Conductance studies of sodium salts of some aliphatic carboxylic acids in water at different temperatures

Zdzisław Kinart

Department of Physical Chemistry, Faculty of Chemistry, University of Lodz, 90 – 236 Lodz, Pomorska 163/165, Poland

ARTICLE INFO

ABSTRACT

In this paper the electric conductivities of aqueous solutions of sodium salts of aliphatic carboxylic acids of the $H(CH_2)_nCOOH$ [n = 0 to 9] types were measured from T/K = (283.15 to 313.15) in the concentration range $0.0005 < c/mol \cdot dm^{-3} < 0.0200$. In terms of the limiting molar conductance (Λ_o) the measured conductance results have been analyzed by the Fuoss – Justice conductance-concentration equation. From these data the carboxylic ionic contributions ($\lambda_{H(CH_2)_nCOO^{-}}^{o}$) to the limiting molar conductance have been estimated. In this paper the Eyring's enthalpies of activation of charge transport ΔH^* were also calculated. The variations of $\lambda_{H(CH_2)_nCOO^{-}}^{o}$ and ΔH^* as the function of the aliphatic chain length of analyzed carboxylic acids were determined.

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1. Introduction

In our previous paper the apparent molar volumes V_{Φ} of sodium salts of the $H(CH_2)_nCOONa$ [n = 1 to 9] types in dilute aqueous solution were determined by density measurements, at T = 298.15 K [1]. From these data the values of limiting partial molal volume $\overline{V_2^0}$ were calculated and the variations of $\overline{V_2^0}$ values with aliphatic chain length of analyzed salts of carboxylic acids were determined. This paper is a continuation of our studies of physicochemical and thermodynamic properties of carboxylic acids and their salts in water [1–5].

The review of the literature shows that only some studies on the electric conductivities of sodium salts of carboxylic acids in water, at 298.15 K, were made [6–8]. Until now, there have not been carried out studies on the analysis of changes of limiting molar conductance linked with the structure and length of the hydrophobic chain of the studied electrolytes.

Therefore in the present studies, the electric conductivities of aqueous solutions of sodium salts of aliphatic carboxylic acids of the $H(CH_2)_nCOOH$ [n = 0 to 9] types were measured from T/K = (283.15 to 313.15) (in the steps of 5 K) in the concentration range $0.0005 < c/mol \cdot dm^{-3} < 0.0200$. From these data the carboxylic ionic contributions ($\lambda_{H(CH_2)_nCOO^{-}}^{o}$) to the limiting molar conductance and Eyring's enthalpies of activation of charge transport ΔH^* have been estimated. The variations of $\lambda_{H(CH_2)_nCOO^{-}}^{o}$ and ΔH^* with aliphatic chain length of all analyzed carboxylic acids were determined.

2. Experimental section

2.1. Materials

Sodium salts of methanoic, ethanoic, propanoic, butanoic, pentanoic, hexanoic, heptanoic, octanoic and nonanoic acid of high degree of purity were used (all information of purity and Producers of these salts are presented in Table 1). All analyzed sodium salts of carboxylic acids were twice recrystallized from aqueous ethanol solutions and dried under vacuum at T = 383 K for 2 days before use.

Potassium chloride (0.9999, Merck) was dried for 2 days at T = 373 K and was used to determine the conductance cell constant.

Double distilled, deionized and degassed water with a specific conductance better than $0.5 \cdot 10^{-6}$ S·cm^{-1} was used for the preparation of the mixed solvents.

2.2. Methods

All the solutions were prepared by mass using an analytical balance (Sartorius RC 210D) with an uncertainty of $\pm 1 \cdot 10^{-5}$ g. The experimental procedure for conductometric measurements were described in our previous papers [9,10]. Conductance measurements were carried out on a RLC Wayner- Kerr 6430B conductivity meter an uncertainty of 0.02%, using a three-electrode cell (similar to that which is described in work [11]). The cell was calibrated by using aqueous potassium chloride solution [12]. For all measurements the calibration thermostat BU 20F (Lauda, Germany) type with a stability better than 0.005 K was used. In addition, the temperature was controlled using a thermometer Amarell 3000TH AD (Germany). The thermostat has been connected with a through-flow cooler DLK 25 (Lauda, Germany). Conductivity

E-mail address: zkinart@uni.lodz.pl.

