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RESEARCH ARTICLE

THERMODYNAMICS OF ELECTROLYTE SYSTEM WITH BINARY MIXTURE OF 1, 2-DIMETHOXYETHANE AND ETHYLENE CARBONATE

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ABSTRACT

Studies on the thermodynamics of binary electrolyte system containing mixture of 1,2-dimethoxyethane (DME) and ethylene carbonate (EC) at various temperatures (25, 40, 50, 60 and 70 °C) and mole fractions (1.000, 0.7197, 0.5982, 0.4905, 0.3910, 0.2997 and 0.2157 of DME) was carried out. The parameters (density, viscosity, excess volume, free energy of activation of flow, excess viscosity, d-parameter, excess Gibbs free energy, change in free energy, enthalpy and entropy) of mixing were examined. Generally, the values of density, viscosity, excess viscosity and free energy of activation of flow increased with decrease in mole fraction of DME. Result analyses showed that the trend in values of excess volume, change in free energy and entropy of mixing for mole ratios of DME is: $0.7197 > 0.5982 > 0.4905 < 0.3910 < 0.2997 < 0.2157$; while that of d-parameter is: $0.7197 < 0.5982 < 0.4905 > 0.3910 > 0.2997 > 0.2157$. In the whole temperature range studied, the negative values of change in free energy of mixing indicate the spontaneous and exergonic nature of the system; whereas the positive values of entropy of mixing suggests the disorderly behavior of the binary electrolyte system. The enthalpy of mixing is zero, which affirms the involvement of non-bonding interactions.

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INTRODUCTION

Electrolyte systems comprise of electrolyte which are molten ionic compounds or solutions having solute particles dissociated in a suitable solvent medium to produce solvated positive and negative charged mobile species that permit the in and out flow of electric current. These solutions have been adopted successfully by Technologists, Scientists and Engineers for experimental studies, process design and theoretical advances in fields associated with electro-dialysis, electro-coagulation (Suneetha *et al.*, 2015) electro-flotation, electro-deionization (Dzyazko *et al.*, 2016), electro-deposition (Fu and Wang, 2011), electro-oxidation (Chen, 2004), electro-gravimetry, coulometry, voltammetry, amperometry, potentiometry, conductometry, titration (Uzoukwu, 2009) and so on. In electrochemical processes therefore, single or multi-component solvents are tailored to the needs of specific electrolyte solution for purpose of cell durability. The thermodynamics of mixing 1,2-dimethoxyethane and ethylene carbonate in an electrolyte system was investigated at various temperatures and concentrations.

In the design of high density chemical source of current, properties of electrolyte solution such as viscous nature, dielectric behavior, dissociation strength, ionic conductivity, thermal stability, thermodynamic parameters and kinetic characteristics are kept on the front burner of the plan. Knowledge of these properties is essential for understanding and predicting the chemical and phase equilibria in electrolyte systems with varying solvent environment (Wang *et al.*, 2002 and 2006; and Jaworski *et al.*, 2011). Solutions of mixed electrolyte exhibit non ideal behavior as a result of forces associated with positive and negative solvated ions, intra- and inter-bonding of species, geometric orientation and size of species. The purpose of this study is to provide insight into the mixing tendencies of different mole proportions of 1,2-dimethoxyethane and ethylene carbonate solvent molecules through the interpretation of resolved parameters (density, viscosity, excess volume, free energy of activation of flow, excess viscosity, d-parameter, excess Gibbs free energy, change in free energy, enthalpy and entropy) of mixing; and to implement the experimental resolutions in the ultimate process design and production of high capacity power source.

