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Vyacheslav F. Nikolaev^a; Guzel I. Ismagilova^a; Adel N. Satgaraev^b

^a A.E. Arbuzov Institute of Organic and Physical Chemistry of Russian Academy of Sciences, Kazan, Russia ^b Kazan State Technological University, Kazan, Russia

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Mixing enthalpies of molecular liquids: model of contribution balance of imaginary endo- and exotherms

Vyacheslav F. Nikolaev^{a*}, Guzel I. Ismagilova^a and Adel N. Satgaraev^b

^a*A.E. Arbutov Institute of Organic and Physical Chemistry of Russian Academy of Sciences, Kazan, Russia;* ^b*Kazan State Technological University, Kazan, Russia*

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To analyse enthalpy isotherms of mixing and other physicochemical properties of binary mixtures, there has been introduced a model of contribution balance of imaginary endo- and exotherms that can be used in solving a number of practical interpolation problems connected with predicting properties of binary mixtures of the composition (constant component 1–variable component 2 (homologous series)) in the absence of data for any member of homologous series, when prognosticating the temperature changes in isotherms of physicochemical properties of binary mixtures, as well as when predicting the temperatures of their stratification. As invariants of the model, there has been proposed a sum of extremums of imaginary endo- and exotherms, which is in linear connection with the integral of mixing enthalpy over the whole region of molar fractions, as well as the arithmetic mean of positions of endo- and exotherm extremums on the axis of molar fractions. For mixing enthalpies of binary systems (water–variable primary *n*-alcohol), linear correlations of extremum sums of endo- and exotherms with the molar volume of alcohol have been determined. Linear correlations of invariants with temperature have been ascertained. Deviations from this linear dependence point out on the occurrence of microheterogeneity in binary mixture and on its approach to stratification.

Keywords: empirical model; enthalpy; water; alcohol; temperature dependence

1. Introduction

Investigation of mixing enthalpies is one of the efficient methods of physicochemical analyses of liquid mixtures. On the basis of the dependences of mixing enthalpies on concentration, temperature, as well as the change of the second component, which is the member of some homologous series, one can draw conclusions about the character of change of intermolecular interactions [1]. As a basis for analysing isotherms, there can be used different empirical models. For formal description of experimental data in literature, there have been widely known the polynomial

*Corresponding author. Email: nikolaev_vf@iopc.knc.ru

Redlich–Kister model [2]:

$$Q^E = Q_{\text{exp}} - Q_{\text{calc}} = f_1 f_2 \sum_{i=0}^n A_i (f_1 - f_2)^i$$

$$= f_1 f_2 A_0 + f_1 f_2 A_1 (f_1 - f_2) + f_1 f_2 A_2 (f_1 - f_2)^2 + \dots + f_1 f_2 A_n (f_1 - f_2)^n, \quad (1)$$

where Q^E denotes excess physicochemical property of mixture, f_1, f_2 denote molar fractions of components of binary mixture ($f_1 + f_2 = 1$) and A_i denotes the model coefficients.

However, the coefficients of model (1) cannot be used for estimating the character of intermolecular interactions in solutions, since the required exponent of polynomial and, accordingly, the number of the used coefficients A_n is not constant but is defined by the necessary accuracy in describing experimental isotherm.

For performing such analysis, the polynomial model suggested in [3] appears to be more pertinent. Hwang et al. [3] paid attention to the presence of asymmetric contributions in isotherms of physicochemical properties of binary mixtures with extremums close to molar fractions of components ($f_1 = 0.2$ and 0.8). This model is characterised by a fixed number of coefficients and represents an isotherm of excess property of binary mixture in the form of a sum of three components:

$$Q^E = f_1 \times f_2 \times [B_0 + B_1 \times f_1^3 + B_2 \times f_2^3], \quad (2)$$

where B_i denotes constant coefficients, f_i denotes molar fractions of components ($f_1 + f_2 = 1$). The first contribution $f_1 \cdot f_2 \cdot B_0$ is symmetrical with regard to the equimolar point of composition, two other ones $B_1 \cdot f_1^4 \cdot f_2$ and $B_2 \cdot f_1 \cdot f_2^4$ are asymmetric and have extremums at $f_1 = 0.8$ and 0.2 , respectively.

