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*Alina Piekarska, Stefania Taniewska-Osińska*

# RELATIVE PARTIAL MOLAL ENTHALPY OF NaI SOLUTIONS IN THE MIXTURES OF N,N-DIMETHYLFORMAMIDE WITH WATER

The values of the relative partial molal enthalpy of mixed solvent ($\bar{L}_1$) and electrolyte ($\bar{L}_2$) in NaI-water-N,N-dimethylformamide systems were obtained.

The course of the function of the $\bar{L}_1$, $\bar{L}_2$ and $\bar{L}_1/T$ vs concentration has been discussed.

In our previous works [1, 2] we have presented the data on integral solution heat ($\Delta H_m$) of NaI in the mixtures of N,N-dimethylformamide with water within the whole composition range of the mixed solvent over the temperature 5-35°C.

All isotherms of integral NaI solution heat in the examined mixtures were described by the equation

$$-\Delta H_m = A_o + \frac{A_1}{m + s} + \frac{A_2}{(m + s)^2} + \frac{A_3}{(m + s)^3} \qquad (1)$$

where:

s - assumed parameter;

$A_o$, $A_1$, $A_2$, $A_3$ - polynomial coefficients.

We have assumed the parameter "s" in such a way as to make the difference between $\Delta H_m$ (calculated) and $\Delta H_m$ (experimental) as small as possible.

Using the functions $\Delta H_m = f(m)$ determined in such a way we have calculated relative partial molal enthalpy of the solvent ($\bar{L}_1$) and NaI ($\bar{L}_2$). We have treated the examined three

component system: N,N-dimethylformamide-water-NaI as pseudobinary, assuming that each of the examined mixtures of a given composition is a separate solvent.

We have calculated the values $(\bar{L}_1)$ and $(\bar{L}_2)$ from the formula:

$$\bar{L}_1 = \frac{m\sqrt{m}}{2}\,\frac{M}{1000}\,\frac{d\,(\Delta H_m^o)}{d\sqrt{m}} \qquad (2)$$

$$\bar{L}_2 = -\Delta H_m^o - \frac{\sqrt{m}}{2}\,\frac{d\,(\Delta H_m^o)}{d\sqrt{m}} \qquad (3)$$

where:

m - the electrolyte concentration in mol./kg of the solvent;

$\Delta H_m^o$ - integral dilution heat.

The value of the derivative $d(\Delta H_m^o)/d\sqrt{m}$ was calculated by analytic method using the computer "Odra 1013".

The calculated values $\bar{L}_1$ and $\bar{L}_2$ are presented in tab. 1-4 and fig. 1 and 2 in the form of the function $\bar{L}_1$, $\bar{L}_2 = f(m)$.

It follows from the figures, that alike in the case of previously discussed NaI-water-formamide [3] system the course of the function $\bar{L}_1 = f(m)$ depends on the composition of the mixed solvent. In all the cases three types of isotherms can be distinguished approximately corresponding to three types of isotherms of NaI solution heat in the discussed mixtures.

In the solutions containing from 0 to 30 mol. % DMF the course of relation $\bar{L}_1 = f(m)$ is analogous to that in aqueous NaI solutions. The curves belong to the III type according to M i s h c h e n k o and P r o n i n a' s classification [4]. In the solutions containing above 67 mol. % DMF the course of the discussed function is typical for nonaqueous solvents; the isotherms belong to I-st type [4]. The shape of the discussed function in the solutions containing from ~30 to ~ 67 mol. % DMF may be considered as intermediate between the two described above forms of the curve $\bar{L}_1 = f(m)$.

Similar observations can be made on the basis of the ana-

Table 1

Relative partial molal enthalpy of solvent ($\bar{L}_1$) and NaI ($\bar{L}_2$) in NaI solutions in water-N,N-dimethylformamide mixtures (t = 5°C)

| m [mol. NaI / kg] | Mol. % of DMF in the mixed solvent | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | 0 | 5 | 15 | 30 | 50 | 67 | 80 | 95 | 100 |
| | | | | $L_1$ [cal/mol.] | | | | | |
| 0.03 | -0,011 | -0.021 | -0.028 | -0.113 | -0.103 | -0.174 | -0.263 | -0.551 | -0.681 |
| 0.05 | -0.023 | -0.050 | -0.065 | -0.210 | -0.202 | -0.345 | -0.549 | -0.800 | -1.096 |
| 0.10 | -0.053 | -0.093 | -0.154 | -0.281 | -0.421 | -0.790 | -1.202 | -1.480 | -1.815 |
| 0.20 | +0.121 | +0.137 | -0.174 | +0.240 | -0.843 | -2.049 | -1.982 | -3.653 | -6.690 |
| 0.50 | +1.53 | +1.892 | +0.579 | +2.834 | -2.430 | -7.375 | -- | -- | -- |
| 1.00 | +8.01 | +4.260 | +1.952 | +5.666 | -- | -- | -- | -- | -- |
| | | | | $L_2$ [cal/mol.] | | | | | |
| 0.03 | +55 | +67 | +79 | +262 | +182 | +261 | +317 | +734 | +875 |
| 0.05 | +65 | +102 | +114 | +335 | +238 | +341 | +434 | +828 | +1023 |
| 0.10 | +89 | +134 | +162 | +369 | +305 | +453 | +582 | +847 | +1158 |
| 0.20 | +42 | +65 | +171 | +272 | +369 | +608 | +673 | +1091 | +1587 |
| 0.50 | -155 | -186 | +89 | +42 | +475 | +903 | -- | -- | -- |
| 1.00 | -655 | -349 | +15 | -75 | -- | -- | -- | -- | -- |

