JOURNAL OF CHEMICAL & ENGINEERING DATA

ARTICLE

pubs.acs.org/jced

Salt Diffusion Coefficients, Concentration Dependence of Cell Potentials, and Transference Numbers of Lithium Difluoromono(oxalato)borate-Based Solutions

Sandra Zugmann,[†] Matthias Fleischmann,[‡] Marius Amereller,[§] Ruth M. Gschwind,[‡] Martin Winter,[§] and Heiner J. Gores*[,†,§]

[†]Institute of Physical and Theoretical Chemistry, University of Regensburg, 93040 Regensburg, Germany
[‡]Institute of Organic Chemistry, University of Regensburg, 93040 Regensburg, Germany
[§]Institute of Physical Chemistry, Westfälische Wilhelms-Universität, 48149 Münster, Germany

ABSTRACT: Several properties of the electrolyte solution lithium difluoromono(oxalato)borate (LiDFOB) in ethylene carbonate diethylcarbonate (EC/DEC (3:7 mass ratio)) are given, including transference numbers, diffusion coefficients, concentration dependence of cell potentials, and the Haven ratio. The concentration range of the salt covers 0.1 mol·kg^{-1} to 1.0 mol·kg^{-1}. In comparison to the standard salt LiPF$_6$ currently in use in lithium ion cells LiDFOB has higher transference numbers. However, the Haven ratio related to ion pair formation in an electrolyte indicates a high amount of aggregates in concentrated LiDFOB/EC/DEC (3:7) electrolytes.

INTRODUCTION

Electrolytes with the salt lithium difluoromono(oxalato)borate (LiDFOB) offer promising results for its use in lithium ion batteries.[1−3] The currently used salt lithium hexafluorophosphate (LiPF$_6$) has some drawbacks, such as HF formation with traces of water[4] and decomposition upon thermal stress at rather low temperatures,[5−7] triggering the search for possible substitutes. LiDFOB has a higher thermal stability,[1] shows a stable solid electrolyte interface (SEI) formation,[8] possesses an adequate conductivity,[2] has a sufficient stability window, and offers excellent Al-corrosion prevention.[9]

The characterization of a battery electrolyte requires the knowledge of conductivity, diffusion coefficients, and in particular, understanding of charge transport that is represented by the transference number.[10−14] Recently we published a comparative study for the determination of transference numbers with three electrochemical methods (potentiostatic polarization method, PP, galvanostatic polarization method, GP, and electromotive force, emf, method) and NMR spectroscopy.[3] Transference numbers of concentrated nonaqueous solutions can accurately be determined by the galvanostatic polarization method.[3,15] By combining measurements of three different parameters, that is, the measurement of the cell potential after galvanostatic polarization, the detection of the salt diffusion coefficient, and the determination of the concentration dependence of the cell potential difference, the cationic transference number is given by:[16]

$$t_+ = 1 - \frac{m_\Phi F c_\infty \sqrt{\pi D}}{4(\mathrm{d}\Phi/\mathrm{d}\ln c)} \tag{1}$$

where m_Φ is the slope of a plot of the cell potential at the time of current interruption obtained from $\Delta\Phi$ vs $jt_i^{1/2}$ fits, where j is the current density and t_i the polarization time, c_∞ the bulk concentration of the salt, F the Faraday constant, D the salt diffusion coefficient, and $\mathrm{d}\Phi/\mathrm{d}\ln c$ the concentration dependence of the potential Φ.

The salt diffusion coefficient is evaluated by the restricted diffusion technique.[17,18] The diffusion coefficient is determined from a plot of $\ln(\Delta\Phi)$ versus time[19] where the slope m_D is linear for long times $t \gg L^2/D$. The slope is given by

$$m_D = -\frac{\pi^2 D}{L^2} \tag{2}$$

where L is the distance between the two electrodes.

