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## **THERMODYNAMIC MODELING OF FLUX REFINING OF PRIMARY ALUMINUM IN A BUCKET**

*This article provides a thermodynamic analysis of the process of processing primary aluminum with boric acid in a ladle. HSC Chemistry 9.0 software from Outotec Technologies was used for calculations. In the calculations, the temperature range under study corresponded to the production conditions of Kazakhstan Electrolysis Plant JSC within the range of 650–950 °C. The pressure range was taken based on the technological conditions for flux processing of aluminum in a transfer ladle with a working height of 2 m and was in the range of 101.33–148.99 kPa. The presence of the solid phase and low kinetics of the processes are characterized by the lower limit of the specified temperature range. The upper temperature limit is the closest to real operating conditions during electrolysis. The pressure at the metal mirror is represented by the lower limit in the pressure range, the upper limit is the pressure at the immersion depth of the flux equivalent to 2 m. The immersion depth of the sample in the calculations varied in the range of 0.5–2 m.*

*The research was carried out within the framework of grant funding from the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan under the «Zhas Galym» competition under the project IRN AR19175493 «Development of technology for refining primary aluminum with filters with an active surface».*

*Thermodynamic analysis in the studied range of temperatures and pressures clearly indicates that vanadium boride phases are more stable compared to aluminum borides, and, therefore, they will form preferentially in this temperature range. The order of stability also suggests that vanadium can be easily removed from aluminum melts by adding boron.*

*Keywords:* aluminum, thermodynamic modeling, flux, boric acid, refining.

### **Introduction**

In the production of anodes for electrolysis baths in the conditions of Kazakhstan Electrolysis Plant JSC (KEZ JSC), coke produced by UPNK-PV LLP (Pavlodar) is partially involved in the production. Locally produced coke contains a high degree of vanadium and other impurities, which in turn limits its use at the enterprise.

A literature review showed that there are methods for refining primary aluminum from vanadium impurities with boron compounds, which have not received widespread industrial use [1–15].

From works [13 – 15] it is known that the occurrence of chemical reactions in the Al-V-B system is complex.  $\text{VB}_2$  compounds are thermodynamically stable at operating temperatures of the electrolysis and aluminum casting process of 950–650 °C. The kinetics of chemical reactions with the formation of  $\text{VB}_2$  compounds during refining with Al-B based alloys is characterized by a low rate due to the formation of an insoluble ring of reaction products consisting of vanadium diboride ( $\text{VB}_2$ ).

### Materials and methods

In this work, a thermodynamic analysis of the process of processing primary aluminum with boric acid was carried out according to the method described in [16]. HSC Chemistry 9.0 software from Outotec Technologies was used for calculations.

When refining a molten raw aluminum with boric acid at a temperature in the range of 650–950 °C and a pressure in the range of 101.33 kPa, boric acid ( $\text{H}_3\text{BO}_3$ ) undergoes a transformation into boron oxide ( $\text{B}_2\text{O}_3$ ), which in turn interacts with the molten aluminum and then with compounds vanadium according to the following formulas:



In the calculations, the temperature range under study corresponded to the production conditions of KEZ JSC within the range of 650–950 °C. The pressure range was taken based on the technological conditions for flux processing of aluminum in a transfer ladle with a working height of 2 m in the range of 101.33–148.99 kPa. The presence of a solid phase and low kinetics of processes are characterized by the lower limit of the specified range. The upper limit is the closest to real operating conditions during electrolysis. The pressure at the metal mirror is represented by the lower limit in the range, the upper limit is the pressure at the immersion depth of the flux equivalent to 2 m. The immersion depth of the sample varies in accordance with the height of the ladle and was in the range of 0.5–2 m. The temperature range was taken in accordance with the operating conditions in the workshop: from 650 °C to 950 °C, corresponding to the highest density of aluminum melt (2380 kg/m<sup>3</sup>) in the considered temperature range.