Table 2

Relative partial molal enthalpy of solvent ($\bar{L}_1$) and NaI ($\bar{L}_2$) in NaI solutions in water-N,N-dimethylformamide mixtures (t = 15°C)

| m [mol. NaI / kg] | Mol. % of DMF in the mixed solvent | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | 0 | 5 | 15 | 30 | 50 | 67 | 80 | 95 | 100 |
| | | | | $L_1$ [cal/mol.] | | | | | |
| 0.03 | -0.011 | -0.029 | -0.052 | -0.119 | -0.098 | -0.239 | -0.322 | -0.686 | -0.722 |
| 0.05 | -0.023 | -0.058 | -0.102 | -0.235 | -0.204 | -0.450 | -0.596 | -1.098 | -1.118 |
| 0.10 | -0.031 | -0.084 | -0.169 | -0.394 | -0.434 | -0.883 | -1.051 | -1.307 | -1.957 |
| 0.20 | +0.081 | +0.106 | -0.022 | -0.050 | -0.694 | -1.909 | -1.694 | -2.201 | -9.216 |
| 0.50 | +0.890 | +1.244 | +1.014 | +2.480 | -1.191 | -6.862 | -- | -- | -- |
| 1.00 | +5.94 | +2.639 | +2.261 | +5.609 | -- | -- | -- | -- | -- |
| | | | | $L_2$ [cal/mol.] | | | | | |
| 0.03 | +62 | +106 | +157 | +261 | +169 | +356 | +423 | +870 | +906 |
| 0.05 | +75 | +141 | +206 | +347 | +228 | +455 | +537 | +1023 | +1048 |
| 0.10 | +87 | +162 | +244 | +417 | +299 | +566 | +642 | +1073 | +1199 |
| 0.20 | +57 | +104 | +210 | +356 | +341 | +693 | +713 | +1147 | +1839 |
| 0.50 | -70 | -61 | +91 | +135 | +374 | +964 | -- | -- | -- |
| 1.00 | -410 | -158 | +23 | +5 | -- | -- | -- | -- | -- |

Table 3

Relative partial molal enthalpy of solvent ($\bar{L}_1$) and NaI ($\bar{L}_2$) in NaI solutions in water-N,N-dimethylformamide mixtures ($t = 25^{\circ}C$)

| m [mol. NaI / kg] | Mol. % of DMF in the mixed solvent | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | 0 | 5 | 15 | 30 | 50 | 67 | 80 | 95 | 100 |
| | | | | $L_1$ [cal/mol.] | | | | | |
| 0.03 | -0.012 | -0.029 | -0.055 | -0.117 | -0.087 | -0.267 | -0.390 | -0.817 | -0.949 |
| 0.05 | -0.036 | -0.056 | -0.088 | -0.216 | -0.162 | -0.536 | -0.703 | -1.210 | -1.548 |
| 0.10 | -0.025 | -0.077 | -0.066 | -0.325 | -0.281 | -1.080 | -1.112 | -1.655 | -1.906 |
| 0.20 | +0.049 | +0.081 | +0.179 | -0.129 | -0.503 | -1.816 | -1.832 | -2.677 | -2.731 |
| 0.50 | +0.808 | +0.963 | +0.656 | +0.745 | -2.549 | -5.032 | -4.166 | -- | -- |
| 1.00 | +4.50 | +2.009 | +0.700 | +1.400 | -- | -- | -- | -- | -- |
| | | | | $L_2$ [cal/mol.] | | | | | |
| 0.03 | +96 | +130 | +209 | +273 | +153 | +373 | +546 | +1038 | +1168 |
| 0.05 | +120 | +163 | +242 | +347 | +195 | +498 | +676 | +1187 | +1382 |
| 0.10 | +104 | +180 | +235 | +395 | +233 | +639 | +774 | +1180 | +1463 |
| 0.20 | +86 | +131 | +172 | +360 | +265 | +734 | +806 | +1306 | +1527 |
| 0.50 | -60 | +2 | +111 | +280 | +394 | +906 | +933 | -- | -- |
| 1.00 | -320 | -69 | +106 | +253 | -- | -- | -- | -- | -- |