Experimental

The 1,2-dimethoxyethane (> 99 %) and Ethylene carbonate (98 %) were obtained from Sigma Aldrich, and used as supplied

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(Ibezim-Ezeani *et al.*, 2017a). Measurements of pure and mixed solvents were performed with 10 ml Pycknometer bottle at 25, 40, 50, 60 and 70 °C using analytical electronic balance (Adam AAA balance with a precision of ±0.001g). Varying proportions of binary mixtures of the DME and EC solvents were prepared with mole fractions: 1.000, 0.7197, 0.5982, 0.4905, 0.3910, 0.2997 and 0.2157 of DME. The viscosity of the solvent mixtures were measured based on ASTMD 445-74 method using an Ubbelholde type glass capillary tube suspended level viscometer in a thermostatic water bath at 25, 40, 50, 60 and 70 °C. Distilled water was used in calibrating the viscometer. Replicate flow time was taken for each sample and overall accuracy estimated to be ±0.003 cP. Parameters of mixing such as density (equation 1), viscosity (equation 2) (Dash and Swain, 1994), molar volume (equation 3), excess volume (equation 4) (Dikio *et al.*, 2012), d-parameter (equation 5) (Jha *et al.*, 2010), excess Gibbs free energy of activation of flow (equation 6) (Dash and Swain, 1994), excess viscosity (equation 7) (Ibezim-Ezeani *et al.*, 2017b), free energy of activation of flow (equation 8) (Jha *et al.*, 2010), change in free energy (equation 9) (Engel and Reid, 2006), enthalpy (equation 10) and entropy (equation 11) (Ibezim-Ezeani and Obi, 2017) of mixing in the various DME-EC solvent proportions at 25, 40, 50, 60 and 70 °C were deduced (Tables 1 – 5) with the following equations:

$$\rho = \frac{m}{V} \qquad \dots\dots (1)$$

Where ρ = density (kgm^{-3}), m = mass (kg) and V = Volume (m^3)

$$\eta = v\rho = Kt\rho \qquad \dots\dots (2)$$

where v is kinematic viscosity (m^2s^{-1}), η is dynamic viscosity ($kgm^{-1}s^{-1}$), K is viscometer constant (m^2s^{-2}) and t is flow time (s).

$$V_m = \frac{(X_1M_1 + X_2M_2)}{\rho_m} \qquad \dots\dots (3)$$

$$V^E = X_1M_1\left(\frac{1}{\rho} - \frac{1}{\rho_1}\right) + X_2M_2\left(\frac{1}{\rho} - \frac{1}{\rho_2}\right) \qquad \dots\dots (4)$$

where V_m = Molar volume (m^3mol^{-1}), V^E = Molar excess volume (m^3mol^{-1}), X_1 = Mole fraction of component 1, M_1 = Molar Mass of component 1 ($kgmol^{-1}$), ρ_1 = Density of component 1 (kgm^{-3}), ρ = Densities of the mixtures (kgm^{-3}), X_2 = Mole fraction of component 2, M_2 = Molar mass of component 2 ($kgmol^{-1}$), ρ_2 = Density of component 2 (kgm^{-3}).

$$d = \ln\eta - \frac{(X_1\ln\eta_1 + X_2\ln\eta_2)}{X_1X_2} \qquad \dots\dots (5)$$

where η = viscosity of mixture ($kgm^{-1}s^{-1}$), η_1 = viscosity of pure component 1 ($kgm^{-1}s^{-1}$), η_2 = viscosity of pure component 2 ($kgm^{-1}s^{-1}$), X_1 = mole fraction of component 1, X_2 = mole fraction of component 2.

$$\Delta G^E = RT\left[\ln\eta V_m - \left(X_1\ln\eta_1 V_1 + X_2\ln\eta_2 V_2\right)\right] \qquad \dots\dots (6)$$

$$\eta^E = \eta - (X_1\eta_1 + X_2\eta_2) \qquad \dots\dots (7)$$

where R is the universal constant of gases ($JK^{-1}mol^{-1}$), T is the temperature (K), ΔG^E is the excess Gibbs free energy ($kJmol^{-1}$), h is Planck's constant (Js), N is Avogadro number (mol^{-1}), V_1 and V_2 are the molar volumes (m^3) of component 1 and 2 respectively, η^E = Excess viscosity ($kgm^{-1}s^{-1}$), η = Viscosity of mixed electrolyte ($kgm^{-1}s^{-1}$), η_1 = Viscosity of pure component 1 ($kgm^{-1}s^{-1}$), η_2 = Viscosity of pure component 2 ($kgm^{-1}s^{-1}$), X_1 = Mole fraction of component 1 and X_2 = Mole fraction of component 2.

