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Application of Zwanzig theory for sodium diethyldithiocarbamate and alkali metal ions in aqueous and non-aqueous solvents at 25°C

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ABSTRACT

The Zwanzig theory of dielectric friction was applied to sodium diethyldithiocarbamate and alkali metal ions (Li^+, K^+, Rb^+ and Cs^+) in different pure solvents at 25° C. It was found that the value of (r_i) obtained from the slope increases with increasing of the size from Na^+ ion to diethyldithiocarbamate ion (DDC^-) However, the value of (r_i) obtained from the intercept decreases with increasing the size from Na^+ ion to (DDC^-) ion. The same trend is exist in case of alkali metal ions (Li^+, K^+, Rb^+ and Cs^+) where the value of (r_i) obtained from the intercept decreases with increasing the size from Li^+ to Cs^+ ions but, The value of (r_i) obtained from the slope increases with increasing of the size from Na^+ to Cs^+ ions. Such differences were investigated and discussed on the basis of the assumptions of Zwanzig theory. There was a scattering of the resulting points around the straight lines for the given ions in different solvents. The scattered values may be attributed to certain specific factors such as the uncertainty in the values of the relaxation time (τ) and solvation which are ignored in the continuum model and these play an important role in determining ionic mobility.

Keywords: Zwanzig theory, hydrated radius, hydrodynamic radius, Conductance, sodium diethyldithiocarbamate and alkali metal ions (Li^+, K^+, Rb^+ and Cs^+).

INTRODUCTION

Ionic conductance of sodium diethyldithiocarbamate in water, methanol, ethanol, n-propanol, n- butanol and acetonitrile has been measured at 25°C and ionic conductance of alkali metal ions (Li^+, K^+, Rb^+ and Cs^+) in water, methanol, ethanol, n-propanol and acetonitrile has been reported at 25°C [1-9].

Fuoss [10] has given a relationship between the ionic conductivity of an ion in different solvents with solvent properties. Boyd [11] and Zwanzig [12] have improved it and expressed λ_i^o as

$$\lambda_i^o = F^2 / N [6\pi\eta_o r_i + (2e^2 \tau / 3r_i^3) / [(\epsilon_o - \epsilon_\infty) / \epsilon_o^2)] \quad (1)$$

It was considered interesting to examine the applicability of Fuoss-Boyd-Zwanzig (F.B.Z) theory to present data. Equation (1) can be used most conveniently in the linear form suggested by Atkinson and Mori [1], where results in a number of solvents are to be compared. When equation (1) is inverted and rearranged with insertion of the numerical constant and radii in A^0, we obtain

$$(F^2/ N\eta_o\lambda^\circ_i) = 6\pi r_i + (2e^2/ 3r^3_i) [(\tau/\eta_o)(\epsilon_o-\epsilon_\infty)/\epsilon_o^2]$$

Or

$$(15.5/\eta_\circ \lambda^\circ_i) = 18.8 r_i + (15.3x10^{12} / r^3_i) [(\tau/\eta_\circ)(\epsilon_\circ-\epsilon_\infty)/\epsilon_\circ^2] \quad (2)$$

For convenience in plotting, this can be written as:

$$L^* = 18.8 r_i + (15.3x10^{12} / r^3_i) R^* \quad (3)$$

Where $L^* = 15.5/\eta_\circ \lambda^\circ_i$ and $R^* = (\tau/\eta_\circ)(\epsilon_\circ-\epsilon_\infty)/\epsilon_\circ^2$

So that a plot of L* vs. R* should be linear and r_i can be obtained from both intercept and slope.

The aim of the present work, is applying the Zwanzig theory to investigate the values of the slope and the intercept for different ions in pure solvents at 25°C. The obtained new data were interpreted.

All chemicals were highly purified reagent grade and used without further purification. Purification of conductivity water, alcohols and acetonitrile was reported in literatures[13-22].The dielectric constant (ϵ_o) for pure solvents at 25°C , the values of Optical dielectric constant (ϵ_∞), relaxation time (τ) and Bulk viscosity (η_o) for pure solvents at 25°C were taken from previous investigations [1, 23-24].

