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Қазақстан Республикасы Ұлттық ғылым академиясы "ҚР ҰҒА Хабарлары. Химия және технология сериясы" ғылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Web of Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Химия және технология сериясы Emerging Sources Citation Index-ке енуі біздің қоғамдастық үшін ең өзекті және беделді химиялық ғылымдар бойынша контентке адалдығымызды білдіреді.

НАН РК сообщает, что научный журнал «Известия НАН РК. Серия химии и технологий» был принят для индексирования в Emerging Sources Citation Index, обновленной версии Web of Science. Содержание в этом индексировании находится в стадии рассмотрения компанией Clarivate Analytics для дальнейшего принятия журнала в the Science Citation Index Expanded, the Social Sciences Citation Index и the Arts & Humanities Citation Index. Web of Science предлагает качество и глубину контента для исследователей, авторов, издателей и учреждений. Включение Известия НАН РК в Emerging Sources Citation Index демонстрирует нашу приверженность к наиболее актуальному и влиятельному контенту по химическим наукам для нашего сообщества.

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THE RMODYNAMIC ASSESSMENT OF SMELTING OF MANGANESE AND CHROMIUM FERROALLOYS BASED ON THE ANALYSIS OF THEIR STATE DIAGRAMS

Abstract. This paper proposes a technique for extracting thermodynamic data from state diagrams, which are based on the modified Schroeder-Le Chatelier equation allowing to describe the lines of phase equilibria of binary systems by mathematical expressions. We showed the possibility to apply the equation of dependence of the Bjerrum-Guggenheim coefficient for the area of iron crystallization on the activity of iron of an ideal solution α - γ for binary systems Fe-Si, Fe-Cr, Fe-Mn, Fe-Al. The calculated partial excess entropy, enthalpy and the Gibbs energies of mixing of the liquidus components of the said systems are proposed.

Key words: complex alloys, ferrosilicoaluminum, modified Schroeder- Le Chatelier equation, Bjerrum-Guggenheim coefficient, partial excess enthalpy (ΔH^E), entropy (ΔS^E) and the Gibbs energy of mixing (G^M) of liquidus component.

When smelting steel and special alloys, the ferroalloys obtained by alloying ferrosilicium and aluminum, produced from very scarce materials such as quartzite, metal shavings, bauxite, etc., are used as a reducing agent and a deoxidizing agent. For the first time in the world practice, a process for obtaining complex silicon-aluminum alloys under the general name of ferrosilicoaluminum has been developed.

When using ferrosilicoaluminum as a metal reducing agent while smelting refined grades of Mn, Cr and Ti the question arises as to how exactly aluminum and silicon in a complex will behave themselves in these alloys, since an alien metal (aluminum - when smelting ferromanganese and a low-carbon ferrochrome, and silicium - when smelting ferrotitanium) is additionally introduced into the system. Whether strong heteropolar (ionic, covalent, metallic) bonds of aluminum in the melts of Mn-Si-Fe, Si-Cr-Fe systems, and those of silicium - in the melt of Al-Ti-Fe system will be formed. If in the corresponding systems in the field of compositions of traditional refined ferroalloys they will experience negative deviations from ideality and they are not inclined to form various complex associates, then there will not be any obstacle in using a novel ferrosilicoaluminum in metallurgy as a metal reducing agent.

The state diagrams clearly reflect the nature of the interaction of components in the melt, its phase composition and the structure of alloys under conditions of stable thermodynamic equilibrium. Consequently, they contain all the fundamental and important data on the thermodynamic properties of phases and components.

The analysis of the state diagrams and the developed technique of mathematical description of the phase equilibrium line in terms of the Bjerrum-Guggenheim concept (Φ) showed that since this coefficient

characterizes a degree of deviation of the energy state of a given component under equilibrium conditions from ideality, then using it, it is possible to determine a magnitude and a sign of the exchange energy between the components, and this deviation being the greater, the stronger the interparticle interaction between the components is ($\Phi > 1$ is a negative deviation from ideality, $\Phi < 1$, is a positive deviation from ideality) [1-3].

The developed technique of mathematical description of the lines of phase equilibria allowed to solve the direct Gibbs problem correctly, namely, to obtain an analytical dependence of the phase compositions on the temperature at phase equilibria based on the laws of phase formation. It is shown that by means of the osmotic Bjerrum-Guggenheim coefficient (Φ_i), a real equilibrium in the system under study can be determined in detail and its correct analytical expression be obtained [4-6].