An analogous approach has been used by us in working out non-stoichiometric model of competing van der Waals interactions for binary mixtures with universal intermolecular interactions [4]. The basic constituents of non-stoichiometric model are represented by volume-dispersive contribution, which is symmetrical with regard to the equimolar point of composition, as well as by two asymmetric exponential contributions, displayed as bursts in the excess regions of each of the mixture components, conditioned by the processes of structuring and mutual destructuring of components:

$$Q^E = A_{12} f_2 \exp(B_{12} f_2) + C_{12} f_1 f_2 + A_{21} f_1 \exp(B_{21} f_1), \quad (3)$$

where Q^E denotes excess thermodynamic function or some excess molar physicochemical property of binary system, f_1, f_2 denote molar fractions of components 1 and 2 ($f_1 + f_2 = 1$), $A_{12} f_2 \exp(B_{12} f_2)$, $A_{21} f_1 \exp(B_{21} f_1)$ denote structurally exponential contributions, having extremums, equal to $-A_{12}/(B_{12} \cdot e)$ in the point $f_2 = -1/B_{12}$ and $-A_{21}/(B_{21} \cdot e)$ in the point $f_1 = -1/B_{21}$, $C_{12} \cdot f_1 \cdot f_2$ —volumetrically dispersing contribution.

In a number of cases, there can be used asymmetric variant of recording volume-dispersive contribution $f_1 \cdot f_2 / (C_1 f_1 + C_2 f_2)$ that comes from Hildebrand–Scatchard relation for mixing enthalpies of binary mixtures with non-polar components [5]. The coefficients of model (3) correlate with polarity, polarisability and molar volume of components. The distinction of model (3) from Hwang model (2) lies in the fact that the given model does not establish the exact position of

extremums of asymmetric contributions. Analysis of experimental data on mixing enthalpies of systems (cyclohexane (component 1)–variable component 2) with the use of model (3) has shown that the greater the polarity of the component 2, the greater is the shift of the extremum $-A_{12}/(B_{12} \cdot e)$ to the region of lesser molar fractions of component 2. The greater the difference in dipole moments of components, the greater is the disturbing effect of components on each other and the greater is the values of extremums of exponential contributions.

Isotherms of physicochemical properties of binary mixtures with associated components have their peculiarities and represent a complex balance of processes of intra-component associations (interactions of the types 1–1 and 2–2) and inter-component complex formation (interactions of the types 1–2). Equilibrium in the solution is characterised with a multiple set of step equilibrium constants, reflecting the process of formation of chained, treelike, cyclic and combined structures [6]. In the given work (Section 3) for describing molar mixing enthalpies and isotherms of other physicochemical properties of binary mixtures with associated components, a model of contribution balance of imaginary endo- and exotherms has been proposed. For the systems constant component 1–variable component 2 (members of homologous series) and for the systems constant component 1–constant component 2 at varying temperatures, linear correlations of sums of extremums of endo- and exotherms with molar volume of variable component and with temperature have been determined accordingly. Correlations of the sum of extremums with integrals of excess properties over the whole region of molar fractions have also been established. The values of this integral and the sum of extremums can be used as a generalised quantitative criterion of intermolecular interactions in binary mixtures while carrying out the statistical analysis of experimental bulk of data on mixing enthalpies.

2. Database

As initial data for checking up the operability of balance model of contributions of imaginary endo- and exotherms, we have used data on mixing enthalpies of water with primary normal alcohols, as well as hexane with methanol, which are given in [1,7].

3. Model of contribution balance of imaginary endo- and exotherms

Model of contribution balance of imaginary endo- and exotherms uses simplified record of stoichiometric approach to the description of the properties of mixtures with the associated components and allows on the empirical level to analyse and to model the most complex processes of self-association of components and complex formation.