Table 4

Relative partial molal enthalpy of solvent ($\bar{L}_1$) and NaI ($\bar{L}_2$) in NaI solutions in water-N,N-dimethylformamide mixtures (t = 35°C)

| m [mol. NaI / kg] | Mol. % of DMF in the mixed solvent | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | 0 | 5 | 15 | 30 | 50 | 67 | 80 | 95 | 100 |
| | | | | $L_1$ [cal/mol.] | | | | | |
| 0.03 | -0.012 | -0.030 | -0.060 | -0.114 | -0.078 | -0.231 | -0.444 | -0.823 | -1.038 |
| 0.05 | -0.022 | -0.061 | -0.097 | -0.206 | -0.159 | -0.528 | -0.876 | -1.255 | -1.765 |
| 0.10 | -0.034 | -0.106 | -0.068 | -0.285 | -0.337 | -1.339 | -1.097 | -1.264 | -2.572 |
| 0.20 | -0.048 | +0.009 | +0.269 | +0.001 | -0.667 | -1.451 | -1.887 | -2.437 | -4.480 |
| 0.50 | +0.522 | +0.983 | +1.180 | +1.133 | -2.206 | -2.845 | -3.851 | -- | -- |
| 1.00 | +3.72 | +2.346 | +1.763 | +2.034 | -- | -- | -- | -- | -- |
| | | | | $L_2$ [cal/mol.] | | | | | |
| 0.03 | +92 | +110 | +196 | +269 | +121 | +294 | +563 | +1037 | +1206 |
| 0.05 | +112 | +149 | +233 | +338 | +166 | +431 | +742 | +1199 | +1465 |
| 0.10 | +109 | +181 | +223 | +375 | +221 | +637 | +931 | +1212 | +1629 |
| 0.20 | +93 | +147 | +136 | +322 | +271 | +784 | +1013 | +1305 | +1795 |
| 0.50 | -3 | +8 | +25 | +219 | +371 | +817 | +1056 | -- | -- |
| 1.00 | -240 | -85 | -8 | +181 | -- | -- | -- | -- | -- |

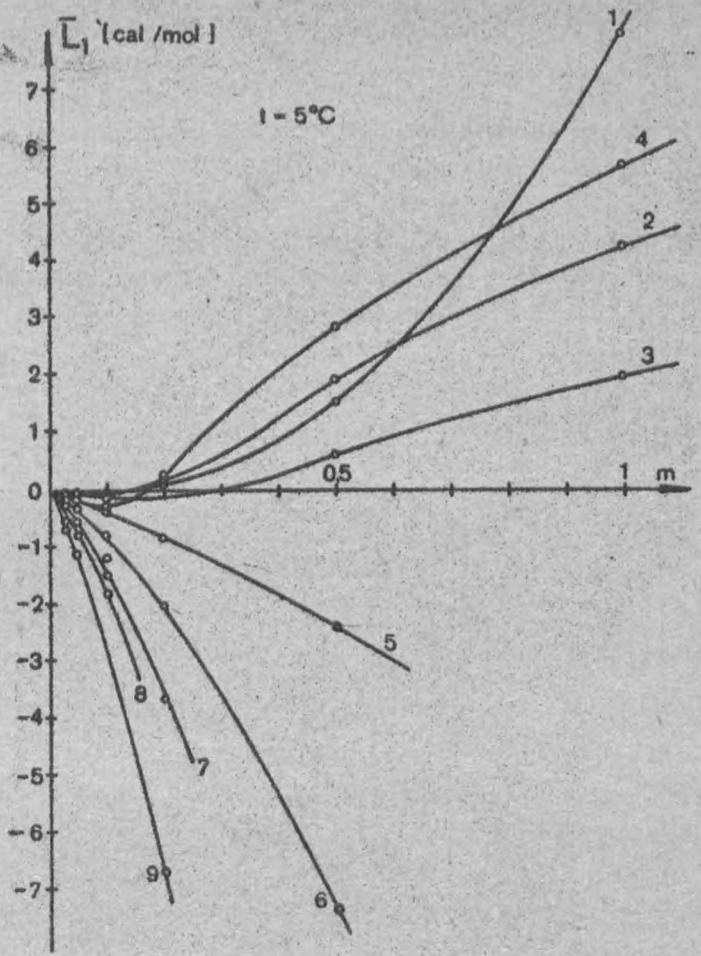


Fig. 1. The course of relation $\bar{L}_1 = f(m)$ of NaI solution in the mixtures N,N-dimethylformamide-water at temp. 5°C. The DMF contents in the mixtures (in mol. %) are: 1 - 0%, 2 - 5%, 3 - 15%, 4 - 30%, 5 - 50%, 6 - 67%, 7 - 80%, 8 - 95%, 9 - 100%

lysis of the concentration dependence of relative partial molal enthalpy of NaI ($\bar{L}_2$) in the examined mixtures (tab. 1-4, fig. 3, 4).