RESULTS AND DISCUSSION

Table-1 summarizes the ionic conductances of sodium and diethyldithiocarbamate in some pure solvents (water, methanol, ethanol, n-propanol, n- butanol and acetonitrile) and **Table-2** shows the ionic conductances of alkali metal ions in water, methanol, ethanol, n-propanol, and acetonitrile; while **Table-3** shows the solvent parameters needed for the F.B.Z calculations. **Tables 4-9** have depicted the parameters obtained from F.B.Z calculations. **Figures (1-6)** represent a plot L* vs. R*(10^{11}), where the least square method was used to calculate the values.

Table 1: Ionic equivalent Conductance of Na^+ and DDC^- in pure solvents at 25°C

Ionic equivalent Conductance	Water	Acetonitrile	Methanol	Ethanol	n-Propanol	n-Butanol
$\lambda_\circ$ Na^+ (Sodium Ion)	50.10	86.40	45.17	20.30	10.51	6.99
$\lambda_\circ$ DDC^- (Diethyldithiocarbamate)	58.31	93.59	53.98	25.33	17.55	11.47

Table 2: Ionic equivalent Conductance of Li^+, K^+, Rb^+ and Cs^+ in pure solvents at 25°C [1-9]

Ionic equivalent Conductance	Water	Acetonitrile	Methanol	Ethanol	n-Propanol
$\lambda_\circ$ Li^+	38.70	77.25	39.55	17.07	9.62
$\lambda_\circ$ K^+	73.50	102.92	52.44	23.55	12.92
$\lambda_\circ$ Rb^+	77.80	85.73	57.38	25.43	13.11
$\lambda_\circ$ Cs^+	77.30	101.63	60.83	26.46	14.33

Table 3: Pure solvent parameters at 25°C [1, 23-24]

Solvent	$\epsilon_\circ$	ϵ_∞	$10^{11}\tau$ (sec)	$10^2\eta_\circ$ (*poise*)
Water	78.54	5.5	0.83	0.8903
Acetonitrile	36.61	1.81	3.8	0.3409
Methanol	33.05	5.3	6.9	0.5448
Ethanol	24.77	4.5	14.4	1.0830
n-Propanol	20.33	3.24	37.12	1.9700
n-Butanol	17.51	2.95	47.73	2.6200

$\epsilon_\circ$: Static (low frequency) dielectric constant of pure solvent at 25°C
ϵ_∞: Optical (infinite frequency) dielectric constant of pure solvent at 25°C
τ: Dielectric relaxation time of pure solvent at 25°C
η_0 : Bulk viscosity of pure solvent at 25°C

The points for the given ions were found to be scattered around the straight lines. The scattered values may be attributed to the uncertainty in the values of the relaxation time (τ). **Table-10** includes values of radii (r_i) of sodium and diethyldithiocarbamate ions calculated from the intercepts and slopes of the straight line graph according to equation (3) as shown in **Figures (1-2)** while **Table-11** includes values of radii (r_i) of alkali metal ions (Li^+, K^+, Rb^+ and Cs^+) calculated from the intercepts and slopes of the straight line graph according to equation (3) as shown in **Figures (3-6)**. A quantitative test for equation (3) is to compare the values of (r_i) obtained from the slope and intercept of the straight lines of each ion. It is noticed that the two values are different; the value of (r_i) obtained from the slope increases with increasing of the size from Na^+ ion to diethyldithiocarbamate ion (DDC^-), but the value of (r_i) obtained from the intercept decreases with increasing the size from Na^+ ion to (DDC^-) ion.