$$G = RT\ln\left(\frac{\eta V_m}{hN}\right) \qquad \dots\dots (8)$$

$$\Delta G^M - nRT(X_1\ln X_1 + X_2\ln X_2) \qquad \dots\dots (9)$$

$$\Delta S^M = -nR(X_1\ln X_1 + X_2\ln X_2) \qquad \dots\dots (10)$$

$$\Delta H^M = \Delta G^M + T\Delta S^M \qquad \dots\dots (11)$$

where the change in free energy is ΔG^M ($kJmol^{-1}$), entropy is ΔS^M ($JK^{-1}mol^{-1}$) and enthalpy ΔH^M ($kJmol^{-1}$) of mixing.

RESULTS AND DISCUSSION

Density

Density measurement reflects the ratio of the total energy content of a substance to that of unit space occupied by it. The measured values of density for the various mole fraction of DME at temperatures of 25, 40, 50, 60 and 70 °C is presented in Tables 1-5. There was a general increase in density with decrease in the mole fraction of DME and increase in that of the EC. Considering that the density of a substance is its mass per unit volume; then, the addition of pure EC with larger density (1.321 g/cm^3) to replace the same quantity of DME will lead to mixed system of an average density higher than that of pure DME. This explanation is in line with the sequential increase in the mole fraction of EC which corresponds to increase in the average density of the mixture at the temperatures studied. Again, the increase in the mole ratio of EC could have lead to the formation of bulkier molecule with longer chain structure as compared to the pure DME. This blending could also result to the increase in forces of attraction between solvent-solvent and ion-solvent to yield species with higher molecular weight and mass, as well as increased density of the system. Furthermore, for the body of mass, m with acceleration, a; the force acting on the system is F = ma; and if density, ρ = m/V, then ρ = F/Va. Thus, density is directly proportional to the attractive force by the equation: ρ = F/Va. This possibly infer that increase in attractive forces due to dipole and dipole-induced dipole interactions between dissimilar molecules could result in formation of species with larger mass in the system which will certainly increase the density of the medium.

Viscosity

Viscosity parameter quantifies the degree of opposition to the flow of fluid. The nature and magnitude of forces acting between molecules in the fluid, influences its viscosity at any given condition. The values in Tables 1 – 5 reveal an increase in viscosity with decrease in mole fraction of DME and increase in mole fraction EC.

Table 1. Parameters of mixing DME and EC at 25 ^oC

$X_{1\ (DME)}$	1	0.7197	0.5982	0.4905	0.3910	0.2997	0.2157
$X_{2\ (EC)}$	0	0.2802	0.4018	0.5094	0.6089	0.7002	0.7842
(kg/m^3)	868	945	995	1035	1085	1139	1185
η (kgm^{-1}s^{-1})	3.56E-04	4.44E-04	5.16E-04	6.82E-04	7.49E-04	8.58E-04	9.66E-04
V^M (m^3 mol^{-1})	1.038E-04	9.475E-05	8.974E-05	8.605E-05	8.190E-05	7.785E-05	7.468E-05
V^E (m^3 mol^{-1})	0	1.33E-06	8.28E-07	1.14E-06	6.79E-07	1.96E-08	-3.09E-08
d	-	-1.228	-0.899	-0.805	-1.149	-1.382	-1.843
ΔG^E	-37.286	-36.966	-36.520	-36.141	-36.031	-35.820	-35.629
η^E (kgm^{-1}s^{-1})	0.00E+00	-3.42E-04	-4.11E-04	-4.55E-04	-541E-04	-5.72E-04	-5.93E-04
G (kJmol^{-1})	11.226	11.546	11.992	12.372	12.481	12.692	12.883
ΔG^M (kJmol^{-1})	-	-2.940	-3.338	-3.434	-3.316	-3.026	-2.584
ΔS^M (JK^{-1}mol^{-1})	-	9.858	11.197	11.517	11.122	10.149	8.667
ΔH^M (kJmol^{-1})	0	0	0	0	0	0	0