The function illustrating the structural changes in the solution in the best way is entropy [5, 6].

This quantity is connected with relative partial molal enthalpy of solvent ($\bar{L}_1$) by the following relation:

$$\Delta \bar{S}_1^E = \frac{\bar{L}_1 - RT \ln \frac{a_1}{x_1}}{T} \qquad (4)$$

In order to determine the excess of relative partial molal entropy of the solvent using the formula (4) the knowledge of activity of the solvent in the solution is necessary.

In literature we have not found the data concerning the activity of NaI solutions in the mixtures of DMF with water. For this reason we could not calculate the value $\Delta \bar{S}_1^E$ in the examined systems. M i s h c h e n k o and S o k o ł o v' s

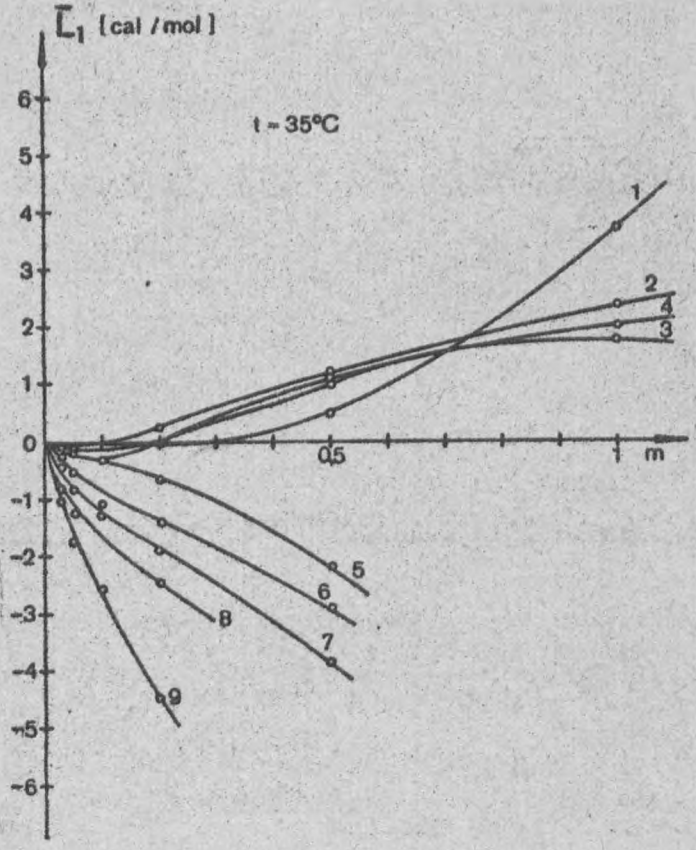


Fig. 2. The course of relation $\bar{L}_1 = f(m)$ of NaI solution in the mixtures N,N-dimethylformamide-water at temp. 35°C. The DMF contents in the mixtures (in mol. %) are; 1 - 0%, 2 - 5%, 3 - 15%, 4 - 30%, 5 - 50%, 6 - 67%, 7 - 80%, 8 - 95%, 9 - 100%

servation [7] which let us assume that the course of relation $\bar{L}_1/T = f(m, T)$ illustrates the effect of the dissolved electrolyte on the solvent structure in a similar way as the course of the curves $\Delta \bar{S}_1^E = f(m, T)$, allowed us to calculate the values $\bar{L}_1/T$ of NaI solutions in DMF-water mixtures. The results are presented in tab. 5-8 and in fig. 5, 6 as the relation $\bar{L}_1/T = f(m)$.