As it is known, isotherm of any physicochemical property of mixture is the result of energy balance of contributions of different types of intermolecular attraction and repulsion [8]. Although the model for describing isotherms on the basis of balance of contributions of imaginary endo- and exotherms is a simulation one, the analysis of experimental data with the use of the given model has shown that the model coefficients and their derivatives (extremum values of endo- and exotherms, their sum, positions of extremums of endo- and exotherms on the concentration axis of

molar fractions, other distinguishing points on endo- and exotherms) can be efficiently used in the analysis and prediction of temperature behaviour of isotherms of physicochemical properties of binary mixtures, including prediction of regions of microheterogeneity occurrence in the solution bulk, becoming apparent in model invariants much earlier than the visually registered stratification of phases.

The model can be used in the analysis both as mixing enthalpies and as other physicochemical properties of mixtures of the composition (constant component 1–variable component 2), where the members of some homologous series are acting as component 2.

Excess of molar physicochemical property of mixture is presented in the model as a sum of two contributions, the signs of which have been set opposite during optimisation:

$$Q^E = C_{\text{exo}}f^n(1-f)^m + C_{\text{endo}}f^{n_1}(1-f)^{m_1}, \quad (4)$$

where f denotes molar fraction of one of the components of binary mixture, C_{exo} , C_{endo} , n , m , n_1 , m_1 denote the model coefficients, defined by the statistical processing of couples of data f_i and Q_i^E . In the case of analysis of mixing enthalpies, the negative constituent can be called imaginary exotherm and the positive one, imaginary endotherm. Mathematical form of imaginary endo- and exotherms is similar to the record of relation for calculating the concentration of the forming complex via equilibrium constant and concentration of components, forming the complex with stoichiometric coefficients n and m (for exotherm) and n_1 and m_1 (for endotherm). Since the sum of molar fractions in binary mixture equals unity, each member of the model simulates the conditions of the experiment in defining the complex composition by the method of isomolar series [9], with the only difference that molar concentrations of components are substituted for their molar fractions, and complex formation proceeds without the background of the (inert) solvent.

Position and value of the minimum of exotherm $C_{\text{exo}}f^n(1-f)^m$ ($C_{\text{exo}} < 0$) are equal to $f_{\text{exo}} = n/(n+m)$ and $Q_{\text{exo}} = C_{\text{exo}}[n/(n+m)]^n[m/(n+m)]^m$, respectively, and position and value of maximum of endotherms $C_{\text{endo}}f^{n_1}(1-f)^{m_1}$ ($C_{\text{endo}} > 0$) are equal to $f_{\text{endo}} = n_1/n_1+m_1$ and $Q_{\text{endo}} = C_{\text{endo}}[n_1/(n_1+m_1)]^{n_1}[m_1/(n_1+m_1)]^{m_1}$. For determining the correlation of the type of isotherms with the properties of components, it is more preferable not to use the model coefficients, but apply their invariants: sum of extremums of endo- and exotherms $Q_{\text{sum}} = Q_{\text{endo}} + Q_{\text{exo}}$ and the arithmetic mean of their positions on the axis of molar fractions of the diagram (composition property) $f_{\text{mid}} = (f_{\text{endo}} + f_{\text{exo}})/2$. These invariants are more resistant to interruption of iteration procedure in determining the model coefficients with the help of different statistical programs, as well as in analysing the experimental data for identical binary mixtures, obtained by different authors. The proposed model (even with all diversity of experimental isotherms: parabolic, asymmetric, S-shaped) allows to reduce their analysis to the analysis of the change of model invariants Q_{sum} and f_{mid} , as well as the integral (Int) of mixing enthalpy ΔH or other molar excess physicochemical property of mixture Q^E over the whole region of molar fractions:

$$\text{Int} = \int_0^1 Q^E(f)df = \int_0^1 (C_{\text{exo}}f^n(1-f)^m + C_{\text{endo}}f^{n_1}(1-f)^{m_1})df \quad (5)$$

from variable factors (molar volume or number of carbon atoms in variable component 2, variable temperature, etc.). As a rule, the model invariants Q_{sum} , f_{mid} and Int , if binary mixture is homogeneous and stays far from the region of stratification, display linear connection with the listed above variable factors. Additional criteria of nearness of binary mixture to the region of microheterogeneity and stratification is the substantial remoteness of f_{mid} from the equimolar point $f=0.5$.