Self-diffusion coefficients can easily be determined by the pulsed field gradient nuclear magnetic resonance (pfg-NMR) method[20−24] by means of the SE or STE (spin echo or stimulated echo) or with the more sophisticated BPLED pulse sequence (longitudinal eddy current delay with bipolar gradient pulses stimulated echo). The corresponding diffusion coefficients are distinguished from salt diffusion coefficients determined by the electrochemical method, because NMR averages the diffusion coefficients of all of the nuclei in the solution, even associates such as uncharged ion pairs and quadruplets. From this deviation, it is possible to characterize the ionicity and thereby the proportion of ion pair formation in the electrolyte at high concentrations, by the Haven ratio H_r:[25]

$$H_r = \frac{\Lambda^0_m}{\Lambda^0_{NMR}} \tag{3}$$

where $\Lambda^0{}_m$ is the molar conductivity determined from the specific conductivity κ and $\Lambda^0{}_{NMR}$ the molar conductivity

Special Issue: Kenneth N. Marsh Festschrift

Received: July 23, 2011
Accepted: October 11, 2011
Published: October 26, 2011

dx.doi.org/10.1021/je2007814 | J. Chem. Eng. Data 2011, 56, 4786–4789

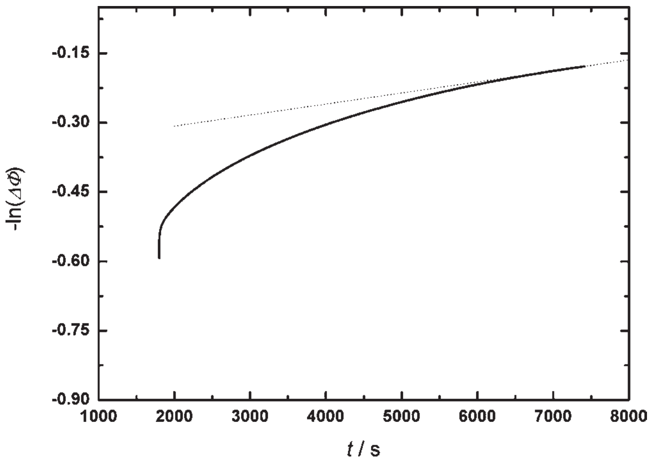

Figure 1. Plot of $-\ln(\Delta\Phi)$ vs relaxation time t for 0.68 $\text{mol}\cdot\text{kg}^{-1}$ LiDFOB in EC/DEC (3:7).

Table 1. Slopes of a Plot $-\ln(\Delta\Phi)$ vs Relaxation Time t and Salt Diffusion Coefficients for LiDFOB in EC/DEC (3:7)[a]

m	slope	D
$\text{mol}\cdot\text{kg}^{-1}$	s^{-1}	$\text{m}^2\cdot\text{s}^{-1}$
0.1	$1.64\cdot10^{-5}\pm1.1\cdot10^{-6}$	$3.95\cdot10^{-10}\pm3.5\cdot10^{-12}$
0.68	$2.20\cdot10^{-5}\pm8.4\cdot10^{-7}$	$2.27\cdot10^{-10}\pm5.2\cdot10^{-12}$
1.0	$4.26\cdot10^{-5}\pm1.9\cdot10^{-6}$	$1.55\cdot10^{-10}\pm5.7\cdot10^{-13}$

[a] m is the molality of the salt; its standard uncertainty $u(m)$ is 0.001.

calculated from self-diffusion coefficients, given by the Nernst–Einstein equation[26]

$$\Lambda^0_{\text{NMR}} = \frac{|z|eF}{k_B T}(D_+ + D_-) \tag{4}$$

where z is the charge number, e the elementary charge, F the Faraday constant, k_B the Boltzmann constant, T the temperature, and D_+ and D_- the self-diffusion coefficients of the cation and anion, respectively.

In the present study, the lithium ion transference number of nonaqueous electrolytes containing the lithium salts lithium difluoromono(oxalato)borate (LiDFOB) and lithium hexafluorophosphate (LiPF_6) was determined by the galvanostatic polarization (GP) method. In addition, ion pair formation of LiDFOB based solutions was characterized by the Haven ratio.

