Science and Technology Captain Elechi Amadi Polytechnic Rumuola, Port-Harcourt Rivers-State, Nigeria.
Author E-mail: chioma.don-lawson@portharcourtpoly.edu.ng

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ABSTRACT: The chemistry of mixed organic solvents formed by mixing dimethylformamide (DMF) and propylene carbonate (PC) solvents with magnesium per chlorate $Mg(ClO_4)_2$ salt was investigated. The mixtures were prepared and their physical and thermodynamic properties were determined. Conductivity, dielectric constant, and cell voltage are examples of these properties. The cells of binary mixtures with Mg^{2+} ion-containing electrolytic solutions were also investigated. Molar conductivity measurements at 25ºC revealed that mixed systems have greater electrochemical stability and better properties than pure solvents. Ion-solvent and solvent-solvent interactions influenced the properties of the mixed electrolytes. The results were interpreted based on the intermolecular interactions between the system's component molecules. The organic solvent mixing ratio for optimal battery performance has been established to be between 50-70 percent PC for the battery system because the system gave the highest conductivity and electromotive force at this range.

Keywords: Electrolyte, dielectric system, cell voltage, binary mixture

INTRODUCTION

The suitability of a solvent or solvent mixture as an electrolyte necessitates the presence of a high dielectric constant and a low viscosity, which are rarely combined in a single solvent. Because of their high energy density, lithium-ion batteries have received more attention than any other rechargeable battery in the past. In recent years, the battery market has shifted rapidly toward other new-generation batteries, such as battery electric vehicles (BEV) and plug-in hybrid electric vehicles (PHEV). Researchers are developing a magnesium-based battery capable of storing double the energy stored by lithium ion cells, which would translate to more mileage on a single charge as part of global automakers' efforts to improve ways of powering electric cars (Ichitsubo et al., 2011). Toyota Motor Corporation, led by Jeffrey Makarewicz, is one example (Motin, 2007). The purpose of this study was to determine the development and applicability of binary systems of dimethylformamide (DMF) and propylene carbonate (PC) in the development of high energy density magnesium ion batteries. Binary mixtures, with one component chosen for its high dielectric constant and the other for its low viscosity, are commonly used to create high conductivity electrolytes for batteries in order to achieve a balance between these two properties. Carbonates are typically chosen for their high dielectric constant and low viscosity (Thirumaran and Sathish 2011). Since the discovery of non-aqueous electrolytes, a wide range of polar solvents has been studied, with the majority of them belonging to the organic esters or ether families. However, the chemistry of mixed organic solvents has piqued the interest of many researchers because these solvents have properties that may be more useful than pure solvents in reducing or eliminating high industrial pollution and environmental destruction. Because a high dielectric constant and a low viscosity cannot coexist in a single solvent. Binary mixtures, with one component chosen for its high dielectric constant and the other for its low viscosity, are commonly used to create high conductivity electrolytes for batteries in order to achieve a balance between these two properties (Nageshewar and Anil, 2010). Magnesium is widely used in batteries, laptops, and mobile phones due to its low weight and good mechanical and electrical properties. Conductivities, melting points, boiling points, dielectric constant, density, flash point, resistivity, transport number, and solubility are some of the properties that can be used to characterize organic solvents. Magnesium ion battery is one of the improved types of battery innovation, but it has not been widely established in comparison to previous work on Nickel-Iron battery (NiFe) and lithium-ion battery. Batteries are technical, the combination of two or more cells, electrically connected to transform chemical energy to electrical energy (Zamir, 2004). The cell is made up of two electrodes, the positive electrode (cathode) and a negative electrode (anode) which are immersed in an electrolyte. Batteries are available in a variety of sizes, shapes, voltages, and configurations (Bin et al., 2013). The battery market is rapidly shifting to new-generation batteries such as battery electric vehicles (BEV) and plug-in hybrid electric vehicles (PHEV). Researchers are developing a magnesium-based battery capable of storing double the energy stored by lithium ion cells, which would translate to more mileage on a single charge as part of global automakers' efforts to improve ways of powering electric cars. Toyota Motor Corporation, for example, is led by Jeffrey Makarewicz (Motin, 2007). The purpose of this study was to determine the development and applicability of a binary system of dimethylformamide (DMF) and propylene carbonate (PC) in the development of high energy density magnesium ion batteries.

MATERIAL AND METHOD

Materials

The solvents, Propylene Carbonate (99.5%), Dimethylformamide (99.5%), and the salt, Magnesium Perchlorate ($Mg(ClO_4)_2$ dried (99%) were all obtained commercially.

Sample preparation

Binary mixtures of DMF/PC were prepared in varying proportions of 100, 70, 50, 40, 30,20,,10 and 0% of PC, corresponding to mole fractions of 1.0,0.680, 0.477, 0.378, 0.280, 0.185, 0.091 and 0.00 of PC. The working temperatures were 25, 40, 50, 60, and 70ºC. Different concentrations of 0.1M, 0.5M and 1.0M of $Mg(ClO4)_2$ solutions of the binary system were also prepared. The weighing were done on Adam AAA electronic balance with a precision of ±0.001g the capillary and K= viscometer constant. The conductivities were measured at different concentrations with the use of a conductivity meter and a multimeter for the cell voltage.