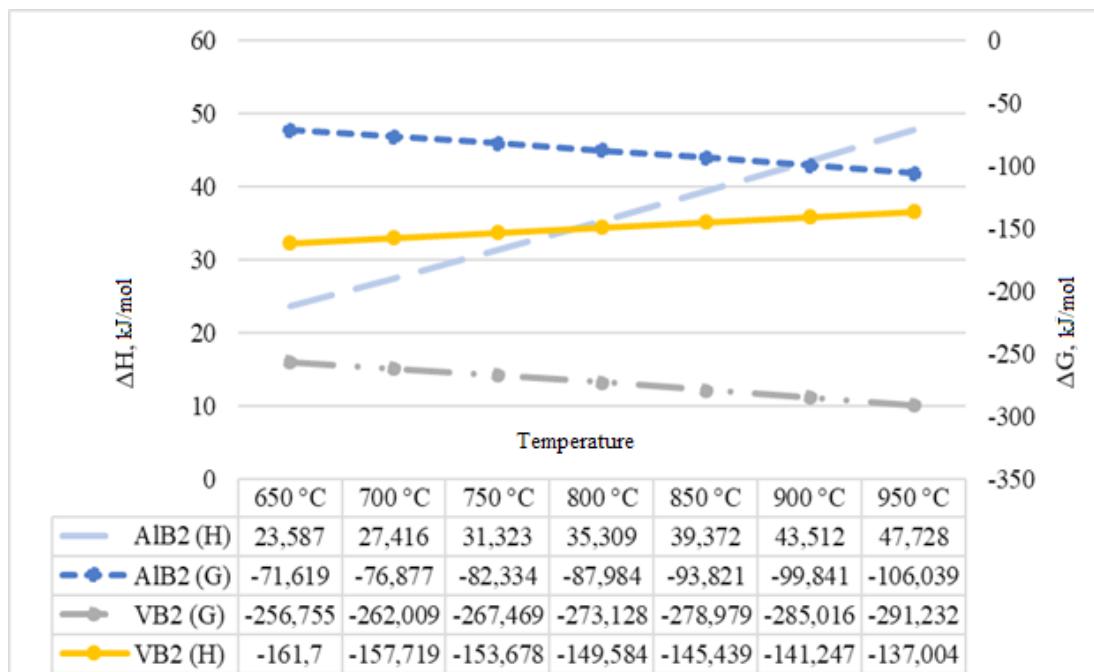
### Results and discussion

The thermodynamic parameters were calculated for compounds  $\text{AlB}_2$  and  $\text{VB}_2$ . Table 1 and Figure 1 show the calculated values  $C_p$ ,  $\Delta G$ ,  $\Delta H$ ,  $\Delta S$ .

Table 1 – Calculated values  $C_p$ ,  $\Delta G$ ,  $\Delta H$ ,  $\Delta S$  for compounds  $AlB_2$  and  $VB_2$ 

Compound	Parameter	Design temperature range, °C (at a pressure of 102.39 kPa)						
		650 °C	700 °C	750 °C	800 °C	850 °C	900 °C	950 °C
$AlB_2$	$C_p$	75,764	77,363	78,938	80,494	82,033	83,560	85,075
	$\Delta H$	23,587	27,416	31,323	35,309	39,372	43,512	47,728
	S	103,132	107,171	111,086	114,889	118,590	122,196	125,715
	$\Delta G$	-71,619	-76,877	-82,334	-87,984	-93,821	-99,841	-106,039
$VB_2$	$C_p$	79,019	80,232	81,361	82,409	83,381	84,277	85,524
	$\Delta H$	-161,700	-157,719	-153,678	-149,584	-145,439	-141,247	-137,004
	S	102,968	107,168	111,216	115,123	118,898	122,549	126,090
	$\Delta G$	-256,755	-262,009	-267,469	-273,128	-278,979	-285,016	-291,232

At temperatures of 650 – 950 °C and atmospheric pressure, the change in Gibbs energy for compounds  $VB_2$  is -256.755 and – 291.232 kJ/mol, and  $AlB_2$  is -71.619 and -106.039 kJ/mol, respectively. The change in enthalpy is a negative value for the reactions of vanadium diboride; therefore, the formation of this compound is exothermic in nature, while the formation of aluminum diboride, on the contrary, is endothermic in nature due to positive enthalpy.

Figure 1 – Dependence of the change in Gibbs energy and change in enthalpy for  $VB_2$  and  $AlB_2$  on temperature

The work also included a calculation of the thermodynamic parameters of the chemical reaction  $AlB_2 + V = VB_2 + Al$ . Table 2 and Figure 2 show the calculated values  $\Delta G$ ,  $\Delta H$ ,  $\Delta S$ ,  $\log k$ .