The solution of the inverse Gibbs problem is in extracting the thermodynamic information contained in the state diagram provided that the mathematical problem of phase equilibria is solved correctly [7].

We have solved the first stage of the Gibbs inverse problem, i.e., we have obtained the modified Schroeder-Le Chatelier equation, allowing to describe analytically the liquidus and solidus lines. In this connection, let us consider the way of deriving analytical expressions for the partial excess enthalpy ($\Delta H^{M(E)}$) and entropy (ΔS^E) of mixing the liquidus component in the systems with the ratio of $\Phi_i = A_i + B_i \cdot a_i$.

The difference between the thermodynamic mixing function G^M for a real solution and the value of this function for an ideal solution G_{id}^M at the same T and p is called an excess thermodynamic function.

Therefore,

$$\Delta G_i^E = RT \ln j_i. \quad (1)$$

The relationship between the activity coefficient and the Bjerrum-Guggenheim coefficient is established as follows:

$$\Phi_i = \frac{\Delta G_{m,i}^{S \rightarrow L}}{\Delta G_{m,i}^{id}} = \frac{\Delta \mu_i^L - \Delta \mu_i^S}{\Delta \mu_{i,id}^L - \Delta \mu_{i,id}^S} = \frac{\ln(a_i^L/a_i^S)}{\ln(x_i^L/x_i^S)} = 1 + \frac{\ln \gamma_i^L / \ln \gamma_i^S}{\ln x_i^L / \ln x_i^S}, \quad (2)$$

where $\Delta G_{m,i}^{S \rightarrow L}$ is the Gibbs energy of melting of the component relates to its ideal component $\Delta G_{m,i,id}^{S \rightarrow L} = RT \ln X_i^L - RT \ln X_i^S$.

Let us express the coefficient of activity in terms of the Bjerrum-Guggenheim osmotic coefficient. In the physicochemical literature, this coefficient is called an osmotic coefficient [8] and it establishes a relationship between the activity and concentration of the component in the solution, which substantially differs from the analogous relationship between the activity coefficient and concentration:

$$\ln a_i = \Phi_i \ln x_i.$$

Since we are considering the variation of the quantity F_i along the line of phase equilibria, hence, it depends on the temperature and on the composition of equilibrium phases. At the same time, as can be seen from expression (2), this quantity is a dimensionless quantity. Its identity with a well-known osmosis is shown below.

The thermodynamic classification of solutions is based on the nature of the equations for the chemical potentials of the solution components. In an ideal solution, the following expression is true for each component:

$$\mu_i^{id}(T, P, X) = \mu_i^0(T, P) + RT \ln X_i, \quad (3)$$

where μ_i^0 - is a standard chemical potential of an i-th substance in a standard state; X_i - is the molar concentration of the i-component in the solution; member $RT\ln X_i$ - corresponds to a change of the chemical potential due to the mixing (when forming an ideal solution).

By analogy with equation (3), the dependence of chemical potentials of a nonideal solution on the concentration can be written [7] in the form of:

$$\mu_i(T, P, X) = \mu_i^0(T, P) + RT \ln \gamma_i X_i, \quad (4)$$

where γ_i is an activity coefficient.

The general condition of ideality is that for an ideal solution throughout the entire concentration range the following condition is satisfied:

$$\gamma_i(T, P, X_1, \dots, X_{k-1}) = 1 \quad (i = 1, 2, \dots, k).$$

An advantage of using the coefficient of activity is that it allows to preserve a formal similarity of the expressions for the thermodynamic properties of real solutions with the equations for nonideal solutions. The activity coefficient of a solvent in contrast to the activity coefficients of the dissolved components from purely arithmetic considerations is not the most suitable function for measuring deviations from ideality. Therefore, instead of the activity coefficient for the solvent, it is more convenient to use another correction factor called the osmotic coefficient of Bjerrum-Guggenheim [9, 10], which is introduced in the following way:

$$\mu_1(T, P, X) = \mu_1^0(T, P) + \Phi_1 \cdot RT \cdot \ln X_1, \quad (5)$$

where Φ_1 is the osmotic coefficient of Bjerrum and Guggenheim of a solvent $\Phi \rightarrow 1$ at $X_1 \rightarrow 1$ and $X_{2,3} \dots \rightarrow 0$.