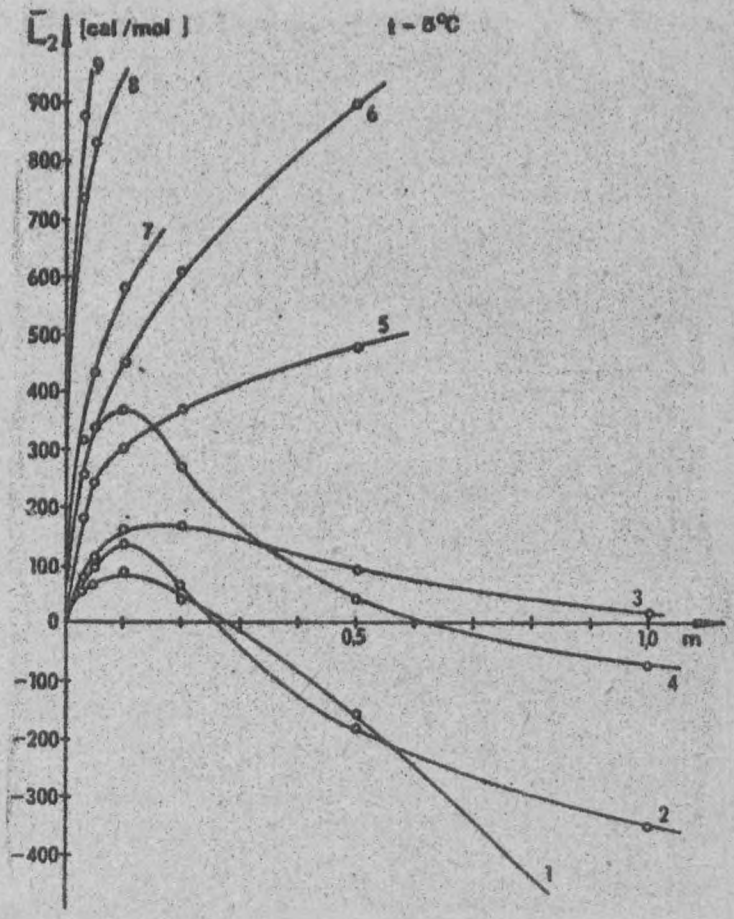


Fig. 3. The course of relation $\bar{L}_2 = f(m)$ of NaI solution in the mixtures N,N-dimethylformamide-water at temp. 5°C. The DMF contents in the mixtures (in mol. %) are: 1 - 0%, 2 - 5%, 3 - 15%, 4 - 30%, 5 - 50%, 6 - 67%, 7 - 80%, 8 - 95%, 9 - 100%

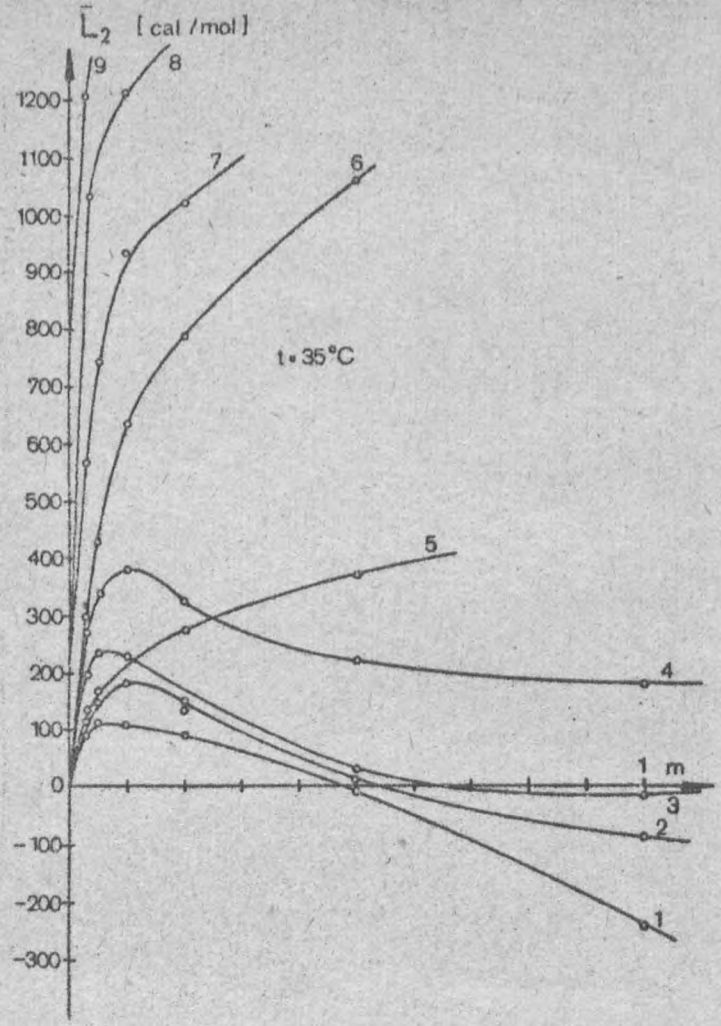


Fig. 4. The course of relation $\bar{L}_2 = f(m)$ of NaI solution in the mixtures N,N-dimethylformamide-water at temp. 35°C. The DMF contents in the mixtures (in mol. %) are: 1 - 0%, 2 - 5%, 3 - 15%, 4 - 30%, 5 - 50%, 6 - 67%, 7 - 80%, 8 - 95%, 9 - 100%

The analysis of the course of the function $\bar{L}_1/T = f(m)$ shows that within the range of small NaI concentrations, in all examined mixtures of DMF with water the above function takes small negative values. The further growth of electrolyte concentration brings about the changes of the course of the