Table 4: Application of Zwanzig equation for sodium ion in different solvents at 25°C

Solvent	$\lambda_\infty Na^+$	$10^2 * \eta^\circ$	$\lambda_\infty Na^+ * \eta_\circ$	$[15.5 / \lambda_\infty Na^+ * \eta_\circ] = L^*$	$10^{11}\tau$	$10^{10}\tau/\eta$	$\epsilon_\circ$	ϵ_∞	$\epsilon_0 - \epsilon_\infty$	$10^{-2}\epsilon_\circ^2$	$10^{10}R^* = 10^{10}\left[\frac{\tau}{\eta_o}(\epsilon_0 - \epsilon_\infty)/\epsilon_0^2)\right]$	$10^{11}R^*$
Water	50.10	0.8903	0.446	34.753	0.83	9.323	78.54	5.5	73.04	61.69	0.110	1.104
Acetonitrile	86.40	0.3409	0.295	52.542	3.8	111.470	36.61	1.81	34.80	13.40	2.894	28.943
Methanol	45.17	0.5448	0.246	63.008	6.9	126.652	33.05	5.3	27.75	10.92	3.218	32.176
Ethanol	20.30	1.0830	0.220	70.455	14.4	132.964	24.77	4.5	20.27	6.136	4.393	43.927
n-Propanol	10.51	1.9700	0.207	74.879	37.12	188.426	20.33	3.24	17.09	4.133	7.791	77.913
n-Butanol	6.99	2.6200	0.183	84.699	47.73	182.176	17.51	2.95	14.56	3.066	8.651	86.513

Table 5: Application of Zwanzig equation for diethyldithiocarbamate ion (DDC) in different solvents at 25°C

Solvent	$\lambda_\infty DDC^-$	$10^2 * \eta^\circ$	$\lambda_\infty DDC^- * \eta_\circ$	$[15.5 / \lambda_\infty DDC^- * \eta_\circ] = L^*$	$10^{11}\tau$	$10^{10}\tau/\eta$	$\epsilon_\circ$	ϵ_∞	$\epsilon_0 - \epsilon_\infty$	$10^{-2}\epsilon_\circ^2$	$10^{10}R^* = 10^{10}\left[\frac{\tau}{\eta_o}(\epsilon_0 - \epsilon_\infty)/\epsilon_0^2)\right]$	$10^{11}R^*$
Water	58.31	0.8903	0.519	29.865	0.83	9.323	78.54	5.5	73.04	61.69	0.110	1.104
Acetonitrile	93.59	0.3409	0.319	48.589	3.8	111.470	36.61	1.81	34.80	13.40	2.894	28.943
Methanol	53.98	0.5448	0.294	52.721	6.9	126.652	33.05	5.3	27.75	10.92	3.218	32.176
Ethanol	25.33	1.0830	0.274	56.569	14.4	132.964	24.77	4.5	20.27	6.136	4.393	43.927
n-Propanol	17.55	1.9700	0.346	44.798	37.12	188.426	20.33	3.24	17.09	4.133	7.791	77.913
n-Butanol	11.47	2.6200	0.301	51.495	47.73	182.176	17.51	2.95	14.56	3.066	8.651	86.513

Table 6: Application of Zwanzig equation for lithium ion in different solvents at 25°C

Solvent	$\lambda_\infty K^+$	$10^2 * \eta^\circ$	$\lambda_\infty K^+ * \eta_\circ$	$[15.5 / \lambda_\infty K^+ * \eta_\circ] = L^*$	$10^{11}\tau$	$10^{10}\tau/\eta$	$\epsilon_\circ$	ϵ_∞	$\epsilon_0 - \epsilon_\infty$	$10^{-2}\epsilon_\circ^2$	$10^{10}R^* = 10^{10}\left[\frac{\tau}{\eta_o}(\epsilon_0 - \epsilon_\infty)/\epsilon_0^2)\right]$	$10^{11}R^*$
Water	73.50	0.8903	0.654	23.700	0.83	9.323	78.54	5.5	73.04	61.69	0.110	1.104
Acetonitrile	102.92	0.3409	0.351	44.160	3.8	111.470	36.61	1.81	34.80	13.40	2.894	28.943
Methanol	52.44	0.5448	0.286	54.196	6.9	126.652	33.05	5.3	27.75	10.92	3.218	32.176
Ethanol	23.55	1.0830	0.255	60.784	14.4	132.964	24.77	4.5	20.27	6.136	4.393	43.927
n-Propanol	12.92	1.9700	0.255	60.784	37.12	188.426	20.33	3.24	17.09	4.133	7.791	77.913