Comparing the expression $\ln a_i = \Phi_1 \ln x_i$ with equation (5), we will find that

$$\Phi - 1 = \frac{\ln \gamma_i}{\ln X_i} \quad \text{or} \quad \Phi = \frac{\ln a_i}{\ln X_i}. \quad (6)$$

Therefore, the activity coefficient can be expressed as follows:

$$\ln \gamma_i = (\Phi - 1) \ln x_i.$$

To calculate the partial excess thermodynamic mixing functions of the liquidus component we have the initial data:

$$\Phi_i = A_i + B_i \cdot a_i; \quad (7)$$

$$a_i = \exp\left[\frac{\Delta H_{m,i}}{R} \left(\frac{1}{T_{m,1}} - \frac{1}{T}\right)\right]; \quad (8)$$

$$\ln x_i = \frac{\Delta H_{m,i}}{R \cdot \Phi_i} \cdot \left(\frac{1}{T_{m,1}} - \frac{1}{T}\right)_0; \quad (9)$$

$$\left(\frac{d\Delta G_i^E}{dT}\right)_{x,p} = -\Delta S_i^E; \quad \Delta G_i^E = \Delta H_i^{E(M)} - T\Delta S_i^E. \quad (10)$$

When differentiating function ΔG_i^E with respect to T at x = const

$$(\Delta G_i^E) = -\Delta S_i^E = [RT(\Phi_i - 1) \ln x] = R \cdot \ln x(\Phi_i - 1 + T \cdot \Phi'_i) = R \ln x(\Phi_i - 1 + T \cdot B_i \cdot a_i \cdot \frac{\Delta H_{m,i}}{R} \cdot \frac{1}{T^2}); \quad (11)$$

$$\Delta S_i^E = -R \ln x(\Phi_i - 1 + B_i \cdot a_i \cdot \frac{\Delta H_{m,i}}{RT}); \quad (12)$$

$$\Delta H_i^{E(M)} = -B_i \cdot a_i \cdot \Delta H_{m,i} \cdot \ln x_i. \quad (13)$$

For Fe-Mn, Fe-Cr, Fe-Si, Fe-Al systems under consideration we assumed the following thermodynamic characteristics of iron: $\Delta H_{\text{melt, Fe}} = 15190.384$ J/mole, $T_{\text{melt, Fe}} = 1811$ K [11].

Calculation of excess thermodynamic mixing functions in Mn-based systems.

The initial data are taken from [12] and presented in Table 1.

Table 1 - Input parameters for calculating thermodynamic data in Fe-Mn system

No. in sequence	T, K	$X_{\text{op. Fe}}^L$	$\ln X_{\text{Fe}}^L$	Φ'_{Fe}	Φ''_{Fe}	a_{Fe}^L	1/T
1	1749	0.875	0	1.991	-0.136	1	5.534
2	1723	0.821	5.1512E-05	2.502	-0.153	0.992	5.577
3	1698	0.755	8.2152E-05	2.869	-0.165	0.986	5.608
4	1673	0.686	0.00010884	3.239	-0.178	0.98	5.64
5	1648	0.617	0.00013234	3.611	-0.190	0.975	5.672
6	1623	0.542	0.00015321	3.984	-0.203	0.969	5.704
7	1598	0.469	0.0001719	4.36	-0.215	0.963	5.737
8	1573	0.401	0.00018877	4.738	-0.228	0.957	5.770
9	1548	0.325	0.00020408	5.118	-0.241	0.951	5.803
10	1523	0.232	0.00021807	5.5	-0.254	0.946	5.837

Equation Φ'' for γ -Fe: $\Phi''_{\text{Fe}} = -2.33 + 2.194 \cdot a_i$

The thermodynamic characteristics of iron in Fe-Mn melt calculated by equations (11-13) are presented in Table 2 and in figure 1.