Table 2 – Calculated values  $\Delta G$ ,  $\Delta H$ ,  $\Delta S$ ,  $\log k$  for a chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$

Chemical reaction	Parameter	Design temperature range, °C (at a pressure of 102.39 kPa)						
		650 °C	700 °C	750 °C	800 °C	850 °C	900 °C	950 °C
$\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$	$\Delta H$	-163,214	-173,692	-173,476	-173,302	-173,176	-173,102	-173,076
	$S$	22,900	11,668	11,885	12,051	12,166	12,232	12,253
	$\Delta G$	-184,354	-185,047	-185,637	-186,235	-186,841	-187,451	-188,063
	$\log k$	10,432	9,933	9,478	9,066	8,690	8,347	8,032

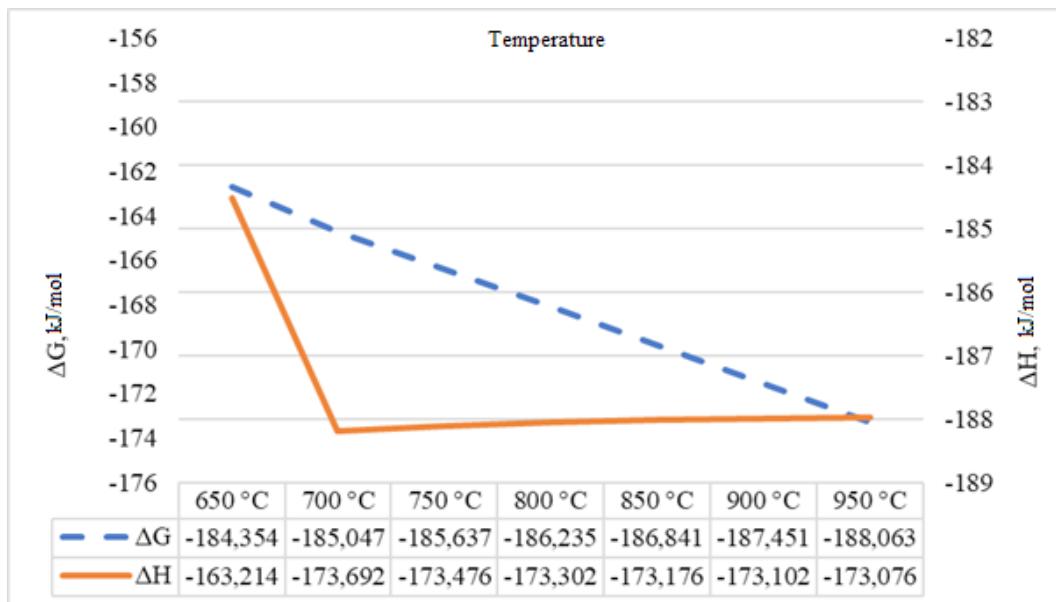


Figure 2 – Graph of changes in Gibbs energies and enthalpy for a chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  depending on temperature

Analysis of thermodynamic parameters confirms that at atmospheric pressure and temperatures from 650 to 950 °C the reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  proceeds with the formation of vanadium diboride ( $\text{VB}_2$ ) the change in Gibbs energy is  $-184.354$  and  $-188.676$  kJ/mol, respectively. An increase in the Gibbs energy with increasing temperature indicates an increase in the stable nature of the occurrence of chemical reactions. The change in enthalpy as a whole is a negative value; therefore, the formation of these compounds is exothermic in nature. It can be concluded that the chemical reaction is thermodynamically favorable.

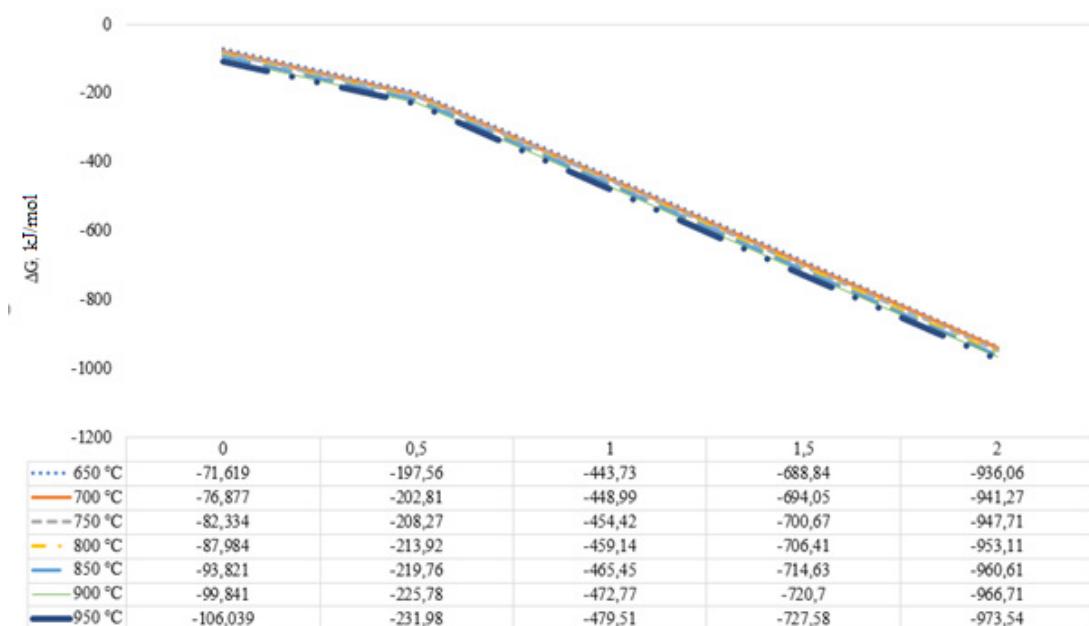
Further in the work, a thermodynamic analysis was carried out for the conditions of immersion of the flux into the melt to a depth of 0.5 m, 1 m, 1.5 m, 2 m.