Table 7: Application of Zwanzig equation for potassium ion in different solvents at 25°C

Solvent	$\lambda_\infty Li^+$	$10^2 * \eta_\circ$	$\lambda_\infty Li^+ * \eta_\circ$	$[15.5 / \lambda_\infty Li^+ * \eta_\circ] = L^*$	$10^{11}\tau$	$10^{10}\tau/\eta_\circ$	$\epsilon_\circ$	ϵ_∞	$\epsilon_0 - \epsilon_\infty$	$10^{-2}\epsilon_\circ^2$	$10^{10}R^* = 10^{10}\left[\frac{\tau}{\eta_o}(\epsilon_0 - \epsilon_\infty)/\epsilon_0^2\right]$	$10^{11}R^*$
Water	38.70	0.8903	0.345	44.928	0.83	9.323	78.54	5.5	73.04	61.69	0.110	1.104
Acetonitrile	77.25	0.3409	0.263	58.935	3.8	111.470	36.61	1.81	34.80	13.40	2.894	28.943
Methanol	39.55	0.5448	0.215	72.093	6.9	126.652	33.05	5.3	27.75	10.92	3.218	32.176
Ethanol	17.07	1.0830	0.185	83.784	14.4	132.964	24.77	4.5	20.27	6.136	4.393	43.927
n-Propanol	9.62	1.9700	0.190	81.579	37.12	188.426	20.33	3.24	17.09	4.133	7.791	77.913

Table 8: Application of Zwanzig equation for rubidium ion in different solvents at 25°C

Solvent	$\lambda_\infty Rb^+$	$10^2 * \eta_\circ$	$\lambda_\infty Rb^+ * \eta_\circ$	$[15.5 / \lambda_\infty Rb^+ * \eta_\circ] = L^*$	$10^{11}\tau$	$10^{10}\tau/\eta_\circ$	$\epsilon_\circ$	ϵ_∞	$\epsilon_0 - \epsilon_\infty$	$10^{-2}\epsilon_\circ^2$	$10^{10}R^* = 10^{10}\left[\frac{\tau}{\eta_o}(\epsilon_0 - \epsilon_\infty)/\epsilon_0^2\right]$	$10^{11}R^*$
Water	77.80	0.8903	0.693	22.367	0.83	9.323	78.54	5.5	73.04	61.69	0.110	1.104
Acetonitrile	85.73	0.3409	0.292	53.082	3.8	111.470	36.61	1.81	34.80	13.40	2.894	28.943
Methanol	57.38	0.5448	0.313	49.521	6.9	126.652	33.05	5.3	27.75	10.92	3.218	32.176
Ethanol	25.43	1.0830	0.275	56.364	14.4	132.964	24.77	4.5	20.27	6.136	4.393	43.927
n-Propanol	13.11	1.9700	0.258	60.078	37.12	188.426	20.33	3.24	17.09	4.133	7.791	77.913

Table 9: Application of Zwanzig equation for cesium ion in different solvents at 25°C

Solvent	$\lambda_\infty Cs^+$	$10^2 * \eta_\circ$	$\lambda_\infty Cs^+ * \eta_\circ$	$[15.5 / \lambda_\infty Cs^+ * \eta_\circ] = L^*$	$10^{11}\tau$	$10^{10}\tau/\eta_\circ$	$\epsilon_\circ$	ϵ_∞	$\epsilon_0 - \epsilon_\infty$	$10^{-2}\epsilon_\circ^2$	$10^{10}R^* = 10^{10}\left[\frac{\tau}{\eta_o}(\epsilon_0 - \epsilon_\infty)/\epsilon_0^2\right]$	$10^{11}R^*$
Water	77.30	0.8903	0.688	22.529	0.83	9.323	78.54	5.5	73.04	61.69	0.110	1.104
Acetonitrile	101.63	0.3409	0.346	44.798	3.8	111.470	36.61	1.81	34.80	13.40	2.894	28.943
Methanol	60.83	0.5448	0.331	46.828	6.9	126.652	33.05	5.3	27.75	10.92	3.218	32.176
Ethanol	26.46	1.0830	0.287	54.007	14.4	132.964	24.77	4.5	20.27	6.136	4.393	43.927
n-Propanol	14.33	1.9700	0.282	54.965	37.12	188.426	20.33	3.24	17.09	4.133	7.791	77.913