Table 2 - Calculated thermodynamic data of γ -Fe in Fe-Mn melt

No. in sequence	T, K	ΔH kJ/g·at.	ΔS kJ/g·at·K	ΔG kJ/mole
1	1749	0	0	0
2	1723	-0.0017	467.954159	-0.07555
3	1698	-0.0027	797.632826	-0.13165
4	1673	-0.00356	1123.464	-0.18956
5	1648	-0.0043	1445.416054	-0.24934
6	1623	-0.00495	1763.457148	-0.31102
7	1598	-0.00552	2077.555229	-0.37464
8	1573	-0.00603	2387.678037	-0.44026
9	1548	-0.00647	2693.793106	-0.50791
10	1523	-0.00688	2995.867768	-0.57764

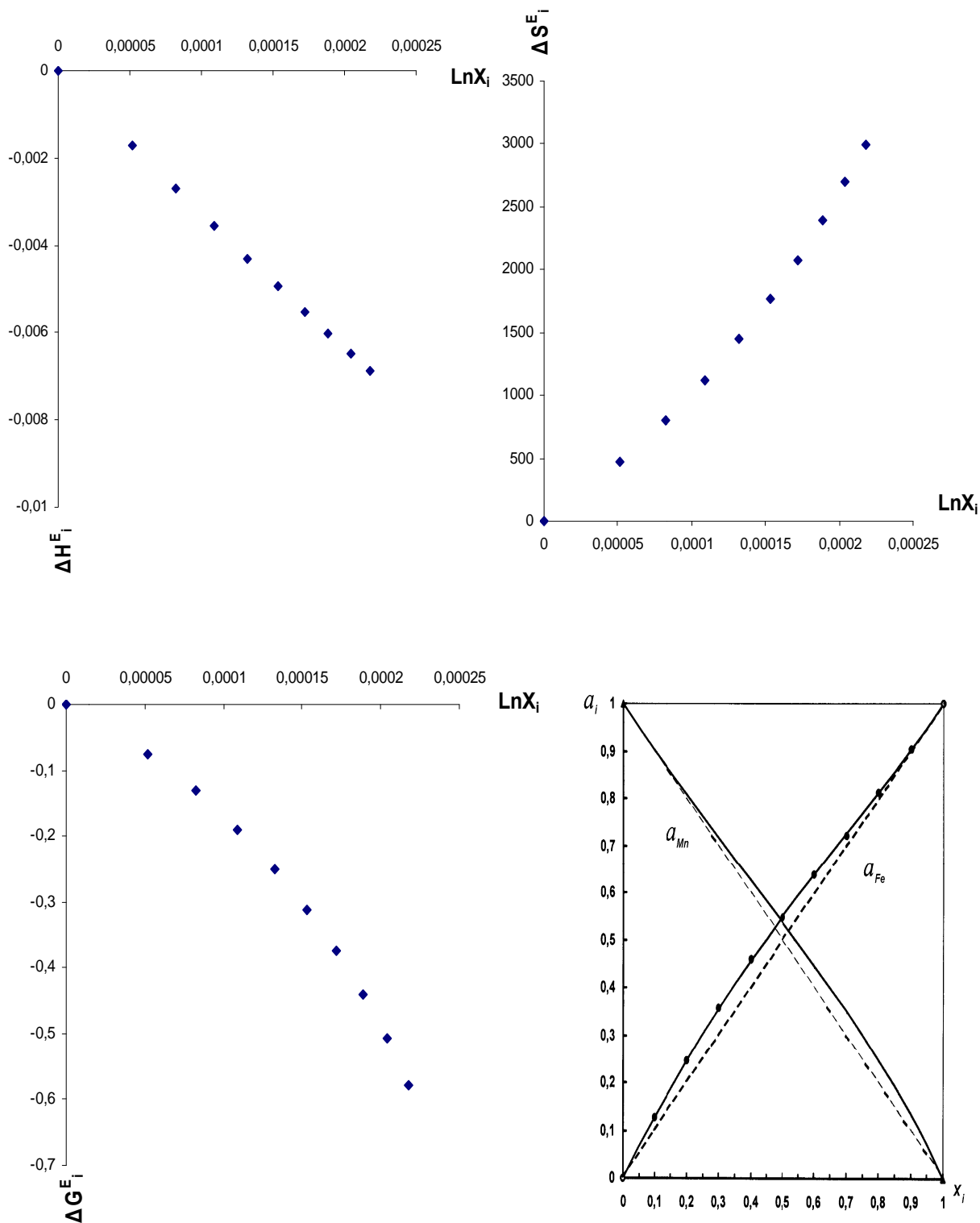


Figure 1 - Partial excess entropy, enthalpy and the Gibbs energy of mixing of the liquidus components in Fe-Mn system

Calculation of excess thermodynamic mixing functions in Cr-based systems

The initial data are taken from [12] and presented in Table 3.