Table 3 and Figure 3 show the calculated values of the Gibbs energy for compounds  $\text{VB}_2$  and  $\text{AlB}_2$ .

Table 3 –  $\Delta G$  for compounds  $\text{AlB}_2$  and  $\text{VB}_2$  at a depth of 0.5 m, 1 m, 1.5 m, 2 m.

Compound	Depth of flux immersion into the melt, m	Design temperature range, °C						
		650 °C	700 °C	750 °C	800 °C	850 °C	900 °C	950 °C
$\text{AlB}_2$	0,5	-197,56	-202,81	-208,27	-213,92	-219,76	-225,78	-231,98
	1,0	-443,73	-448,99	-454,42	-459,14	-465,45	-472,77	-479,51
	1,5	-688,84	-694,05	-700,67	-706,41	-714,63	-720,70	-727,58
	2,0	-936,06	-941,27	-947,71	-953,11	-960,61	-966,71	-973,54
$\text{VB}_2$	0,5	-307,17	-312,42	-317,88	-323,54	-329,39	-335,43	-341,64
	1,0	-689,91	-694,82	-700,28	-706,13	-713,28	-719,41	-726,74
	1,5	-1070,94	-1076,01	-1083,34	-1091,45	-1100,63	-1107,26	-1118,38
	2,0	-1455,38	-1461,57	-1468,48	-1475,58	-1485,12	-1491,89	-1511,17

At a temperature of 650 °C and pressures of 102.39 kPa (at the surface of the melt) and 148.99 kPa (when introducing flux to a depth of 2 m), the change in the Gibbs energy for the formation of compound  $\text{VB}_2$  is -256.755 and -1455.3 kJ/mol, respectively. For the  $\text{AlB}_2$  compound under such conditions, the change in Gibbs energy is -71.619 and -936.06 kJ/mol. for the initial temperature, and -291.232 and -1511.17 kJ/mol for a temperature of 950 °C, respectively, for  $\text{VB}_2$ . For  $\text{AlB}_2$ , the change in Gibbs energy is -106.039 and -973.54 kJ/mol.

Figure 3 – Dependence of the change in Gibbs energy for  $\text{VB}_2$  and  $\text{AlB}_2$  on temperature and immersion of boron-based flux into the aluminum melt at different temperatures

Further in the work, similar thermodynamic calculations were carried out to change the isobaric-isothermal potential of the system for a chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$ .

Table 4 and Figure 4 show the calculated Gibbs energy values for the chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$

Table 4 –  $\Delta G$  for the chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  at a depth of 0.5 m, 1 m, 1.5 m, 2 m

Compound	Depth of flux immersion into the melt, m	Design temperature range, °C						
		650 °C	700 °C	750 °C	800 °C	850 °C	900 °C	950 °C
$\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$	0	-184,354	-185,047	-185,637	-186,235	-186,841	-187,451	-188,063
	0,5	-276,08	-278,13	-279,25	-279,90	-280,55	-281,12	-282,55
	1,0	-511,98	-515,85	-520,12	-526,85	-533,35	-540,22	-545,23
	1,5	-794,788	-799,556	-810,453	-830,612	-850,463	-856,123	-860,462
	2,0	-1080,034	-1100,236	-1122,123	-1135,185	-1145,252	-1148,22	-1173,54