The given model is not exacting to the choice of the first approximation. Thus, for C_{exo} and C_{endo} as the first approximation, there are chosen numbers equal in absolute values, which exceed the maximum value of the analysed excess property Q^E (or ΔH), but opposite in sign, that is $C_{\text{endo}} = -C_{\text{exo}}$. For exponents n , m , n_1 and m_1 , the first approximation is taken as a unit.

4. Results and discussion

4.1. The analysis of isotherms of mixing enthalpies of binary mixtures with associated components (component 2-homologous series)

As it has been noted above, the model coefficients are extremely sensitive to the quality of experimental data and to interruption of iteration procedure in statistical data processing. In Table 1, there are given the coefficients of model (4) for mixing enthalpies of binary mixtures (water-primary n -alcohol) on the basis of statistical processing of experimental enthalpies, obtained by different authors [1,7]. It can be seen that with the rather good quality of describing experimental data, the coefficients of model for the same mixtures essentially differ, differences in coefficients C_{endo} and C_{exo} being especially great. At the same time, the model invariants Q_{sum} and f_{mid} (Table 2) turn out to be rather close. This circumstance allows to use Q_{sum} and f_{mid} as a stable characteristic of isotherms in analysing experimental data.

Figure 1 shows experimental isotherms of mixing enthalpies ΔH^{298} of mixtures water-primary n -alcohol and isotherms, plotted with the use of model (4) coefficients, given in Table 1. Between the model invariants Q_{sum} and f_{mid} , there also exists an interconnection close to linear dependence.

It is seen from Figure 2 that with the growth of size of alkyl radical in variable n -alcohol, the midposition of the extremums of endo- and exotherms f_{mid} is increasingly shifted from the point of equimolarity $f=0.5$. Deviation of the 1-propanol point from the line, on which there are points for methanol and ethanol, is apparently connected with the system approaching to the region of microheterogeneity and stratification [10]. Indeed, the next member of homologous series of n -alcohols- n -butanol in equimolar mixture with water at 298.15 K already does not form a homogeneous solution.

Thus, with model (4) for describing isotherms of mixture enthalpies of constant component 1-variable component 2, if as a component 2, the members of some homologous series are used, and it is possible to determine the correlations of model invariants with the molar volume of variable component (Figure 3) or with the number of carbon atoms in alkyl chain. The latter is allowing us to predict the appearance of isotherms for the member of homologous series, the experimental data for which are absent. Besides, it is possible to define the homologue, starting from

Table 1. Coefficients of model (4) for isotherms of mixing enthalpies ΔH (J mol^{-1}) of systems water–primary n -alcohol at 298.15 K.

	Alcohol (component 2)	C_{exo} (J mol^{-1})	n	m	C_{endo} (J mol^{-1})	n_1	m_1	r	s
1	Methanol	-2,510,261.0	1.288545	1.483929	2512537.0	1.291099	1.485833	99.99	5.02
2	Methanol	-894 196.1	1.315262	1.544215	897135.2	1.323056	1.550267	99.99	4.00
3	Methanol	-3,740,339.0	1.335854	1.561577	3,743,54.0	1.337795	1.563148	99.98	7.08
4	Ethanol	-8,101,021.0	1.263168	1.796510	8109618.0	1.264363	1.797403	99.82	15.19
5	Ethanol	-7,844,070.0	1.177660	1.672257	7850106.0	1.178628	1.672930	99.66	22.40
6	Ethanol	-2,812,348.0	1.247861	1.780535	2819971.0	1.251071	1.782924	99.82	14.82
7	Ethanol	-8,101,061.0	1.231388	1.847789	8,109,761.0	1.232575	1.848729	99.93	10.40
8	1-Propanol	-3,277,206.0	0.847183	1.586702	3,279,507.0	0.847863	1.586697	99.37	22.53

Table 2. Extremums of imaginary endo- and exotherms and model invariants Q_{sum} and f_{mid} .