In case of alkali metal ions (Li^+, K^+, Rb^+ and Cs^+), the value of (r_i) obtained from the intercept decreases with increasing the size from Li^+ to Cs^+ ions but, The value of (r_i) obtained from the slope increases with increasing of the size from Na^+ to Cs^+ ions.

The increasing in the values of hydrated radii obtained from slope more than hydrodynamic radii from intercept refers to occurrence of solvation.

Nightingale [25] and kay et al. [26], have calculated the hydrated radii and hydrodynamic radii for tetra-alkyl ammonuim ions. They found that the hydrated radius increases with increasing ion size. The intercept value reflects factor closely related to the ion solvent interaction for a particular ion. Evan and Gardam. [27], applied F.B.Z equation for tetra alkylammonuim salts in MeOH, EtOH , n-PrOH and n-BuOH at 25°C. They attributed the scattering of the points and difference between the (r_i intercept) and (r_i slope) values to certain specific factors such as solvation which are ignored in the continuum model and these play an important role in determining ionic mobility. El-Hammamy et al. [24], calculated the hydrated radii and hydrodynamic radii for acetylcholine ion and Cl^-, Br^-, I^-, ClO_4^- and s-alkyl isothiouronium ions (s-$Meis^+$, s-n-$Buis^+$, s-n-$Amis^+$ and s-n-$Heptis^+$) in H_2O, MeOH, n-PrOH and CH_3CN at 25°C.

They found that the radii from the slope increase with increase in size for both cation and anions. The increase in the value of hydrodynamic radii (r_i intercept) for (s-$Meis^+$, s-n-$Buis^+$, s-n-$Amis^+$ and s-n-$Heptis^+$) with increase in ion size may also be attributed to ion solvation Interaction.

El-Hammamy et al.[28], have applied Zwanzig theory of dielectric friction for chloride ion in different solvents at 25°C. They compared between the values of radius (r_i) obtained from the slope and from the intercept of the straight line for Cl^- ion and it was noticed that the radius from both of slope and intercept slightly decreases with increasing the ionic size from Cl^- to I^-. This may be attributed to the decrease in solvation.

El-Hammamy et al.[29], have determined the values of the radius from slope (hydrated radius) and intercept (hydrodynamic radius) for Br^-, I^- and ClO_4^- in water, methanol and acetonitrile at 30°C by applying Zwanzig theory. They found that the radius from slope and intercept increases with increasing the size of ions. This was due to the solvation of ions.