Table 1
Specification of chemical samples.

Chemical name	Chemical formula	Source	CAS No.	Mass fraction purity
Sodium formate	$HCOONa$	Sigma Aldrich	141-53-7	≥0.99
Sodium acetate	CH_3COONa	Sigma Aldrich	127-09-3	≥0.99
Sodium propionate	$CH_3(CH_2)COONa$	Sigma Aldrich	137-40-6	≥0.99
Sodium butyrate	$CH_3(CH_2)_2COONa$	Sigma Aldrich	156-54-7	≥0.985
Sodium pentanoic	$CH_3(CH_2)_3COONa$	Toronto Research Chemicals	6106-41-8	≥0.99
Sodium hexanoate	$CH_3(CH_2)_4COONa$	Sigma Aldrich	10,051-44-2	0.99–1
Sodium heptanoate	$CH_3(CH_2)_5COONa$	Shanghai jinjinle Industry Co	10,051-45-3	>0.99
Sodium octanoate	$CH_3(CH_2)_6COONa$	Sigma Aldrich	1984-06-1	≥0.99
Sodium nonanoate	$CH_3(CH_2)_7COONa$	Tokyo Chemical Industry	14,047-60-0	>0.98
Sodium decanoate	$CH_3(CH_2)_8COONa$	Sigma Aldrich	1002-62-6	≥0.98

measurements were performed at the different frequencies, ν, (0.2, 0.5, 1, 1.5, 2, 3, 5, 10, 20) kHz.

All measured conductance values, $\lambda = 1/R_\infty$, were the results of an extrapolation of the cell resistance, $R_\infty(\nu)$, to infinite frequency, $R_\infty = \lim_{\nu \to \infty} R(\nu)$ using the empirical function $R(\nu) = R_\infty + A/\nu$, where parameter A is specific to the cell. Considering the sources of error (calibration, purity of samples, measurements), the estimated uncertainty of the measured values of conductivity was estimated to be ±0.05%.

3. Results and discussion

The experimental molar conductances (Λ) and corresponding molalities (m) for the investigated salts in the mixed solvent are collected in Table S 1.

For convert molonity, $\tilde{m}$, (moles of electrolyte per kilogram of solution) into molarity, c, the values of density gradients, b, have been determined on the basis of density solutions measurements from the equation:

$$c/\tilde{m} = \rho = \rho_o + b \cdot \tilde{m} \tag{1a}$$

where ρ_o is the density of the solvent. The values of measured densities of water and the salts solutions as a function of temperature are presented in Table S 1. The density gradients and the molar conductances Λ as a function of molality m (moles of electrolyte per kilogram of solvent) and temperature are presented in Table S 1. The relationship among m, $\tilde{m}$, and c is the following:

$$\tilde{m} = c/\rho = m/(1 + m \cdot M) \tag{1b}$$

where M is the molar mass of electrolyte.

The conductance data were analyzed using the Fuoss-Justice equation [13,14] in the form:

$$\Lambda = \alpha\left[\Lambda_o - S(\alpha c)^{1/2} + E(\alpha c)\,\ln(\alpha c) + J(\alpha c) + J_{3/2}(\alpha c)^{3/2}\right] \tag{2}$$

together with

$$K_A = \frac{1-\alpha}{\alpha^2 c \gamma_\pm^2} \tag{3}$$

and

$$\ln\gamma_\pm = -\frac{A\alpha^{1/2}c^{1/2}}{1 + BR\alpha^{1/2}c^{1/2}} \tag{4}$$

In these equations, Λ_o is the limiting molar conductance; α is the dissociation degree of an electrolyte; K_A is the ionic association constant; R is the distance parameter of ions; $\gamma_\pm$ is the activity coefficient of ions on the molar scale; and A and B are the coefficients of the Debye Hückel

equation. The analytical form of the parameters S, E, J, and $J_{3/2}$ were presented previously [15–17].

The values of Λ_o for the all investigated salts in all measured temperatures were obtained using the well-known procedure given by Fuoss [13,14]. The calculations were carried out assuming that $R = q$ (q - Bjerrum distance [18]). Example of the graphical variations of $\Lambda(\exp)$ – $\Lambda(calc)$ as a function of concentration is shown in Fig. 1.