Table 4 - Calculated thermodynamic data of α -Fe in Fe-Cr melt

No. in sequence	T,K	ΔH kJ/g-at.	ΔS kJ/g-at-K	ΔG kJ/mole
1	1812	0	0	0
2	1803	0.00192	319.5202762	-0.03789
3	1793	0.004722	670.7393355	-0.08159
4	1783	0.008638	1017.922868	-0.12702
5	1773	0.014515	1361.037675	-0.17422
6	1763	0.024346	1700.050331	-0.22322
7	1753	0.044214	2034.927181	-0.27408
8	1743	0.105791	2365.63435	-0.32683

Calculation of excess thermodynamic mixing functions in Si-based systems

The initial data are taken from [12] and presented in Table 5.

Table 5 - Input parameters for calculating thermodynamic data in Fe-Si system

No. in sequence	T,K	$X_{opt. Fe}^L$	$\ln X_{Fe}^L$	Φ'_{Fe}	Φ''_{Fe}	a_{Fe}^L	1/T
1	2	3	4	5	6	7	8
2	1811	1	0	1.151	0.39	1	5.552
3	1773	0.955	-3.80722E-05	1.345	0.420	0.984	5.640
4	1733	0.91	-8.75179E-05	1.624	0.454	0.961	5.770
5	1693	0.87	-0.00013	1.913	0.489	0.937	5.906
6	1653	0.835	-0.00017	2.218	0.525	0.913	6.049
7	1613	0.805	-0.00021	2.514	0.561	0.888	6.199
8	1573	0.775	-0.00024	2.826	0.598	0.863	6.357
9	1533	0.748	-0.00027	3.174	0.633	0.837	6.523
10	1493	0.705	-0.00032	3.841	0.644	0.811	6.697
11	1485	0.69	-0.00033	4.012	0.647	0.804	6.743

The crystallization area of α -Fe is described by the equation:

$$\Phi''_{Fe} = 1.8685 + 1.4711 \cdot a_i$$

and that for the area of the ordered phase α_2 -Fe by

$$\Phi''_{Fe} = 0.98655 + 0.4216 \cdot a_i$$

The thermodynamic characteristics of iron in Fe-Si melt calculated by equations (11-13) are presented in Table 6 and in Fig. 3.

Table 6 - Calculated thermodynamic data of α and α_2 -Fe in Fe-Si melt

No. in sequence	T,K	ΔH kJ/g-at.	ΔS kJ/g-at-K	ΔG kJ/mole
1	1811	0	0	0
2	1773	-0.000837	-624.71	-0.077
3	1733	-0.00187	-1481.49	-0.180
4	1693	-0.00276	-2295.06	-0.275
5	1653	-0.00352	-3064.01	-0.359
6	1613	-0.00418	-3786.88	-0.432
7	1573	-0.00473	-4462.19	-0.492
8	1533	-0.0015	-1458.67	-0.541
9	1493	-0.00168	-1623.72	-0.619
10	1485	-0.00173	-1662.67	-0.638