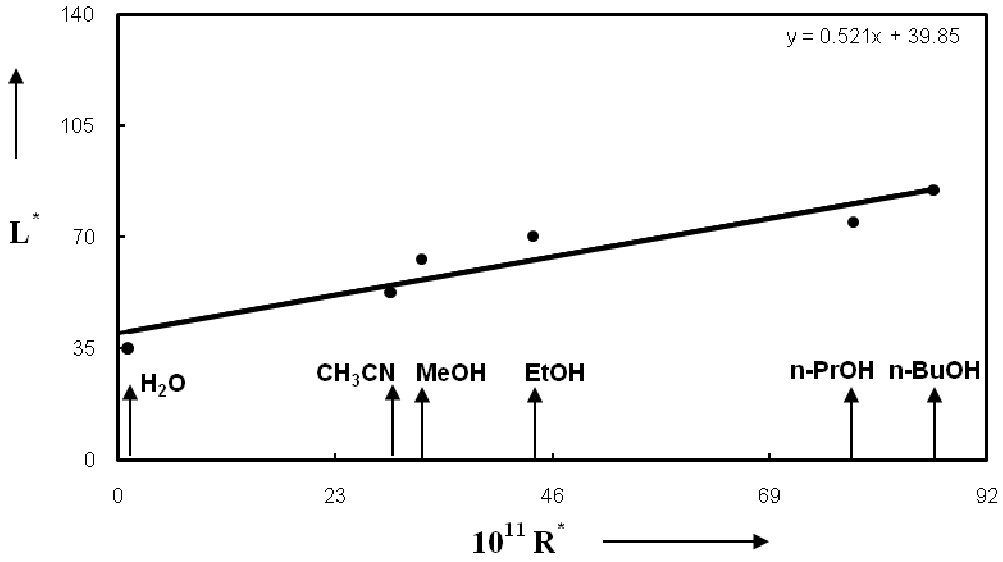

Fig. 1: A plot of eqn. (3) for the for sodium ion (Na^+) in water, methanol, ethanol, n-propanol, n-butanol and acetonitrile at 25°C

Table 10: Hydrodynamic radii (from intercept) and hydrated radii (from slope) of F.B.Z equation for Na^+ and DDC^-

Ion	$r_i A^\circ$ "Hydrodynamic radii" (intercept)	$r_i A^\circ$ "Hydrated radii" (Slope)	$r_i A^\circ$ (Pauling)
λ_∞ Na^+ (Sodium ion)	2.12	6.65	0.95
λ_∞ DDC^- (Diethyldithiocarbamate)	1.89	8.37	----

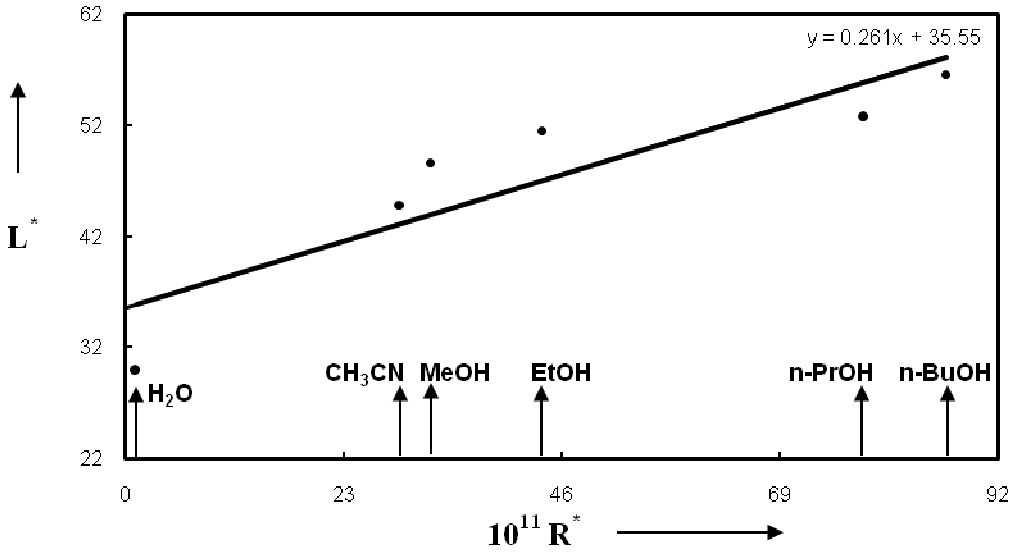

Fig. 2: A plot of eqn. (3) for diethyldithiocarbamate ion $(DDC)^-$ in water, methanol, ethanol, n-propanol, n-butanol and acetonitrile at 25˚C