In Table S 2 have been collected the limiting molar conductance Λ_o and standard deviations $\sigma(\Lambda)$ of sodium salts in water, different temperatures.

The literature data of molar conductivity slightly different for the first representatives of the homologous series of salts of monocarboxylic acids. As we can see on the Fig. S1 the literature values for CH_3COONa [19–23] and on the Fig. S2 for $CH_3(CH_2)COONa$ [23] differ slightly from the values presented in this work. These differences are mainly in the curvature of the presented functions but the values of molar conductivity obtained in this work are comparable to those for the literature. This effect disappears with increasing chain length of the studied salts of aliphatic carboxylic acids and it is shown on the Fig. S3 when our results are presented with comparing of literature values for the $CH_3(CH_2)_2COONa$ [23]. The most likely, that these effects are due by the smaller precision of apparatus for the conductometric measurements used in those years (see papers [19–23]) and the methodology.

Fig. S4 shows a comparison of the course of the function change $\Lambda = f(m)$ for $CH_3(CH_2)_4COONa$ in water at 298.15 K which are presented in this work and literature. As we can see, the values presented in the work [24] have a slightly different slope in relation to the values discussed in
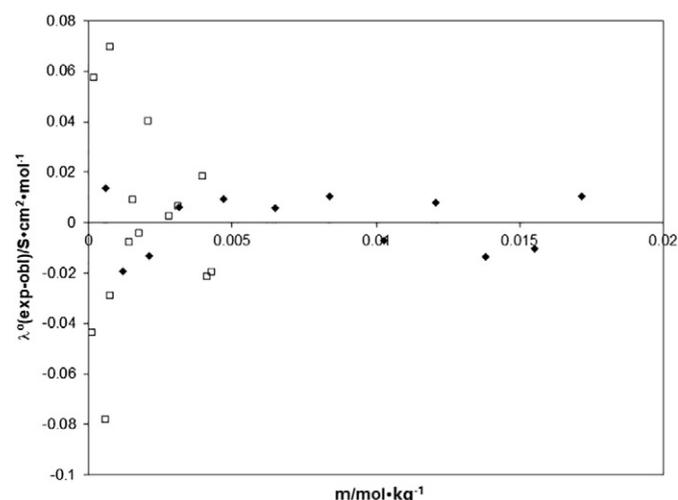

Fig. 1. Plot of the function $\lambda^0(\exp\text{-calk}) = f(m)$ ♦ experimental and □ literature [7].

Table 2
Literature densities ρ_0, viscosities η, relative permittivities ε of water and Bjerrum parameters q at different temperatures, at pressure p = 0.1 MPa.

$\frac{T}{K}$	$\frac{\rho_0}{g \cdot cm^{-3}}$ [a]	$\frac{\eta}{mPa \cdot s}$ [b]	ε [c]	$\frac{q}{10^{-8} \, cm}$	$\frac{\lambda^o_{Na^+}}{S \cdot cm^2 \cdot mol^{-1}}$ [d]
283.15	0.99970	0.01303	83.945	3.52	34.88
288.15	0.99910	0.01374	82.039	3.54	39.72
293.15	0.99821	0.01002	80.176	3.56	44.81
298.15	0.99704	0.00890	78.358	3.58	50.15
303.15	0.99565	0.00797	76.581	3.60	55.72
308.15	0.99404	0.00719	74.846	3.62	61.53
313.15	0.99221	0.00652	73.151	3.65	67.34

[a] Ref. [28].
[b] Ref. [29].
[c] Ref. [30].
[d] Ref. [11].

this work. These differences may be due to different reasons. The most probable explanation this fact may be the use by the authors of paper [24] less accurate measuring instruments, whose measurement error was ± 0.5%. It is likely because, that the same presented and discussed values are cited in [25] and this problem is hardly visible. In other cases presented for $CH_3(CH_2)_6COONa$ on Fig. S5 at 295.15 K and on Fig. S7 at 308.15 K the value adjustment is almost perfect [26]. In the work [27] describing the same values for $CH_3(CH_2)_6COONa$ (see Fig. S6), there is a similar problem with different conductivity values. In the first place, it should be emphasized that the authors of this work do not provide values in low salt concentrations, and we do not know exactly how the course of this relationship will look. The second cause of the presented differences is a larger measurement error (as in work [25]) which can significantly influence the results of the measurement. This error will bind with a less accurate measuring bridge. The authors of discussed works used the conductivity cell (Model 011510, Thermo Orion), which is much less accurate than the one used in this work. It should also be noted, that the authors of the discussed works also benefit from a more simplified model of counting of conductivity. All these discussed factors may affect on differences in the course of changes of the analyzed functions.