EXPERIMENTAL SECTION

Materials. LiDFOB was prepared by the synthesis novel route introduced by Schreiner et al.[27] with a purity of 0.997 mass fraction; LiPF_6 was purchased from Stella (high purity, >0.999 mass fraction) and tetraethylammonium tetrafluoroborate (TEABF_4) from Merck KGaA (selectipure, >0.999 mass fraction). The solvents ethylene carbonate (EC) and diethyl carbonate (DEC) were purchased from Merck KGaA (p.a.) and were used as solvents with the mixture EC/DEC = 3:7 by weight. The water content of the electrolytes was $<5\cdot10^{-5}$, checked by Karl Fischer titration (Mettler, type Karl Fischer Titrator DL18). All solutions were prepared in a glovebox (Mecaplex GB80) with a low mass fraction of water ($<1\cdot10^{-6}$) and oxygen ($<5\cdot10^{-6}$).

Table 2. Diffusion Coefficient, Conductivity, and Cationic Transference Number of 1.0 $\text{mol}\cdot\text{kg}^{-1}$ LiDFOB and LiPF_6 in EC/DEC (3:7), Respectively

	D	κ	
salt	$\text{m}^2\cdot\text{s}^{-1}$	$\text{mS}\cdot\text{cm}^{-1}$	t_+
LiDFOB	$1.55\cdot10^{-10}\pm5.7\cdot10^{-13}$	4.1722 ± 0.0098	0.33 ± 0.050
LiPF_6	$3.47\cdot10^{-10}\pm5.7\cdot10^{-12}$	7.4179 ± 0.0082	0.24 ± 0.087

The cell type and used electrodes for galvanostatic polarization, emf measurements, and restricted diffusion technique are described in ref 3.

Diffusion Coefficient Measurements. The restricted diffusion technique was carried out by polarization measurements using 0.05 mA for 1800 s and detecting the relaxation potential over 100 min following current interruption. As electrode material, active material attached to aluminum foil was used. The foil was cut in a spherical shape and pressed on the electrode tips as described in ref 3.

NMR measurements were performed with a standard Bruker BPLED sequence in a pseudo 2D mode, recorded on a Bruker Avance III 400 Nanobay (400.13 MHz) spectrometer equipped with a $\text{BBFO}^{\text{PLUS}}$ room temperature probe with a z-gradient (maximum gradient strength of 53.5 $\text{G}\cdot\text{cm}^{-1}$), as described in ref 3.

Electromotive Force (emf) Measurements. For emf measurements two glass wool pieces are soaked with electrolytes of different concentrations and brought in contact by glass wool soaked with a nonaqueous salt bridge. A 1 m concentration (molality) was held constant (1 $\text{mol}\cdot\text{kg}^{-1}$), whereas the second glass wool was soaked in several succeeding steps with the same electrolyte but at varying concentration (from 0.9 $\text{mol}\cdot\text{kg}^{-1}$ down to 0.01 $\text{mol}\cdot\text{kg}^{-1}$). As a salt bridge a saturated tetraethylammonium tetrafluoroborate (TEABF_4) solution in EC/DEC (3:7) was used.

Galvanostatic Polarization Measurements. Measurements were carried out with current densities ranging from (0.04 to 0.18) $\text{mA}\cdot\text{cm}^{-2}$ and a polarization time of 400 s.

Conductivity Measurements. Conductivity measurements were performed as described earlier,[28] as well as the thermostat setup which allows a temperature stability of ± 2 mK (monitored by an ASL F-250 MkII thermometer (Automatic Systems Laboratories)) that was described elsewhere.[29,30] The estimated uncertainty of the measured specific conductivities is in the range of (0.2 to 0.4) %.

Density Measurements. Densities were determined with a precision densitometer DMA 60/DMA 602 from Anton Paar at 298 K. Temperature control (better than ± 0.02 K) was maintained by a RK 8 KP thermostat from LAUDA and controlled by a temperature sensor close to the measuring cell. The estimated uncertainty of the measured densities is below 0.01 %.

RESULTS AND DISCUSSION

Diffusion coefficients were determined by the presented electrochemical method as well as by NMR measurements. An example for a measurement by the restricted diffusion method for 0.68 $\text{mol}\cdot\text{kg}^{-1}$ LiDFOB in EC/DEC (3:7) is shown in Figure 1. With the slope m_D of the plot of $-\ln(\Delta\Phi)$ vs relaxation time t the diffusion coefficient can be calculated by eq 2.