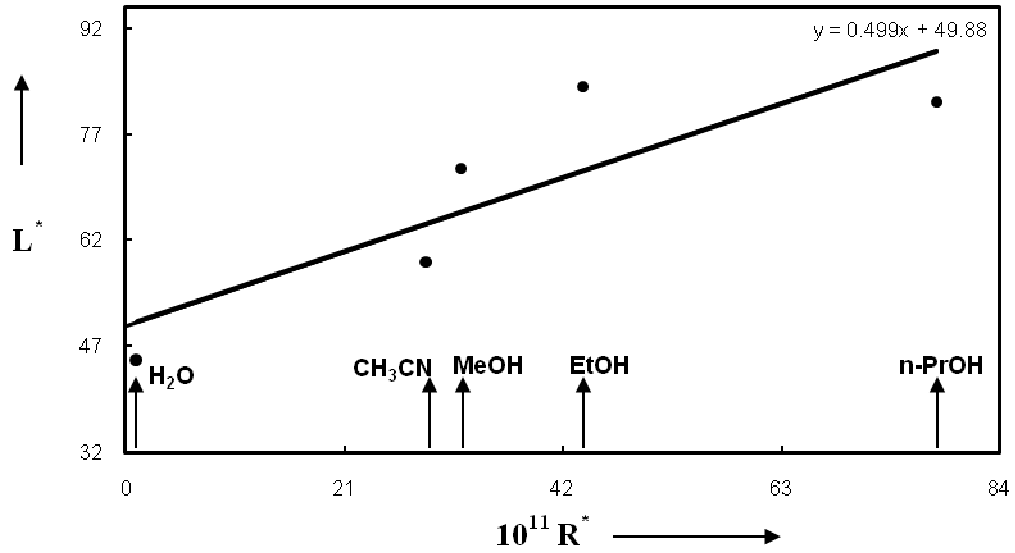

Fig. 3: A plot of eqn. (3) for the for lithium ion (Li^+) in water, acetonitrile, methanol, ethanol and n-propanol at 25˚C

Table 11: Hydrodynamic radii (from intercept) and hydrated radii (from slope) of F.B.Z equation for alkali metal ions (Li^+, Na^+, K^+, Rb^+ and Cs^+

Ion	$r_i A^\circ$ "Hydrodynamic radii" (intercept)	$r_i A^\circ$ "Hydrated radii" (Slope)	$r_i A^\circ$ (Pauling)
Li^+	2.65	6.74	0.60
Na^+	2.12	6.65	0.95
K^+	1.65	6.84	1.33
Rb^+	1.67	6.93	1.48
Cs^+	1.57	7.21	1.69

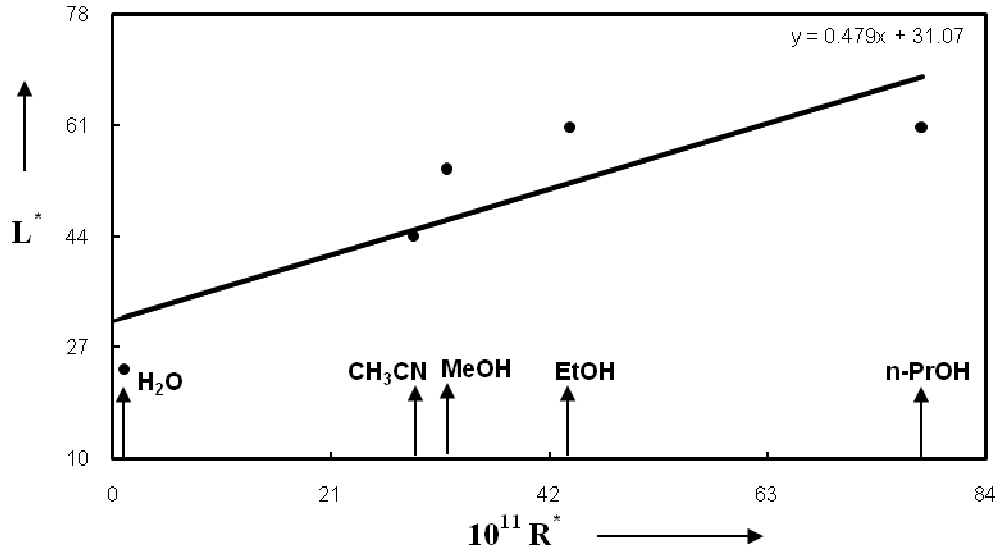

Fig. 4: A plot of eqn. (3) for the for potassium ion (K^+) in water, acetonitrile, methanol, ethanol and n-propanol at 25˚C