Table 5

The $\frac{\bar{L}_1}{T} \cdot 10^3$ function values of NaI solutions in cal mol.$^{-1}$ in N,N-dimethylformamide-water mixtures (t = 5°C)

| Mol. % of DMF | Solution concentrations in mol. NaI/kg of solvent | | | | | |
|---|---|---|---|---|---|---|
| | 0.03 | 0.05 | 0.10 | 0.20 | 0.50 | 1.00 |
| 0 | -0.04 | -0.08 | -0.19 | +0.43 | +5.50 | +28.79 |
| 5 | -0.08 | -0.18 | -0.33 | +0.49 | +6.80 | +15.32 |
| 15 | -0.10 | -0.23 | -0.55 | -0.63 | +2.08 | +7.02 |
| 30 | -0.41 | -0.76 | -1.01 | +0.86 | +10.19 | +20.37 |
| 50 | -0.37 | -0.73 | -1.51 | -3.03 | -8.74 | -- |
| 67 | -0.63 | -1.24 | -2.84 | -7.37 | -26.52 | -- |
| 80 | -0.94 | -1.97 | -4.32 | -7.13 | -- | -- |
| 95 | -1.98 | -2.88 | -5.32 | -13.13 | -- | -- |
| 100 | -2.45 | -3.94 | -6.53 | -24.05 | -- | -- |

Table 6

The $\frac{\bar{L}_1}{T} \cdot 10^3$ function values of NaI solutions in cal mol.$^{-1}$ in N,N-dimethylformamide-water mixtures (t = 15°C)

| Mol. % of DMF | Solution concentrations in mole NaI/kg of solvent | | | | | |
|---|---|---|---|---|---|---|
| | 0.03 | 0.05 | 0.10 | 0.20 | 0.50 | 1.00 |
| 0 | -0.04 | -0.08 | -0.11 | +0.28 | +3.09 | +20.61 |
| 5 | -0.10 | -0.20 | -0.29 | +0.37 | +4.32 | +9.16 |
| 15 | -0.18 | -0.35 | -0.59 | -0.08 | +3.52 | +7.85 |
| 30 | -0.41 | -0.82 | -1.37 | -0.18 | +8.61 | +19.47 |
| 50 | -0.34 | -0.71 | -1.51 | -2.41 | -4.13 | -- |
| 67 | -0.83 | -1.56 | -3.07 | -6.63 | -23.81 | -- |
| 80 | -1.12 | -2.07 | -3.65 | -5.88 | -- | -- |
| 95 | -2.38 | -3.81 | -4.53 | -7.64 | -- | -- |
| 100 | -2.51 | -3.88 | -6.79 | -31.98 | -- | -- |

Table 7

The $\frac{\bar{L}_1}{T} \cdot 10^3$ function values of NaI solutions in cal mol.$^{-1}$ in N,N-dimethylformamide-water mixtures (t = 25°C)

| Mol. % of DMF | Solution concentrations in mol. NaI/kg of solvent | | | | | |
|---|---|---|---|---|---|---|
| | 0.03 | 0.05 | 0.10 | 0.20 | 0.50 | 1.00 |
| 0 | -0.04 | -0.12 | -0.08 | +0.16 | +2.71 | +15.09 |
| 5 | -0.10 | -0.19 | -0.26 | +0.27 | +3.23 | +6.74 |
| 15 | -0.18 | -0.30 | -0.22 | +0.60 | +2.20 | +2.46 |
| 30 | -0.39 | -0.72 | -1.09 | -0.44 | +2.50 | +4.70 |
| 50 | -0.29 | -0.55 | -0.94 | -1.69 | -8.55 | -- |
| 67 | -0.90 | -1.80 | -3.62 | -6.09 | -16.88 | -- |
| 80 | -1.31 | -2.36 | -3.73 | -6.14 | -13.97 | -- |
| 95 | -2.74 | -4.06 | -5.55 | -8.98 | -- | -- |
| 100 | -3.18 | -5.19 | -6.39 | -9.16 | -- | -- |

Table 8

The $\frac{\bar{L}_1}{T} \cdot 10^3$ function values of NaI solutions in cal mol.$^{-1}$ in N,N-dimethylformamide-water mixtures (t = 35°C)

| Mol. % of DMF | Solution concentrations in mol. NaI/kg of solvent | | | | | |
|---|---|---|---|---|---|---|
| | 0.03 | 0.05 | 0.10 | 0.20 | 0.50 | 1.00 |
| 0 | -0.04 | -0.07 | -0.11 | -0.15 | +1.69 | +12.07 |
| 5 | -0.10 | -0.20 | -0.34 | +0.03 | +3.19 | +7.61 |
| 15 | -0.20 | -0.32 | -0.22 | +0.88 | +3.83 | +5.72 |
| 30 | -0.37 | -0.67 | -0.92 | +0.00 | +3.68 | +6.60 |
| 50 | -0.25 | -0.52 | -1.09 | -2.17 | -7.16 | -- |
| 67 | -0.75 | -1.71 | -4.35 | -4.70 | -9.23 | -- |
| 80 | -1.44 | -2.84 | -3.56 | -6.12 | -12.50 | -- |
| 95 | -2.67 | -4.07 | -4.10 | -7.91 | -- | -- |
| 100 | -3.37 | -5.73 | -8.35 | -14.54 | -- | -- |