Table 2. Parameters of mixing DME and EC at 40 ^oC

$X_{1\ (DME)}$	1	0.7197	0.5982	0.4905	0.3910	0.2997	0.2157
$X_{2\ (EC)}$	0	0.2802	0.4018	0.5094	0.6089	0.7002	0.7842
(kg/m^3)	845	932	988	1034	1075	1105	1124
η (kgm^{-1}s^{-1})	2.92E-04	4.09E-04	5.27E-04	6.48E-04	7.68E-04	7.97E-04	8.81E-04
V^M (m^3 mol^{-1})	1.067E-04	9.607E-05	9.038E-05	8.613E-05	8.266E-05	8.024E-05	7.873E-05
V^E (m^3 mol^{-1})	0	-1.14E-07	-1.28E-06	1.50E-06	-1.26E-06	-2.71E-07	1.35E-06
d	-	-0.72118829	-0.42041888	-0.31561781	-0.68118625	-0.95670249	-1.44709466
ΔG^E	-39.267	-38.657	-38.158	-37.742	-37.621	-37.389	-37.177
η^E (kgm^{-1}s^{-1})	-5.00E-07	-2.58E-04	-3.03E-04	-3.25E-04	399E-04	-4.32E-04	-4.60E-04
G (kJmol^{-1})	10.797	11.378	11.855	12.299	12.365	12.585	12.787
ΔG^M (kJmol^{-1})	-	-3.052	-3.466	-3.566	-3.443	-3.142	-2.683
ΔS^M (JK^{-1}mol^{-1})	-	9.746	11.069	11.386	10.996	10.033	8.568
ΔH^M (kJmol^{-1})	0	0	0	0	0	0	0

Table 3. Parameters of mixing DME and EC at 50 ^oC

$X_{1\ (DME)}$	1	0.7197	0.5982	0.4905	0.3910	0.2997	0.2157
$X_{2\ (EC)}$	0	0.2802	0.4018	0.5094	0.6089	0.7002	0.7842
(kg/m^3)	821	921	986	1025	1063	1092	1102
η (kgm^{-1}s^{-1})	2.83E-04	4.64E-04	4.95E-04	5.78E-04	7.01E-04	7.56E-04	8.30E-04
V^M (m^3 mol^{-1})	1.098E-04	9.721E-05	9.056E-05	8.689E-05	8.359E-05	8.120E-05	8.034E-05
V^E (m^3 mol^{-1})	0	-2.01E-06	-4.11E-06	3.72E-06	-3.28E-06	-2.24E-06	2.36E-08
d	-	-0.564	-0.473	-0.559	-0.476	-0.909	-1.407
ΔG^E	-40.422	-39.793	-39.437	-39.133	-38.718	-38.593	-38.372
η^E (kgm^{-1}s^{-1})	0.00E+00	-2.23E-04	-2.81E-04	-3.30E-04	-3.29E-04	-3.86E-04	-4.15E-04
G (kJmol^{-1})	11.700	12.330	12.685	12.991	13.405	13.530	13.751
ΔG^M (kJmol^{-1})	-	-3.148	-3.575	-3.678	-3.551	-3.241	-2.767
ΔS^M (JK^{-1}mol^{-1})	-	9.742	11.064	11.380	10.990	10.029	8.564
ΔH^M (kJmol^{-1})	0	0	0	0	0	0	0