Alcohol (component 2)	Q_{exo} (J mol ⁻¹)	f_{exo}	Q_{endo} (J mol ⁻¹)	f_{endo}	Q_{sum} (J mol ⁻¹)	f_{mid}	Int	V (cm ³)
1 Methanol	-369 924.8	0.46476	369 096.9	0.46494	-827.9	0.46485	-609.9	40.44
2 Methanol	-124 345.3	0.45997	123 539.8	0.46046	-805.5	0.46022	-591.7	40.44
3 Methanol	-506 430.3	0.46105	505 611.4	0.46116	-818.9	0.46111	-601.3	40.44
4 Ethanol	-1,018,066.8	0.41284	1,017,586.4	0.41295	-480.4	0.41290	-393.7	58.41
5 Ethanol	-361 312.2	0.41323	360 804.0	0.41333	-508.2	0.41328	-405.7	58.41
6 Ethanol	-1,135,964.5	0.41205	1,135,459.1	0.41235	-505.4	0.41220	-406.4	58.41
7 Ethanol	-1,019,984.8	0.39991	1,019,480.5	0.40002	-504.3	0.39997	-404.6	58.41
8 1-Propanol	-679 835.4	0.34808	679 826.2	0.34826	-9.2	0.34817	-4.5	74.80

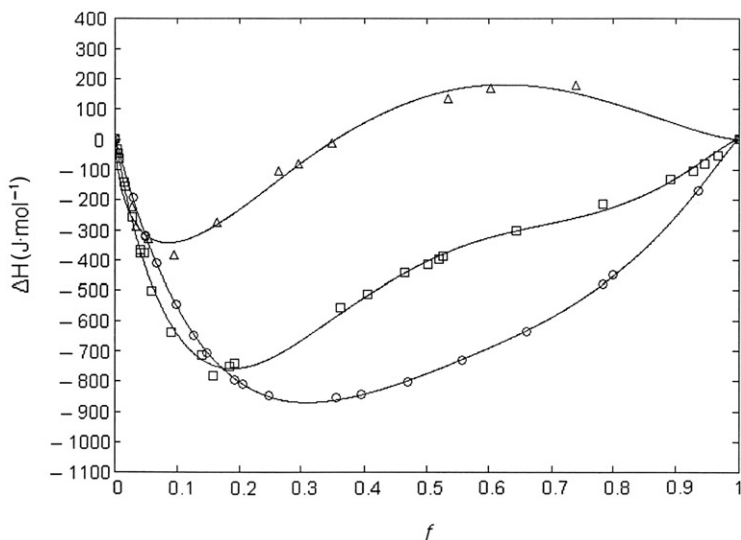


Figure 1. Experimental enthalpy isotherms of binary mixtures water-variable primary n -alcohol (points) and isotherms, approximated by model (4), from the molar fraction of alcohol. Symbols: (○) water-methanol²; (□) water-ethanol⁶; (Δ) water-1-propanol⁸; (—) model of contribution balance of imaginary endo- and exotherms.