The literature values of density, viscosity and relative permittivity of water, which are necessary for the calculation of these parameters, are presented in Table 2.

There are also collected in Table 2 the literature values of limiting molar conductances for Na^+ cation measured in water at seven temperatures. These data enabled us to calculate the limiting ionic conductances of individual carboxylic anions using the following dependences:

$$\lambda^0_{A^-} = \Lambda_o - \lambda^o_{Na^+}$$

In this equation, Λ_o is the limiting molar conductance of sodium salts of the studied carboxylic acids.

The obtained values of $\lambda_{A^-}{}^o$ are collected in Table 3. For comparative purposes values of $\lambda_{A^-}{}^o$ at 298.15 K measured by us and the literature data are brought together in Table 4. The greatest divergences of these values are visible particularly for anions $H(CH_2)_nCOO^-$ when n > 3. These differences can be presumably explained by low precision of that mentioned above data published in the first half of the 20-th century.

The dependences of limiting conductances of studied carboxylic ions ($\lambda_{A^-}{}^o$) as a function of number of – CH_2 – groups in the chain anion of the studied acid (n) are shown at Fig. 2.

Parallely for comparison, there are shown at Fig. 3 the dependences of $\lambda_{A^-}{}^o$ as the function of n at 298.15 K together with the available literature data. The course of changes of discussed functions univocally indicates that for the dependence $\lambda_{A^-}{}^o = f(n)$ based on the literature data it is difficult to describe more determined character.

Whereas, in case of values $\lambda_{A^-}{}^o$ obtained in the present study the dependence $\lambda_{A^-}{}^o = f(n)$ monotonously decreases at all measuring temperatures (see also Fig. 2). The more precise analysis of obtained by us results show that the dependence $\lambda_{A^-}{}^o = f(n)$ can be described by the following dependence:

$$\lambda^0_{A^-} = ae^{-bn} + c \tag{5}$$

The values of parameters a, b and c have been calculated using the nonlinear least square method and obtained data are collected in Table 5.

Table 3
Experimental values of limiting equivalent conductances, $\lambda^0_{(A^-)}$, of studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ in water, literature values of limiting molar conductances, $\lambda^0_{(Na^+)}$, of sodium cation in water, different temperatures at pressure.

T/K	$\frac{\lambda^o(A^-)}{S \cdot cm^2 \cdot mol^{-1}}$									
	n = 0	n = 1	n = 2	n = 3	n = 4	n = 5	n = 6	n = 7	n = 8	n = 9
283.15	38.07	27.01	24.02	21.80	19.90	18.26	16.85	15.64	14.60	13.70
288.15	43.56	31.29	27.98	25.54	23.45	21.65	20.10	18.77	17.63	16.65
293.15	49.05	35.56	31.94	29.28	27.01	25.05	23.36	21.91	20.67	19.59
298.15	54.51	39.81	35.86	33.00	30.53	28.41	26.59	25.02	23.67	22.51
303.15	59.95	44.04	39.77	36.69	34.03	31.75	29.79	28.10	26.65	25.41
308.15	65.43	48.30	43.72	40.42	37.58	35.14	33.04	31.23	29.68	28.34
313.15	70.93	52.60	47.70	44.18	41.15	38.55	36.31	34.39	32.73	31.31

Table 4

Experimental and literature values of limiting equivalent conductances, $\lambda^0_{(A^-)}$, of studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ in water, at 298.15 K.