RESULTS AND DISCUSSION

Electrochemical Cell Voltage (volts)

Cell potentials depend on the concentration of the solution and show the output of the battery system The values of the measured electrochemical cell voltage values are shown in (Table 1) and indicates an improved system due to the mixing. The values obtained are also attributed to the combined influence of low viscosity of DMF and high dielectric constant of PC and ionic mobility. The voltage was highest at 70% of PC showing the highest output of the battery system with a value of 1.02volts.

Conductivity

The conductivity of an electrolyte solution depends on the concentration of the ionic species. The results of the conductivity measurements of solutions $Mg(ClO_4)_2$ in binary mixtures of PC and DMF of different %PC (Table 2) show an increase in molar conductivities with a decrease in % PC. This increase could be attributed to both the high dielectric constant of PC and the low viscosity of DMF. Thus, it may be inferred that the combination of the dielectric constant and viscosity in solvent results in the conductivity of its solution. Such observations have been reported earlier by (maduelosi *et al*., 2014). Maximum conductivity was observed between the 50-70% PC. These results show that ion-solvent and solvent-solvent interactions contribute to the improvement of conductance. This was also observed by (Obunwo and Izonfuo, 1999). It can thus be suggested that an increase in ionic conductivity is achievable in binary mixtures of solvents where one of the components has a high dielectric constant and the other solvent a low viscosity. The highest conductivity values were seen at the maximum concentration of the salt (1.0M). This also explains that an increase in the concentration of the salt increases the conductivity of the system to an extent that is, as long as the ionic mobility is not hindered. According to Nwokobia *et al.(*2013), a net increase in ion conductivity is achieved when solvents of varying properties are mixed. This is described by the viscosity of the electrolytic media.

Dielectric Constant (ε)

Table 3 shows changes in the values of the dielectric constant of the system on mixing.

Table 1: Cell Voltages (Volts) of solution of binary mixtures of PC/DMF and salt.

%PC	E_{Cell} (Volts)
100	0.56
70	1.02
50	0.98
40	0.62
30	0.58
20	0.49
10	0.51
0	0.48

Table 2: Molar conductivities of solutions of $Mg(ClO_4)_2$ in binary mixtures of PC/DMF at various concentrations.

% PC	without salt	0.1M (Scm^2mol^{-1})	0.5M Salt (Scm^2mol^{-1})	1.0M (Scm^2mol^{-1})
100	0.025	2.93	9.23	12.06
70	0.081	3.88	17.64	38.44
50	0.078	3.71	16.90	36.70
40	0.054	3.14	16.99	36.58
30	0.011	3.68	11.91	30.16
20	0.023	2.86	16.31	35.13
10	0.033	2.92	15.30	34.96
0	0.012	2.95	16.49	35.11

Table 3: Dielectric constant of pure and various mixtures OF PC/DMF AT 25ºC.

PC	Dielectric constant
100	36.7
70	70.65
50	69.85
40	53.68
30	56.51
20	59.34
10	62.17
0	65.00

This indicates an improvement in the properties of a solvent when mixed with another. The increase in the values of the dielectric constant of the mixtures with the mole fraction PC is implied since the dielectric constant of a mixture is just an additive factor. Molecular interactions that affect the density and viscosity of a mixture do not necessarily have any effect on the dielectric constant this could mean that interactions and dissociation of solute-solvent are high and there is a minimum ion-pair formation. For a solvated ion to migrate by the influence of an electric field, it must not be allowed to form close ion pairs with its counter ions by the solvating solvent (William, 2005). The efficiency of the solvent molecule in shielding the inter-ionic columbic attraction is determined by the dielectric constant of the solvent. Obviously, with the higher dielectric constant values obtained in (Table 3), the magnesium ions would have a high probability of staying free at a given concentration, and ion association will subsequently be less likely to occur. Studies have shown that solvents with a dielectric constant in the range 20-40 show extensive ion-pair formation (Obunwo and Izuonfo 1999). The attraction between solute and solvent is essentially that of ion-dipole interaction which depends mainly on the ion size and polarity of the solvents. The strength of such interaction also depends on the charge and magnitude of the distance between the ion and dipolar molecule.

Conclusion

Results of experimental measurements of the properties of the mixed solvents have provided significant information regarding the state of affairs in the mixture. The results obtained for solutions of $Mg(ClO_4)_2$ in binary mixtures of PC and DMF at the varying composition of

PC show that the values of the properties studied have intermediate between those of the pure solvents.. The molar conductivity in the mixed solvents is much higher than that in the pure solvents. The mixing ratio of the mixed PC-DMF system for optimum battery performance has been established to be between 50-70% PC for the system studied. .The salt concentration with the highest molar conductivity is 1.0M $Mg(ClO_4)_2$.

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