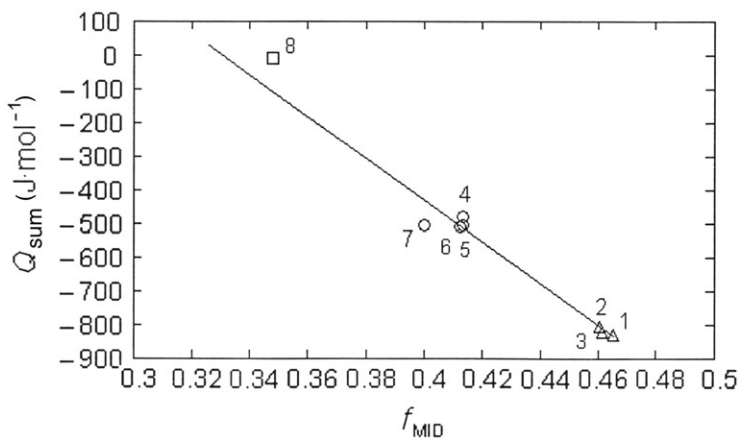


Figure 2. Correlation of invariants Q_{sum} and f_{mid} of model (4) for mixing enthalpies ΔH^{298} of mixtures water-primary n -alcohol. Symbols: (Δ) water-methanol; (○) water-ethanol; (□) water-1-propanol. Values Q_{sum} and f_{mid} have been calculated by using the experimental data of different authors [1,7]. Numerical designations of the systems are given in Table 1.

which there begins stratification of binary mixture. The greater the asymmetry of components of binary mixture, the greater will be the shift of positions of extremums of exotherm f_{exo} and endotherm f_{endo} from the equimolar point of composition $f_{\text{water}} = f_{\text{alcohol}} = 0.5$.

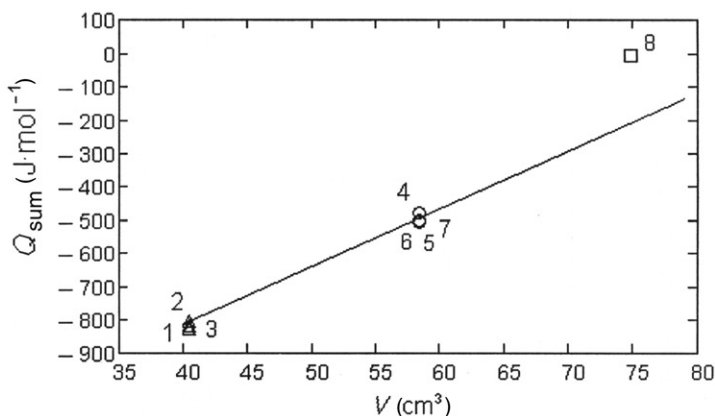


Figure 3. Correlation of Q_{sum} with molar volume, V , of variable primary n -alcohol. Symbols: (Δ) water–methanol; (\circ) water–ethanol; (\square) water–1-propanol. Numerical values of points are given in Table 1.

It is remarkable that there exists an almost linear dependence between Q_{sum} and Int (Figure 4).

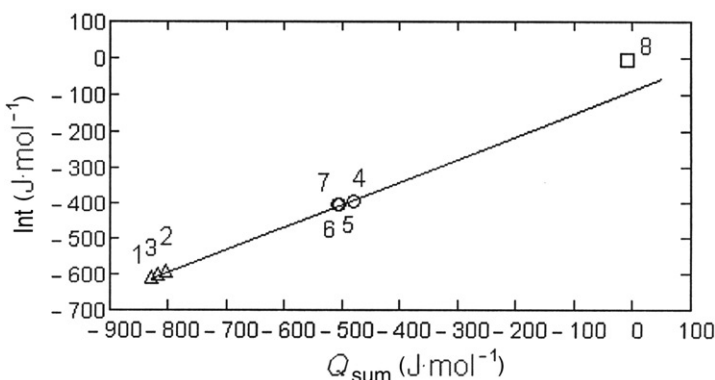


Figure 4. Correlation of invariant Q_{sum} of model (4) and Int of isotherms of mixing enthalpies ΔH^{298} of mixtures water–primary n -alcohol. Symbols: (Δ) water–methanol; (\circ) water–ethanol; (\square) water–1-propanol. Numerical values of points are given in Table 1.

4.2. Analysis of temperature dependences of mixing enthalpies

Model (4) also allows to analyse the influence of temperature on the isotherms of mixing enthalpies (constant component 1–constant component 2). For this case, there have been obtained dependences similar to those given in Section 4.1, with the only difference that temperatures at which mixing enthalpies were determined were used instead of molar volume of variable component 2.