n	Salts	$\dfrac{\lambda^0_{exp}(A^-)}{S \cdot cm^2 \cdot mol^{-1}}$	$\dfrac{\lambda^0_{lit}(A^-)}{S \cdot cm^2 \cdot mol^{-1}} Z$
0	HCOONa	54.51	54.64[a]
1	CH₃COONa	39.81	40.92[b]
2	CH₃CH₂COONa	35.86	36.26[b]
3	CH₃(CH₂)₂COONa	33.00	32.37[b]
4	CH₃(CH₂)₃COONa	30.53	33.68[d,e]
5	CH₃(CH₂)₄COONa	28.41	31.09[c]
6	CH₃(CH₂)₅COONa	26.59	28.14[c]
7	CH₃(CH₂)₆COONa	25.02	28.17[c]
8	CH₃(CH₂)₇COONa	23.67	25.40[c]
9	CH₃(CH₂)₈COONa	22.51	–

[a] Ref. [6].
[b] Ref. [7].
[c] Ref. [8].
[d] Ref. [31].
[e] Ref. [32].

It seems worth mentioning that values of these parameters have been determined without taking into consideration the limiting conductances $\lambda_{A^-}{}^o$ of $HCOO^-$ and CH_3COO^- ions, since they not fulfilled (differently from values $\lambda_{A^-}{}^o$ for other anions) the dependence:

$$\ln\left(\lambda^0_{A^-} - c\right) = \ln a - bn \tag{6}$$

The discussed dependence Eq. (6) may be obtained by simple mathematical transformation of the Eq. (5). The plots of the function $\ln(\lambda_{A^-}{}^0 - c) = \ln a - bn$ are presented on Fig. 4.

Most likely, that the experimental data obtained for the formate ion do not obey Eqs. (5) and (6) are results of this that this ion has no alkyl fragment in structure.

The Eqs. (5) and (6) are much better fulfilled for CH_3COO^- than for the $HCOO^-$ anion. However, the value of $\lambda_{A^-}{}^0$ for formic anion has not been used for calculations of a, b, and c (see Table 4), since as it has been postulated by authors of other works [1,33–35] (where the

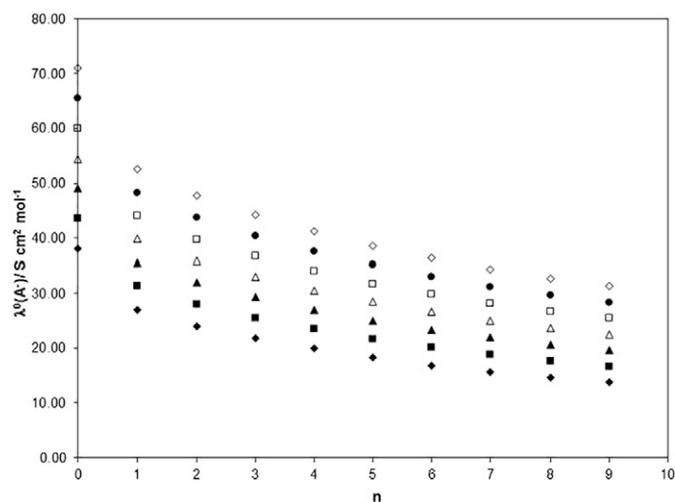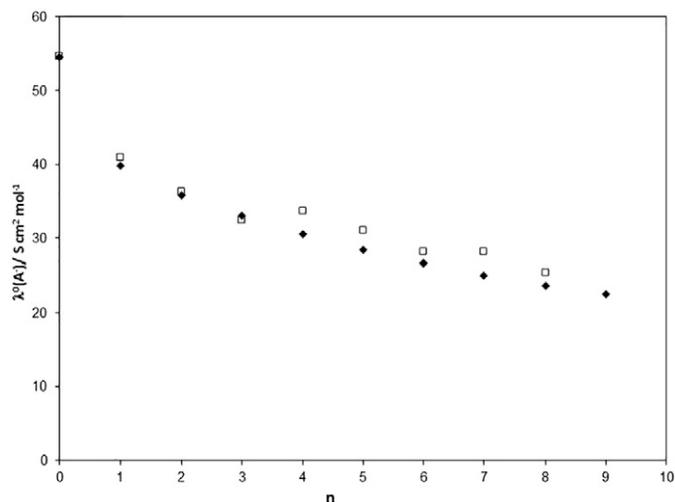**Fig. 3.** The dependences of the ♦ experimental and □ literature [6–8,30,31] values of the limiting ionic conductances of the studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ as a function of carbon atoms (n) in these molecules.

apparent molar volumes of carboxylic anions have been studied), the charged COO^- group influences considerably the hydration sphere of the nearest $-CH_2-$ group.