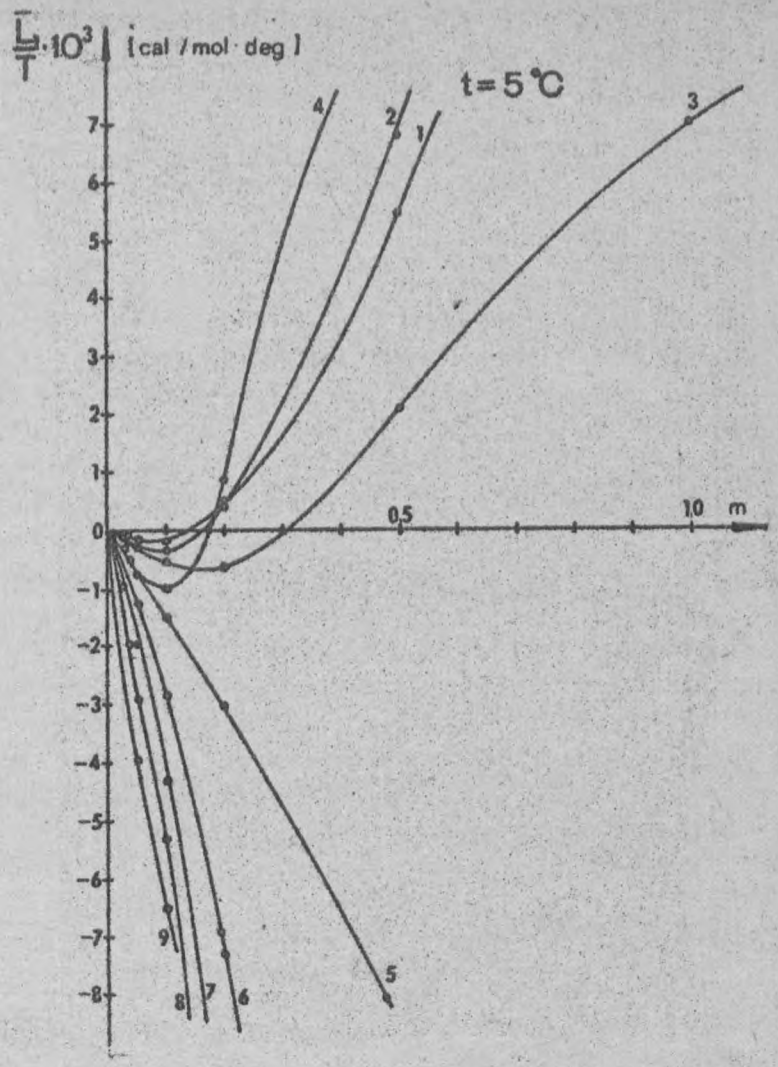


Fig. 5. The course of relation $\bar{L}_1/T = f(m)$ of NaI solution in the mixtures N,N-dimethylformamide-water at temp. 5°C. The DMF contents in the mixtures (in mol. %) are: 1 - 0%, 2 - 5%, 3 - 15%, 4 - 30%, 5 - 50%, 6 - 67%, 7 - 80%, 8 - 95%, 9 - 100%

function $\bar{L}_1/T = f(m)$, relating to the solvent composition. In the mixtures containing 0-30 mol. % DMF the discussed function takes positive values. According to F r a n k and R o b i n s o n' s suggestion [5] we can suppose that NaI disorders the