Table 4. Parameters of mixing DME and EC at 60 ^oC

$X_{1\ (DME)}$	1	0.7197	0.5982	0.4905	0.3910	0.2997	0.2157
$X_{2\ (EC)}$	0	0.2802	0.4018	0.5094	0.6089	0.7002	0.7842
(kg/m^3)	814	912	964	1015	1051	1052	1012
η (kgm^{-1}s^{-1})	2.30E-04	3.43E-0.4	4.54E-04	5.73E-04	6.26E-04	6.93E-04	7.30E-04
V^M (m^3 mol^{-1})	1.107E-04	9.817E-05	9.263E-05	8.795E-05	8.454E-05	8.429E-05	8.746E-05
V^E (m^3 mol^{-1})	0	-2.42E-06	-3.58E-06	-4.57E-06	-4.18E-06	-1.15E-06	5.04E-06
d	-	-0.583	-0.262	-0.115	-0.521	-0.914	-1.743
ΔG^E	-42.289	-41.511	-40.899	-40.402	-40.259	-39.988	-39.741
η^E (kgm^{-1}s^{-1})	0.00E+00	-2.31E-04	-2.71E-04	-2.83E-04	-3.53E-04	-3.98E-04	-4.64E-04
G (kJmol^{-1})	11.508	12.287	12.898	13.396	13.539	13.810	14.057
ΔG^M (kJmol^{-1})	-	-3.282	-3.728	-3.834	-3.703	-3.379	-2.885
ΔS^M (JK^{-1}mol^{-1})	-	9.851	11.189	11.509	11.114	10.141	8.660
ΔH^M (kJmol^{-1})	0	0	0	0	0	0	0

This signifies increased resistance to fluid flow, which is occasioned by the prevalence of interacting forces between unlike molecules. The free energy of activation of flow (G) is the minimum amount of energy which molecules must acquire in order to flow. The values of G in Tables 1 – 5 increased with decrease in mole fraction of DME and increase in that of EC. This indicates that as the viscosity of the medium increased from 0.7197-0.2157, increased free energy is required to activate the flow of fluid.

Excess parameters

The characteristic pattern of interaction between mixed solvent molecules is possibly understood by investigation into the excess parameters such as in solvent-solvent interaction. Excess parameters are therefore influenced by forces involved in the mixing. The available excess volume is a measure of the unoccupied space by the interacting molecules within the volume of mixture as well as relative closeness of the molecules of the constituents.

Table 5. Parameters of mixing DME and EC at 70 °C

$X_{1\ (DME)}$	1	0.7197	0.5982	0.4905	0.3910	0.2997	0.2157
$X_{2\ (EC)}$	0	0.2802	0.4018	0.5094	0.6089	0.7002	0.7842
(kg/m^3)	810	910	961	1010	1022	1029	1001
η (kgm^{-1}s^{-1})	2.93E-04	3.14E-04	4.22-04	5.38E-04	5.79E-04	6.42E-04	6.91E-04
V^M (m^3 mol^{-1})	1.113E-04	9.839E-05	9.292E-05	8.818E-05	8.659E-05	8.841E-05	8.841E-05
V^E (m^3 mol^{-1})	0	-5.18E-06	-7.34E-06	-9.12E-06	-7.63E-06	-5.48E-06	-1.36E-06
d	-	-1.786	-1.039	-0.684	-1.066	-1.362	-2.019
ΔG^E	-42.801	-42.957	-42.217	-41.731	-41.575	-41.280	-41.014
η^E (kgm^{-1}s^{-1})	0.00E+00	-2.75E-04	-2.96E-04	-2.93E-04	-3.60E-04	-3.91E-04	-4.31E-04
G (kJmol^{-1})	12.564	12.408	13.088	13.634	13.991	14.086	14.352
ΔG^M (kJmol^{-1})	-	-3.290	-3.737	-3.844	-3.712	-3.387	-2.893
ΔS^M (JK^{-1}mol^{-1})	-	9.589	10.891	11.202	10.818	9.871	8.430
ΔH^M (kJmol^{-1})	0	0	0	0	0	0	0

The forces at play in solvent- solvent interaction therefore influence the closeness of the mixing parameters to ideality. In the case of ideal mixture, the values of excess parameter are zero, while the non-zero values indicate non-ideal mixing (Dikio *et al.*, 2012). The positive values of excess volume in some cases between 25 and 50 °C suggest strong interactions; while negative values (Tables 1 – 5) of excess volume, excess Gibbs free energy of activation of flow and excess viscosity at temperatures of 25-70 °C, indicate the dominance of forces of dispersion and weak interaction between dissimilar solvent molecules (Ibezim-Ezeani *et al.*, 2017b).