The obtained slopes and calculated diffusion coefficients for three different concentrations are summarized in Table 1.

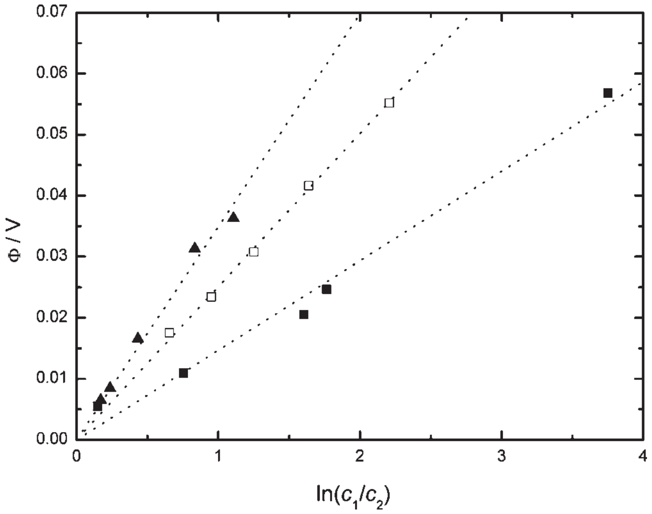

Figure 2. Concentration dependence of the potential Φ for solutions of LiDFOB in EC/DEC (3:7): ■, 0.1 mol·kg^{-1}; □, 0.68 mol·kg^{-1}; ▲, 1.0 mol·kg^{-1}.

Table 3. Concentration Dependence of the Potential dΦ/d ln c and Density of LiDFOB in EC/DEC (3:7)[a]

m	ρ	dΦ/d ln c
mol·kg^{-1}	g·cm^{-3}	V
0.1	$1.05836 \pm 7.9 \cdot 10^{-4}$	$0.0147 \pm 5.49 \cdot 10^{-4}$
0.68	$1.10807 \pm 7.3 \cdot 10^{-4}$	$0.0251 \pm 2.25 \cdot 10^{-4}$
1.0	$1.13776 \pm 6.7 \cdot 10^{-4}$	$0.036 \pm 1.21 \cdot 10^{-3}$

[a] m is the molality of the salt; its standard uncertainty $u(m)$ is 0.001.

Table 4. Cationic Transference Numbers for LiDFOB in EC/DEC (3:7)[a]

m	
mol·kg^{-1}	t_+
0.10	0.38 ± 0.048[3]
0.68	0.34 ± 0.019
1.0	0.33 ± 0.050[3]

[a] m is the molality of the salt; its standard uncertainty $u(m)$ is 0.001.

As expected, the diffusion coefficient decreases with increasing concentration starting with $3.95 \cdot 10^{-10} \pm 3.5 \cdot 10^{-12}$ m^2·s^{-1} for a 0.1 mol·kg^{-1} solution of LiDFOB in EC/DEC (3:7), $2.27 \cdot 10^{-10} \pm 5.2 \cdot 10^{-12}$ m^2·s^{-1} for 0.68 mol·kg^{-1}, and $1.55 \cdot 10^{-10} \pm 5.7 \cdot 10^{-13}$ m^2·s^{-1} for 1.0 mol·kg^{-1}. Compared to the standard salt LiPF$_6$ in EC/DEC (3:7) at the same concentration the salt diffusion coefficient of LiDFOB is smaller (as well as the conductivity of the salt), but the cationic transference number is much higher. The values for diffusion coefficient, conductivity, and transference number of the 1.0 mol·kg^{-1} solutions of LiDFOB and LiPF$_6$ in EC/DEC (3:7) are given in Table 2.

It seems that the major charge transport of LiPF$_6$ in EC/DEC (3:7) is carried out by the anions. Therefore, LiDFOB appears to be an attractive salt for application in lithium ion batteries because of its high cation transport.