4.3. Application of model (4) in defining the region of microheterogeneity and stratification of binary mixtures

In Section 4.2, it has been shown that if determination of enthalpy of binary system is carried out at the temperature far from the temperature of its stratification, then the

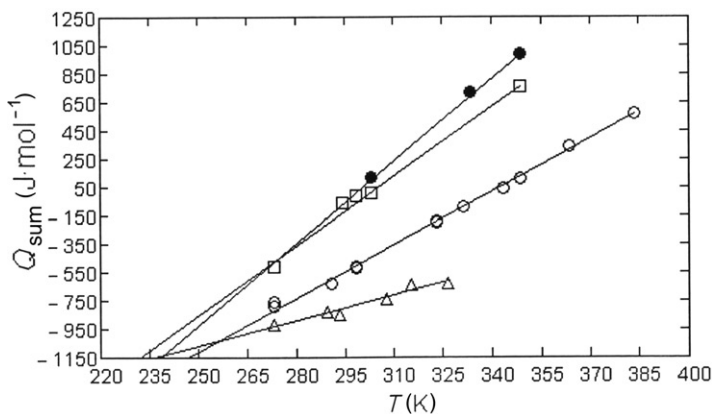


Figure 5. Dependence of invariant Q_{sum} for mixing enthalpies of mixtures water–primary n -alcohol on temperature. Symbols: (Δ) – water–methanol; (\circ) – water–ethanol; (\square) – water–1-propanol, (\bullet) – water–1-butanol. (The experimental data are taken from [1,7]).

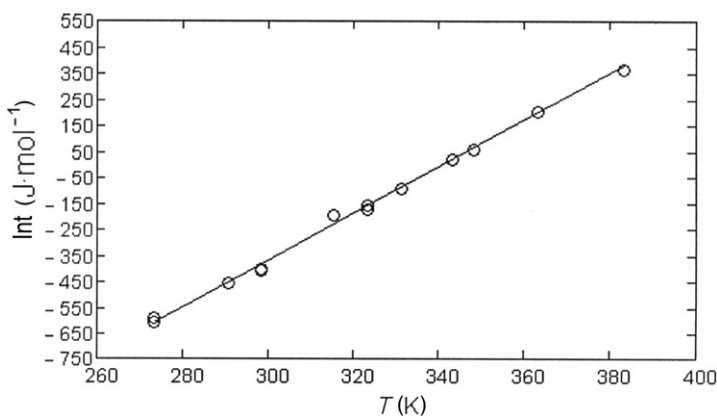


Figure 6. Dependence of Int of isotherms of mixing enthalpies of system water–ethanol on temperature. (The experimental data are taken from [1,7]).

invariant Q_{sum} , as well as the Int , are linearly connected with temperature (Figures 5 and 6). Example of the system with stratification is methanol–hexane. The coefficients of model (4) for this system are given in Table 3. In Table 4 for the same system, the characteristics of extremums of imaginary endo- and exotherms and calculated the model invariants: Q_{sum} , f_{mid} and Int are given.

Figure 7 shows the changes of the invariant f_{mid} with the temperature. In the region of about 306.85 K, the sharp rupture of dependence is observed. More smooth rupture is also observed on the dependence of invariant Q_{sum} on temperature (Figure 8).

This points out that at such temperature the binary system is transformed from the region of homogeneity to the region of microheterogeneity and stratification.

Table 3. Coefficients of model (4) for mixing enthalpies of methanol-hexane at different temperatures.