d-parameter

Grunberg and Nissan derived a partly empirical expression relating the viscosity of the binary component mixture to its mole fraction. The d-parameter lays emphasis on the degree of interaction between the molecular constituents of the binary mixture at a given temperature. The measured values of the dynamic viscosity were applied in the deduction of d-parameter. The results in Tables 1 – 5 showed decrease in the d-parameter with decrease in mole fraction of DME in most cases, signifying stronger interaction of dissimilar molecules at increased mole component of DME (from 0.7197 – 0.4905) and thereafter, decreased with DME mole fractions from 0.4905 – 0.2157. On the overall assessment, the negative value of d-parameter in some cases is suggestive of dipole-dipole and dipole-induced dipole interaction of little magnitude between species of dissimilar forms.

Thermodynamics of mixing

The change in Gibbs free energy of mixing (ΔG^M) reveals the tendency for the solvents to mix. The negative values (Tables 1 – 5) of ΔG^M indicate that the free energy of the pure solvents (reactant) exceeds that of the solvent mixture (product), which led to the increase in the disorderliness of the system and its surrounding. Hence, the more reduced the ΔG^M value, the more the possibility of solvents interaction to form mixture. The ΔG^M values increased in negativity with decrease in mole fractions of DME. Again, the negative sign on ΔG^M values signify the existence of sufficient free energy within the system, which is released in the course of mixing. The varied values of ΔG^M with mole ratios of DME and EC within the experimental temperature, confirms the dependence of ΔG^M on temperature and the amount of the solvent constitutions that form the mixture. Thus, if the free energy change associated with the formation of mixture at a given temperature and pressure is more than that of the component solvents, then the process will be non-spontaneous; on the contrary, the process becomes spontaneous if the free energy value of the product is less than that of the component solvents. Considering that $\Delta G^M = \Delta H^M - T\Delta S^M$; then, $\Delta G^M = \Delta H^M - T\Delta S^M < 0$ is the driving expression for the spontaneous reaction condition. Result analyses revealed that the spontaneity of the process increased with entropy and in line with the value of $\Delta S^M > O$.

Furthermore, the favorability of a reaction depends on the total bond energy (ΔH^M) possessed by the reactants and products, as well as the changing pattern of disorderliness (ΔS^M) of the system. The values of ΔH^M is zero in the whole experiment, indicating the negligible effect of intra- and inter-bonding forces at all temperatures; and dominance of solvent-solvent interactions. The positive values of ΔS^M suggest increased level of disorderliness, which led to a less structured system. It also reveals that the solvent molecules did not cage each other, but are dispersed in the binary system.

Conclusion

Thermodynamic parameters of binary electrolyte system containing mixture of 1,2-dimethoxyethane (DME) and ethylene carbonate (EC) at various temperatures (25, 40, 50, 60 and 70 °C) and mole fractions (1.000, 0.7197, 0.5982, 0.4905, 0.3910, 0.2997 and 0.2157 of DME) has been investigated. Density, viscosity, excess volume, free energy of activation of flow, excess viscosity, d-parameter, excess Gibbs free energy, change in free energy, enthalpy and entropy of mixing are the parameters considered. The values of density, viscosity, excess viscosity and free energy of activation of flow increased with increase in mole fraction of EC. The varied values of density, viscosity, excess volume, free energy of activation of flow, excess viscosity, d-parameter, excess Gibbs free energy, change in free energy and entropy of mixing evaluated for different mole ratios of DME and temperatures revealed the dependence of the reaction system on temperature and concentration of species. The negative values of change in free energy of mixing indicate the feasibility of the reaction system; while the positive values of entropy of mixing indicate the increased randomness of the reaction in the mixed system. The enthalpy of mixing is zero, indicating that dipole-dipole interactions were predominant in the system.

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