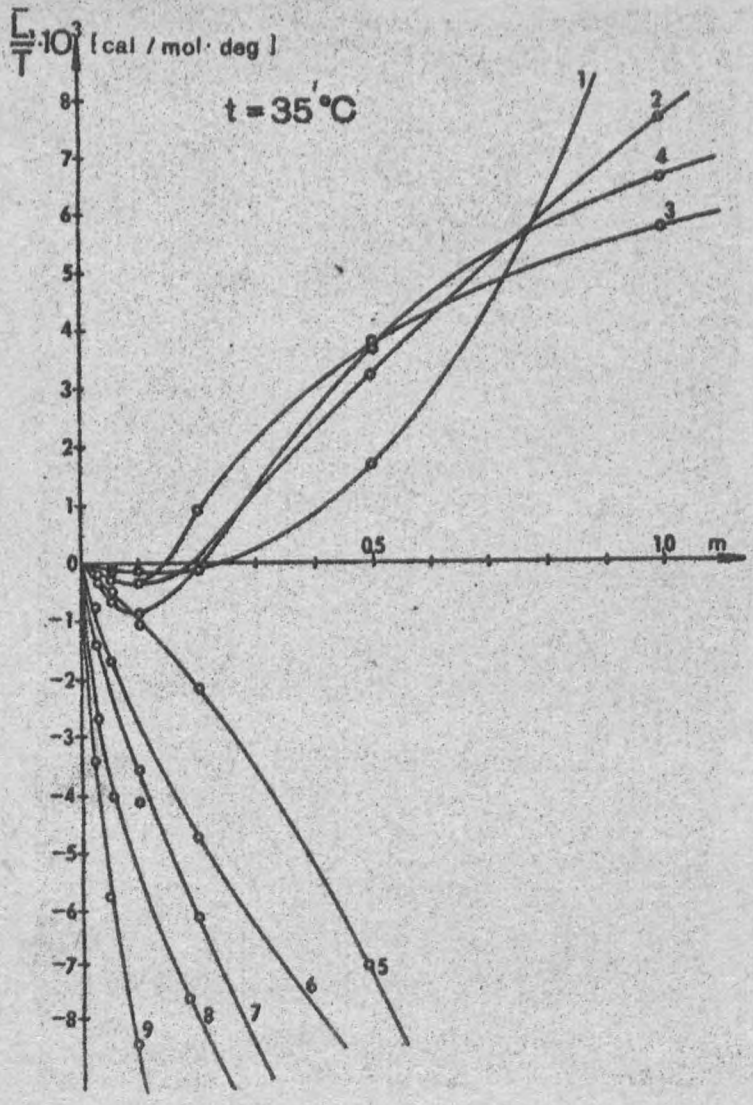


Fig. 6. The course of relation $\bar{L}_1/T = f(m)$ of NaI solution in the mixtures N,N-dimethylformamide-water at temp. 35°C. The DMF contents in the mixtures (in mol. %) are: 1 - 0%, 2 - 5%, 3 - 15%, 4 - 30%, 5 - 50%, 6 - 67%, 7 - 80%, 8 - 95%, 9 - 100%

solvent structure within the range of DMF contents smaller than 30 mol. %. In the solutions containing above 30 mol. % DMF the function $\bar{L}_1/T = f(m)$ takes negative values, which according to

the above suggestion, would point to ordering of solvent structure by the added electrolyte (NaI), within the remaining composition range of the discussed system.

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Institute of Chemistry
University of Łódź

*Alina Piekarska, Stefania Taniewska-Osińska*

WZGLĘDNA CZĄSTKOWA MOLOWA ENTALPIA ROZTWORÓW NaI W MIESZANINACH N,N-DWUMETYLOFORMAMIDU Z WODĄ

Na podstawie zmierzonych [1, 2] entalpii rozpuszczania NaI w mieszaninach N,N-dwumetyloformamidu z wodą w zakresie temperatur 5-35°C obliczono wartość względnej cząstkowej molowej entalpii rozpuszczalnika mieszanego ($\bar{L}_1$) i elektrolitu ($\bar{L}_2$), w zależności od stężenia elektrolitu we wszystkich badanych mieszaninach w temperaturze 5, 15, 25 i 35°C.

Z uzyskanych danych wynika, że kształt funkcji $\bar{L}_1 = f(m)$ i $\bar{L}_2 = f(m)$ zależy od składu mieszanego rozpuszczalnika oraz od temperatury. Ponadto obliczono wartości funkcji $\bar{L}_1/T$ i zanalizowano jej przebieg w zależności od stężenia elektrolitu i temperatury. Analiza przebiegu funkcji $\bar{L}_1/T$, której wartości są bliskie zmian entropii $\Delta\bar{S}_1^E$ wykazuje, że NaI zakłóca strukturę rozpuszczalnika w roztworach o zawartości DMF mniejszej niż 30% mol. W pozostałym zakresie składów omawianego układu wprowadzony elektrolit porządkuje strukturę rozpuszczalnika.

Алина Пекарска, Стефания Таневска-Осинска

## ОТНОСИТЕЛЬНАЯ ПАРЦИАЛЬНАЯ МОЛЯЛЬНАЯ ЭНТАЛЬПИЯ РАСТВОРОВ NaI В СМЕСЯХ N,N-ДИМЕТИЛФОРМАМИДА С ВОДОЙ

На основе полученых нами [1, 2] значений интегральной энтальпии растворения NaI в смесях N,N-диметилформамида с водой в диапазоне температур 5--35°C рассчитана относительная парциальная моляльная энтальпия смешанного растворителя ($\bar{L}_1$) и электролита ($\bar{L}_2$) в зависимости от концентрации электролита во всех изученных смесях при температуре 5, 15, 25, 35°C. На основании полученных данных видно, что характер функции $\bar{L}_1 = f(m)$ и $\bar{L}_2 = f(m)$ зависит от состава смешанного растворителя и от температуры.

Рассчитаны значения функции $\bar{L}_1/T$ и рассмотрен её ход в зависимости от концентрации соли и температуры. Анализ характера функции $\bar{L}/T$, которой значения близки изменениям энтропии $\Delta\bar{S}_1^E$ показывает, что NaI разупорядочивает структуру растворителя в растворах содержащих менее чем 30 мол. % ДМФ. В остальном интервале составов введенный электролит упорядочивает структуру растворителя.