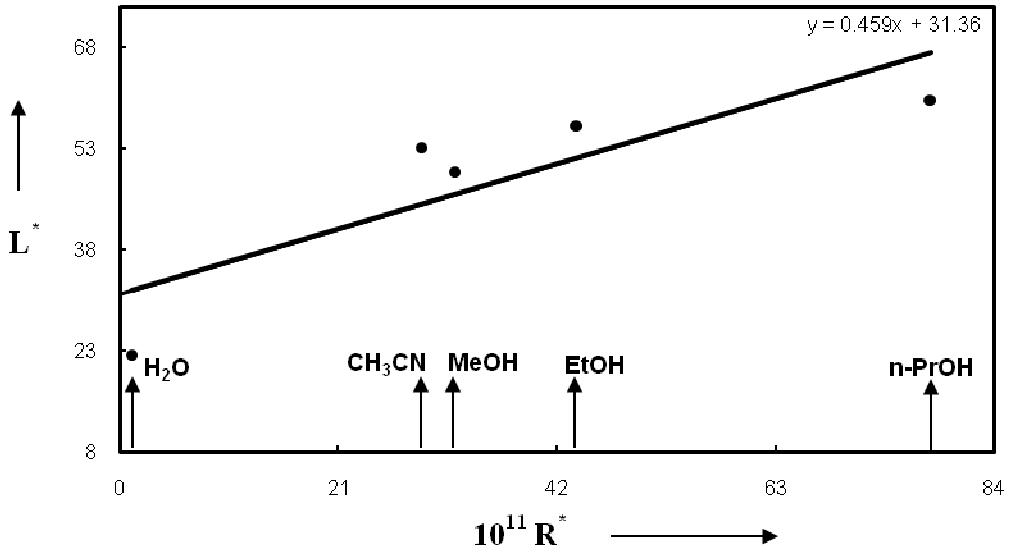

Fig. 5: A plot of eqn. (3) for the for rubidium ion (Rb^+) in water, acetonitrile, methanol, ethanol and n-propanol at 25˚C

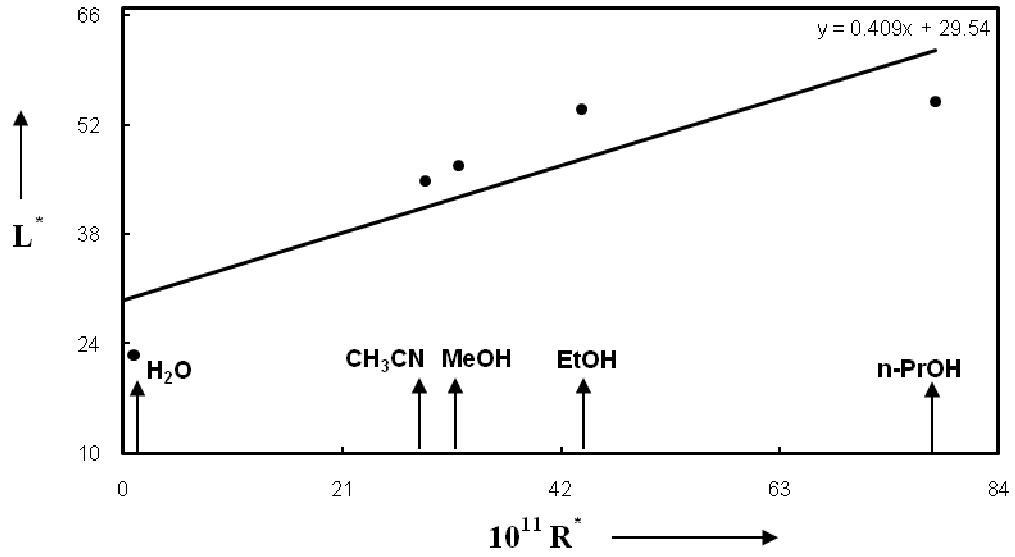

Fig. 6: A plot of eqn. (3) for the for cesium ion (Cs^+) in water, acetonitrile, methanol, ethanol and n-propanol at 25˚C

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