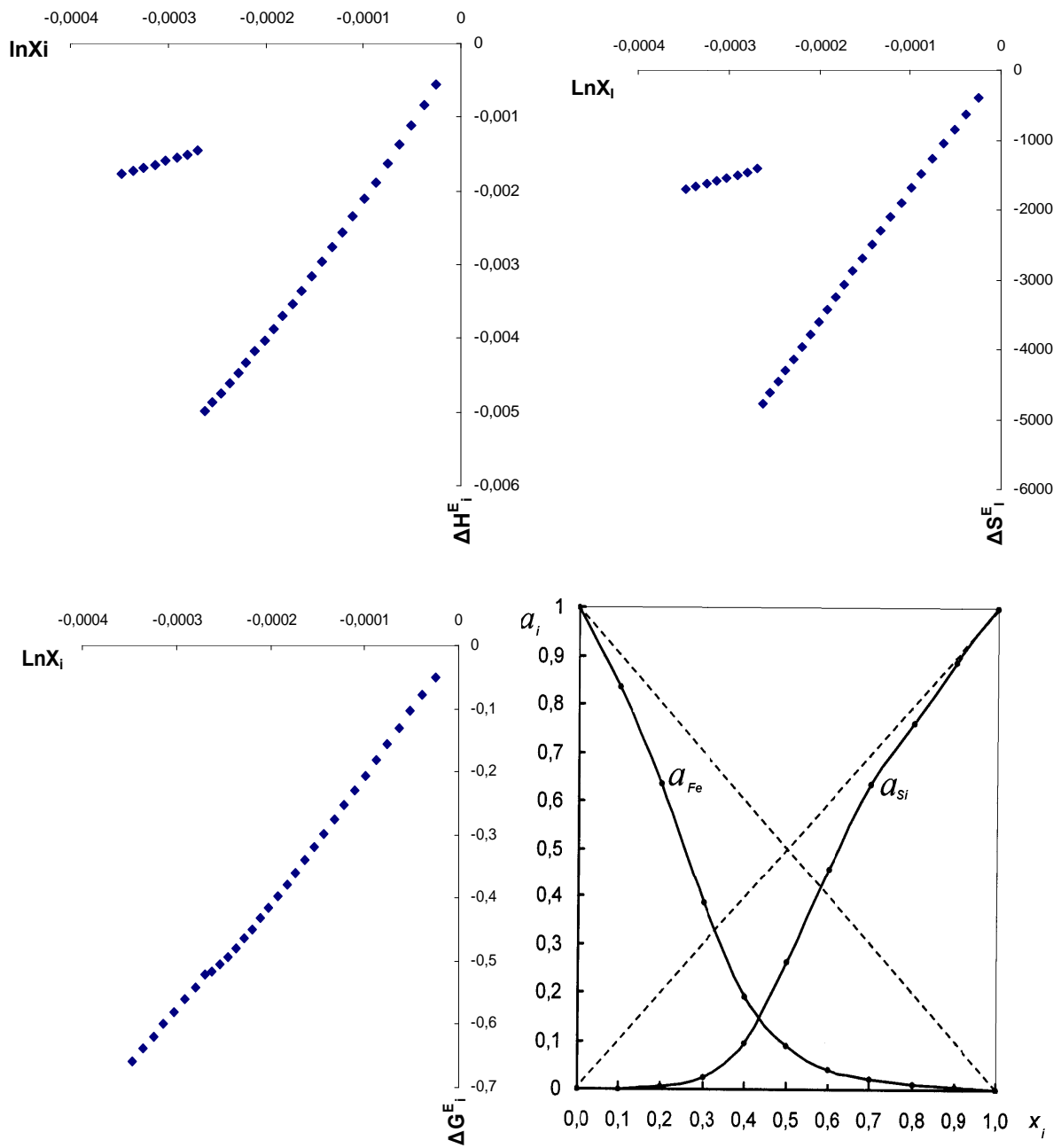


Figure 3 - Partial excess entropy, enthalpy and the Gibbs energy of mixing of the liquidus components in Fe-Si system

Calculation of excess thermodynamic mixing functions in Al-based systems

The initial data are taken from [4] and presented in Table 7. The crystallization area of α -Fe is described by the equation:

$$\Phi''_{Fe} = 0.3989 + 0.0495 \cdot a_i,$$

Table 7 - Input parameters for calculating thermodynamic data in Fe-Al system

No. in sequence	T, K	$X_{\text{opt. Fe}}^L$	$\ln X_{\text{Fe}}^L$	Φ'_{Fe}	Φ''_{Fe}	a_{Fe}^L	1/T
1	1811	1.0	0	0.7033	-0.3494	1	5.534
2	1773	0.808	-4.3E-05	0.764	-0.350	0.980	5.640
3	1733	0.684	-9.7E-05	0.837	-0.351	0.957	5.77
4	1693	0.608	-0.00015	0.911	-0.352	0.934	5.906
5	1653	0.551	-0.00021	0.987	-0.353	0.910	6.049
6	1613	0.505	-0.00027	1.065	-0.355	0.885	6.199
7	1573	0.466	-0.00034	1.116	0.435	0.860	6.357
8	1533	0.428	-0.00041	1.052	0.405	0.834	6.523
9	1493	0.387	-0.00048	0.986	0.374	0.808	6.697
10	1488	0.376	-0.0005	0.969	0.367	0.803	6.723

and that for the region of the ordered phase α_2 -Fe is described by:

$$\Phi''_{\text{Fe}} = -0.5715 + 1.1707 \cdot a_i.$$

The thermodynamic characteristics of iron in Fe-Al melt calculated by equations (11-13) are presented in Fig. 4 and table 8.