As it results from the examination of data which are shown in Table 5, values of parameters a and c systematically increase with the growth of the temperature which is not surprising since with increase of temperature also the conductivity of ions increases. Whereas, the values of parameter b are independent from temperature.

The transformation of the Eq. (5) leads to the following dependence:

$$\Delta\lambda^0 = \lambda^0_{n+1} - \lambda^0_n = A_2 A_1^n \tag{7}$$

where $A_1 = e^{-b}$, $A_2 = a(e^{-b} - 1)$.

This dependence is so far important as it enables the precise determination of changes of the conductivity of the anion with its growth by one $- CH_2 -$ group.

Based on the fact that values of limiting conductances were determined at seven temperatures it was possible to calculate values ΔH^* enthalpy of activation of charge transport from the given below equation [36] using data collected in Table 2:

$$\frac{\partial\left(\ln\lambda^0_{A^-} + \dfrac{2}{3}\ln\rho_0\right)}{\partial T} = -\frac{\Delta H^*}{RT^2} \tag{8}$$

Fig. 2. Plot of the function $\lambda_{A^-}{}^o = f(n)$ for studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ in water, at ♦ 283.15 K; ■ 288.15 K; ▲ 293.15 K; △ 298.15 K; □ 303.15 K; ● 308.15 K and ◇ 313.15 K.

Table 5

Values of a, b and c parameters for studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ in water, at different temperatures.

T/K	a	b	c
283.15	21.391	0.151	8.199
288.15	23.486	0.151	10.613
293.15	25.596	0.151	13.006
298.15	27.682	0.151	15.397
303.15	29.769	0.151	17.772
308.15	31.876	0.151	20.152
313.15	33.975	0.151	22.580
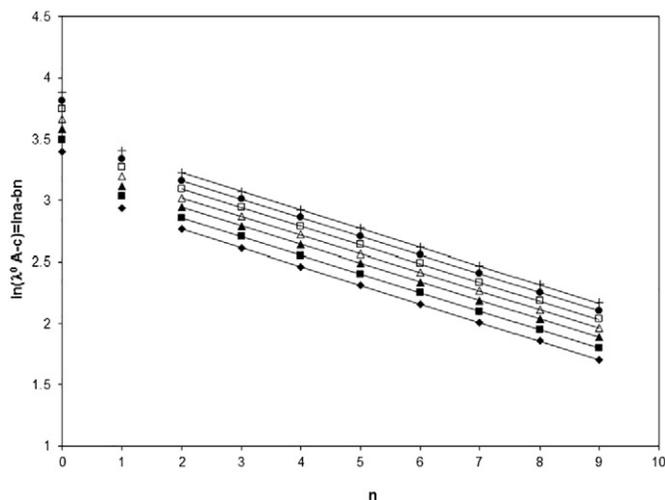
Fig. 4. The dependences of the function $\ln(\lambda_A{}^0 - c) = \ln a - bn$, at ♦ 283.15 K, ■ 288.15 K, ▲ 293.15 K, △ 298.15 K, □ 303.15 K, ● 308.15 K and + 313.15 K.

Table 6
Values of enthalpy of activation of charge transport for studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ in water, at 298.15 K.

n	$\dfrac{\Delta H^*}{kJ \cdot mol^{-1}}$
0	15.088
1	16.144
2	16.614
3	17.098
4	17.575
5	18.073
6	18.558
7	19.037
8	19.500
9	19.959

The plots of the function $\ln \lambda_A^0 + \frac{2}{3} \ln \rho_0 = f(T)$ are presented on Fig. 5. The mentioned above dependences have been represented by the second order polynomial and values ΔH^*, at 298.15 K, have been calculated (see Table 6).

The courses of changes of enthalpy ΔH^* as a function of the number of $-CH_2-$ groups in the alkyl chain of carboxylic anions are shown at Fig. 6. As it can be seen this dependence has a linear character exhibiting a very high correlation coefficient. It indicates that the appearance in the alkyl chain the additional $-CH_2-$ group affects the increase of the value of enthalpy of activation of charge transport by a constant value equal approximately to 0.480 $kJ \cdot mol^{-1}$.