Temperature (K)	C_{exo} (J mol^{-1})	n	m	C_{endo} (J mol^{-1})	n_1	m_1	r	s
298.15	-14,651,760	1.035821	0.568521	14,652,420	1.035732	0.568498	99.98	3.33
303.15	-13,942,610	1.036473	0.607225	13,943,310	1.036376	0.607191	99.98	3.53
306.85	-11,498,270	1.064983	0.733276	11,498,570	1.064831	0.733186	99.77	13.99
313.15	-11,498,050	1.081643	0.697509	11,498,620	1.081493	0.697424	99.98	4.68
318.15	-11,497,960	1.083250	0.683935	11,498,690	1.083098	0.683848	99.98	4.56
323.15	-11,497,910	1.085972	0.682223	11,498,750	1.085816	0.682129	99.99	4.25

Table 4. Characteristics of extremums of endo- and exotherms and invariants of model (4) for enthalpies of methanol–hexane.

Temperature (K)	Q_{exo} (J mol ⁻¹)	f_{exo}	Q_{endo} (J mol ⁻¹)	f_{endo}	Q_{sum} (J mol ⁻¹)	f_{mid}	Int
298.15	-5,163,257.0	0.64564	5,163,813.8	0.64563	556.8	0.64564	462.8
303.15	-4,722,454.4	0.63057	4,723,062.7	0.63057	608.3	0.63057	496.2
306.85	-3,409,302.4	0.59223	3,409,938.1	0.59223	635.7	0.59223	518.5
313.15	-3,492,998.4	0.60795	3,493,710.4	0.60795	712.0	0.60795	569.9
318.15	-3,535,154.9	0.61298	3,535,934.4	0.61298	779.5	0.61298	618.2
323.15	-3,536,194.0	0.61417	3,537,037.9	0.61417	843.9	0.61417	664.3

Note: f denotes molar fraction of hexane.

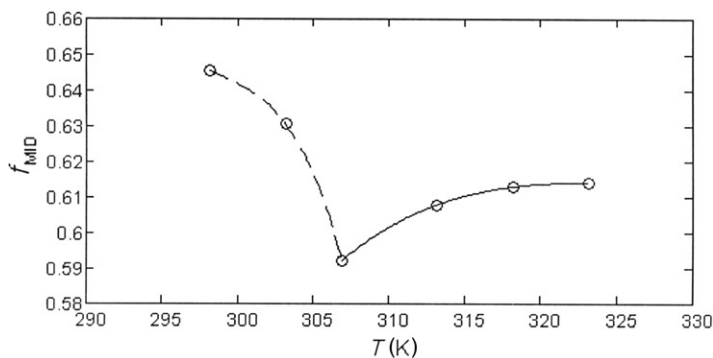


Figure 7. Dependence of the invariant f_{mid} on temperature for methanol-hexane mixture.

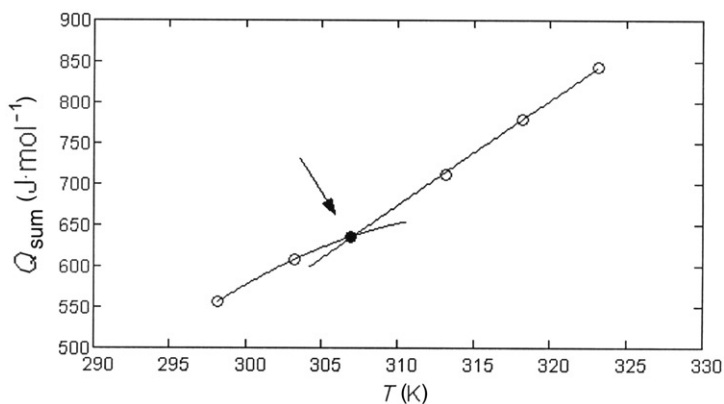


Figure 8. Temperature dependence of the invariant Q_{sum} of model (4) for mixing enthalpies of system methanol-hexane at the transition of the system to the region of stratification (306.85 K).

5. Conclusions

The invariants of the proposed model of balance of contributions of imaginary endo- and exotherms Q_{sum} , f_{mid} together with the value of the Int excess physicochemical property of mixture over the whole region of composition allow to define the simple linear interrelations of characteristics of isotherms with the parameters, varying in the experiment (variation of component 2, variations of temperature). The deviations of experimental points from the obtained linear dependences point out at the approximation of the system to the region of microheterogeneous and stratification.

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