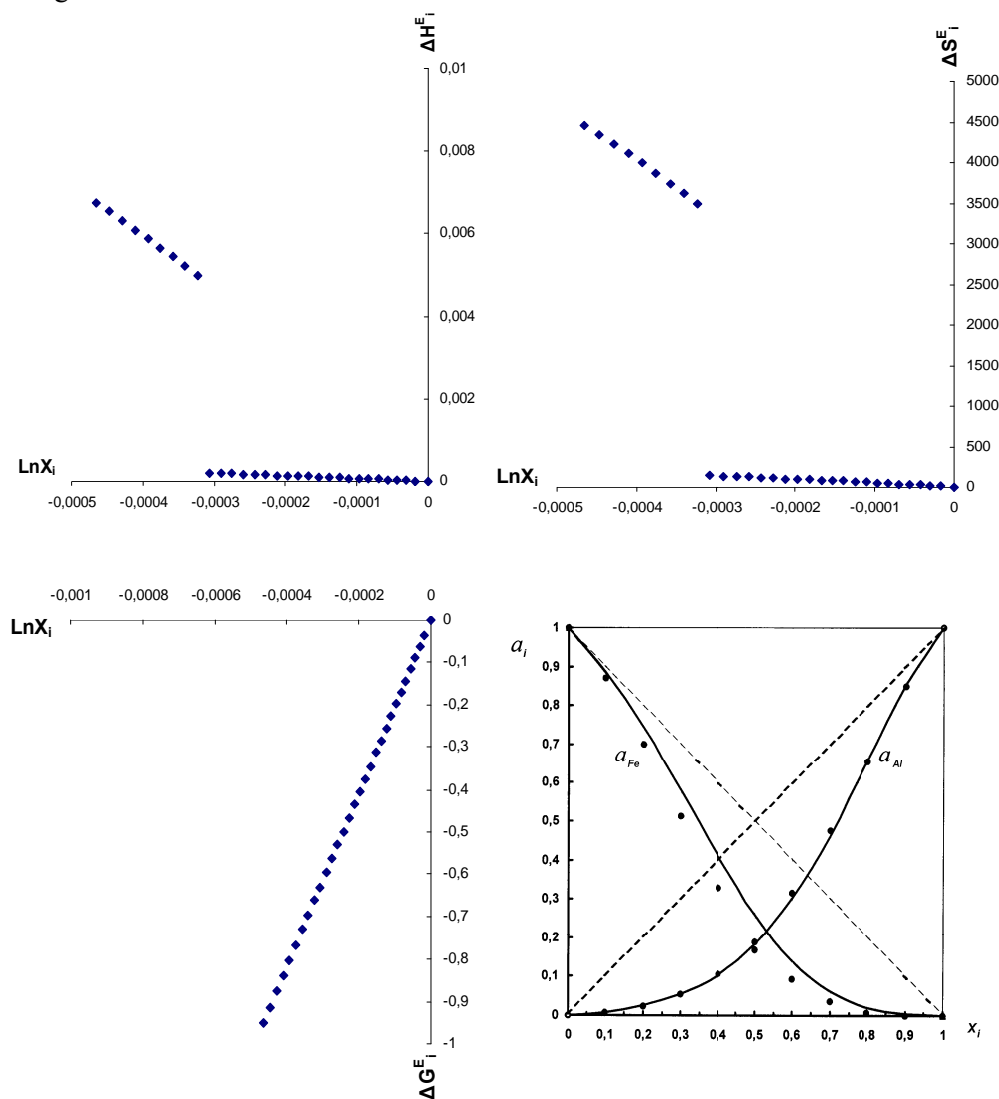


Figure 4 - Partial excess entropy, enthalpy and the Gibbs energy of mixing of the liquidus components in Fe-Al system