4. Conclusions

The electric conductivities of aqueous solutions of sodium salts of aliphatic carboxylic acids of the $H(CH_2)_nCOOH$ [n = 0 to 9] types were measured from T/K = (2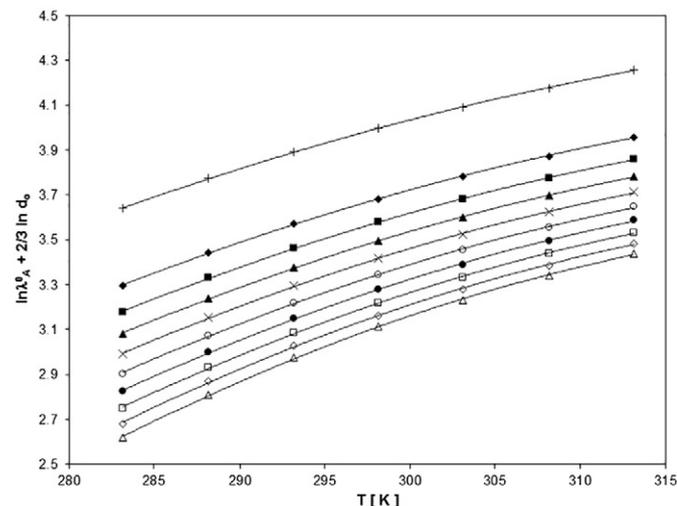83.15 to 313.15) (in the steps of 5 K). The conductance results have been analyzed by the Fuoss – Justice conductance-concentration equation in terms of the limiting molar conductance Λ_0. We have used available in the literature values of limiting molar conductivity of sodium cation to estimate the limiting molar conductivities of carboxylic anions. The equation representing changes of conductivity of these anions as a function of the number of carbon atoms in the aliphatic chain has been proposed. The enthalpies of activation of charge transport ΔH^* have been also calculated and their dependences on the number of carbon atoms (n) and the energy corresponding to a single $-CH_2-$ group has been suggested.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.molliq.2017.10.133.

Fig. 5. Plot of the function $\ln \lambda_{A^-}^0 + \frac{2}{3} \ln \rho_0 = f(T)$ for studied monocarboxylic organic anions $H(CH_2)_nCOO^-$ (+ $HCOO^-$; ♦ CH_3COO^-; ■ $CH_3CH_2COO^-$; ▲ $CH_3(CH_2)_2COO^-$; x $CH_3(CH_2)_3COO^-$; ○ $CH_3(CH_2)_4COO^-$; ● $CH_3(CH_2)_5COO^-$; □ $CH_3(CH_2)_6COO^-$; ◇ $CH_3(CH_2)_7COO^-$; △ $CH_3(CH_2)_8COO^-$) in water.
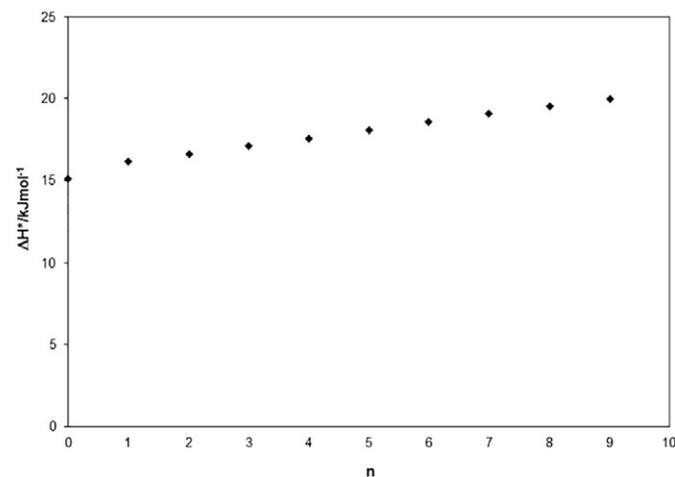

Fig. 6. The dependences of the values of ΔH^* as a function of carbon atoms (n) in the molecules of studied monocarboxylic organic anions $H(CH_2)_nCOO^-$. $\Delta H^* = 0.4796n + 15.664$ (for n = 1 to 9, $R^2 = 0.9999$).

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