The concentration dependence of the potential of LiDFOB in EC/DEC (3:7) determined with transference cells without transference for different concentrations is shown in Figure 2 as well as in Table 3, where the density and the obtained slopes of Figure 2 are given.

The calculation of the transference number by the galvanostatic polarization method is based on molar concentration; nevertheless in Table 3 dΦ/d ln c is given for molal concentrations for comparison. The determination of molar concentration was done by density measurements.

Low concentrations of LiDFOB in EC/DEC (3:7) have stronger concentration dependence than concentrated solutions. The potential in concentrated solutions changes faster with little changes in concentration, which is an important aspect for battery application, especially in the case of concentration polarization. Therefore, high transference numbers are preferred to avoid potential loss. Cationic transference numbers are shown in Table 4. As can be seen, the transference numbers decrease with increasing concentration as expected for transference numbers with a value < 0.5 at infinite concentration.[3,31]

A comparison of transference numbers measured by electrochemical methods or by NMR method[3] shows that ion pair diffusion plays an important role. The Haven ratio can give information about this transport phenomenon. If H_r is larger than 1, ions with opposite charges are moving in the same direction, meaning ion pair diffusion occurs. Data for LiDFOB in EC/DEC (3:7) are given in Table 5.

Even low concentrations show a value higher than 1 for H_r, which implicates a significant ion pair concentration that increases with increasing salt concentration, leading to a Haven ratio over 3 at high concentrations.

Table 5. Molar Conductivity Λ and Λ_{NMR}, Calculated from Specific Conductivity κ and from Self-Diffusion Coefficients, Respectively, and the Haven Ratio H_r for LiDFOB in EC/DEC (3:7)[a]

m	Λ	Λ_{NMR}	
mol·kg^{-1}	mS·cm^2·mol^{-1}	mS·cm^2·mol^{-1}	H_r
0.93	4.3700 ± 0.0098	$1.41297 \pm 2.10 \cdot 10^{-5}$	$3.102 \pm 7.10 \cdot 10^{-3}$
0.83	4.5254 ± 0.0118	$1.54626 \pm 1.28 \cdot 10^{-5}$	$2.916 \pm 7.73 \cdot 10^{-3}$
0.70	4.7098 ± 0.0153	$1.82719 \pm 1.53 \cdot 10^{-5}$	$2.571 \pm 8.47 \cdot 10^{-3}$
0.50	4.8874 ± 0.0246	$2.08887 \pm 1.76 \cdot 10^{-5}$	2.344 ± 0.012
0.30	4.7852 ± 0.0468	$2.50763 \pm 2.14 \cdot 10^{-5}$	1.913 ± 0.019
0.10	3.9372 ± 0.1505	$3.13964 \pm 2.65 \cdot 10^{-5}$	1.258 ± 0.049
0.051	3.2903 ± 0.3139	$3.28293 \pm 2.80 \cdot 10^{-5}$	1.006 ± 0.096

[a] m is the molality of the salt; its standard uncertainty $u(m)$ is 0.001.

■ CONCLUSIONS

Transference numbers, diffusion coefficients, concentration dependence of the concentration cell potential, and the Haven ratio for LiDFOB in EC/DEC (3:7) were presented from diluted to concentrated solutions. LiDFOB has lower diffusion coefficients than the standard salt $LiPF_6$ but higher transference numbers and seems to be an interesting salt for lithium ion batteries. The Haven ratio indicates strong ion pair formation, especially in concentrated electrolytes.

■ AUTHOR INFORMATION

Corresponding Author

*Tel.: +49(0)941/9434746, fax: +49(0)941/9434532. E-mail address: heiner.gores@chemie.uni-regensburg.de, hgore_01@uni-muenster.de.

Funding Sources

For financial support the Deutsche Forschungsgemeinschaft (DFG) in association with Project Initiative PAK 177: Funktionsmaterialien und Materialanalytik zu Lithium-Hochleistungsbatterien (# 544243) is gratefully acknowledged.

■ DEDICATION

Thanks to Ken Marsh for helpful discussions and motivating advice in the past. Thanks for improving our manuscripts submitted to the *Journal of Chemical & Engineering Data*. Best wishes for his future.

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