Table 8 - Calculated thermodynamic data of α and α_2 -Fe in Fe- Al melt

No. in sequence	T, K	ΔH kJ/g.at.	ΔS kJ/g.at-K	ΔG kJ/mole
1	1811	0	0	0
2	1773	3.2E-05	25.2	-0.089
3	1733	6.02E-05	53.6	-0.198
4	1693	8.85E-05	80.6	-0.314
5	1653	0.00012	106.1	-0.435
6	1613	0.00016	130.0	-0.563
7	1573	0.0002	3619.0	-0.696
8	1533	0.00565	4111.1	-0.839
9	1493	0.00653	4563.1	-0.989
10	1488	0.00674	4669.8	-1.028

Thus, by the example of Fe-Cr, Fe-Si, Fe-Al, Fe-Mn state diagrams, we illustrated the technique for solving the inverse Gibbs problem - extraction of the thermodynamic information directly from the state diagram.

The thermodynamic characteristics of solutions with positive and negative deviations from ideality have been calculated. In doing so we have found that in metal systems of Fe-Cr, Fe-Si, Fe-Al impurity elements Si and Al in the region of rich in chromium alloys exhibit positive deviations from ideality.

This fact allows to draw conclusions about the possibility of deep refining of ferrochromium from these elements, especially aluminum, using a complex alloy of ferrosilicoaluminum (FSA), since the above elements have extremely low solubility in the leading element (chromium) and in this respect using of FSA as a reducing agent does not cause certain difficulties.

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ФЕРРОҚОРЫТПАЛАРДЫ БАЛҚЫТУҒА ТЕРМОДИНАМИКАЛЫҚ БАҒАЛАУ**

Аннотация. Бұл жұмыста бинарлы жүйелердің фазалық тепе-теңдік сызықтарын математикалық өрнекпен сипаттауға мүмкіндік беретін Шредер-Ле-Шателье түрлендірілген теңдеуі негізіндегі күй диаграммасынан термодинамикалық деректерді бөліп шығару әдістемесі ұсынылған. Fe-Si, Fe-Cr, Fe-Mn, Fe-Al бинарлы жүйелері үшін идеалды α - γ ерітіндісінің темір белсенділігінен темірдің кристалдану облысына Бьеррум-Гуггенгейм коэффициентінің тәуелділік теңдеуінің жарамдылығы көрсетілген. Аталған жүйелердің есептік парциалды шектен тыс энтропиялары, энтальпиялары және ликвидус компоненттерінің араласуының Гиббс энергиясы ұсынылған.

Түйін сөздер: кешенді қорытпалар, ферросиликоалюминий, Шредер-Ле-Шателье түрлендірілген теңдеуі, Бьеррум-Гуггенгейм коэффициенті, парциалды шектен тыс энтальпия (ΔH^E), энтропия (ΔS^E) және ликвидус компонентінің араласуының Гиббс энергиясы (ΔG^M).

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С.О. Байсанов¹, В.В. Толоконникова^{1*}, Г.И. Нарикбаева¹, И.Я. Корсукова¹, В.И. Жучков²¹Химико-металлургический институт им.Ж.Абишева, Казахстан, Караганда;²Институт металлургии УрО РАН, Екатеринбург, Россия**ТЕРМОДИНАМИЧЕСКАЯ ОЦЕНКА ВЫПЛАВКИ МАРГАНЦЕВЫХ
И ХРОМИСТЫХ ФЕРРОСПЛАВОВ НА ОСНОВЕ АНАЛИЗА ИХ ДИАГРАММ СОСТОЯНИЯ**

Аннотация. В работе предложена методика извлечения термодинамических данных из диаграмм состояния в основе которых лежит модифицированное уравнение Шредера-Ле-Шателье, позволяющее математическими выражениями описывать линии фазовых равновесий бинарных систем. Показана применимость уравнения зависимости коэффициента Бьеррума-Гуггенгейма для области кристаллизации железа от активности железа идеального α - γ раствора для бинарных систем Fe-Si, Fe-Cr, Fe-Mn, Fe-Al. Предложены расчетные парциальные избыточные энтропии, энтальпии и энергии Гиббса смешения ликвидусных компонентов указанных систем.

Ключевые слова: комплексные сплавы, ферросиликоалюминий, модифицированное уравнение Шредера-Ле-Шателье, коэффициент Бьеррума-Гуггенгейма, парциальная избыточная энтальпия (ΔH^E), энтропия (ΔS^E) и энергия Гиббса смешения (ΔG^M) ликвидусного компонента.

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