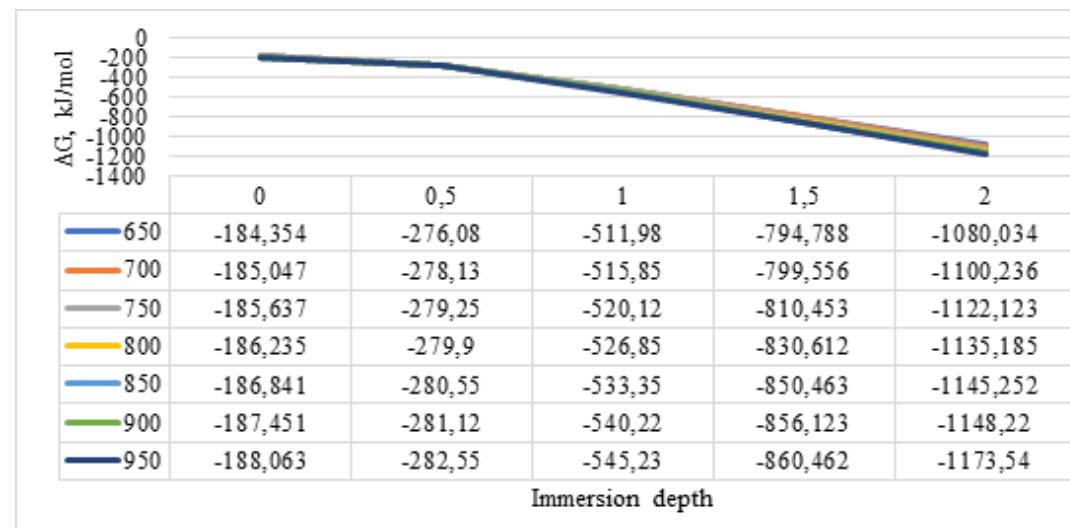


Figure 4 – Dependence of the change in the Gibbs energy for the chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  on the temperature and depth of immersion of a sample of boron-based flux into the aluminum melt at different temperatures

At a temperature of 650 °C (Figures 4 and 5) for pressures of 102.39 kPa (at the melt surface) and 148.99 kPa (at a flux immersion depth of 2 m), the change in Gibbs energy for the chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  is -184.354 and -1080.034 kJ/mol, respectively. For a temperature of 950 °C these values are -188.063 and -1173.54 kJ/mol.

This important observation shows that as pressure increases at constant temperature, the magnitude of the difference in changes in isobaric-isothermal potential decreases, especially noticeably at elevated temperatures. On the other hand, as the temperature decreases at constant pressure, the value of the difference in Gibbs energy changes also decreases, and this trend is more noticeable as the pressure in the system increases.

At a temperature of 650 °C (Figures 4 and 5) for the reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$ , the difference in changes in the Gibbs energy when the pressure in the system increases from 102.39 kPa to 114.04 kPa is -91.726 kJ/mol, from 114.04 kPa to 125.69 kPa

-235.9 kJ/mol, from 125.69 kPa to 137.29 kPa -282.808 kJ/mol, and from 137.29 kPa to 148.99 kPa -285.246 kJ/mol. At 950 °C these Gibbs energy changes are -94.487 kJ/mol, -262.68 kJ/mol, -315.232 kJ/mol and -313.078 kJ/mol respectively.

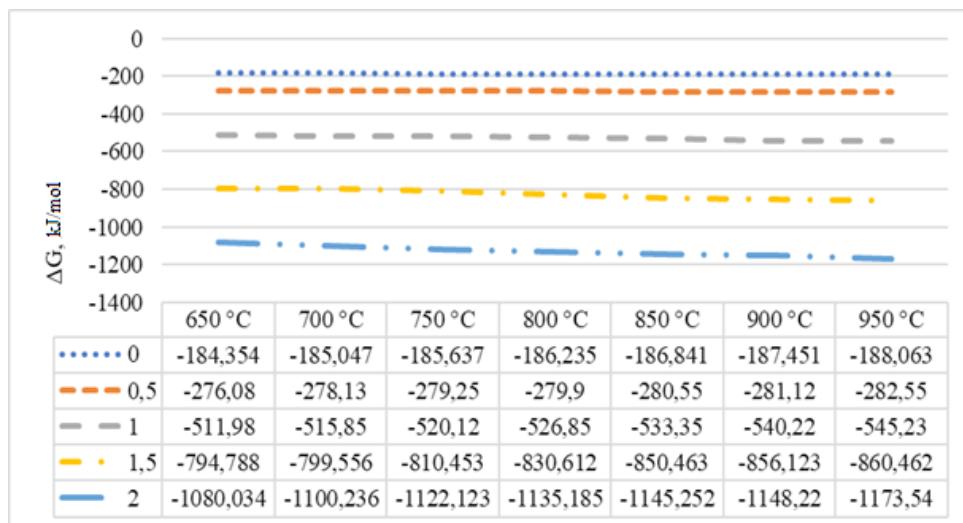


Figure 5 – Graph of the difference in changes  $\Delta G$  for the chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  on temperature with a fixed increase in the depth of immersion of a sample of boron-based flux into the aluminum melt with a change in the depth of immersion.

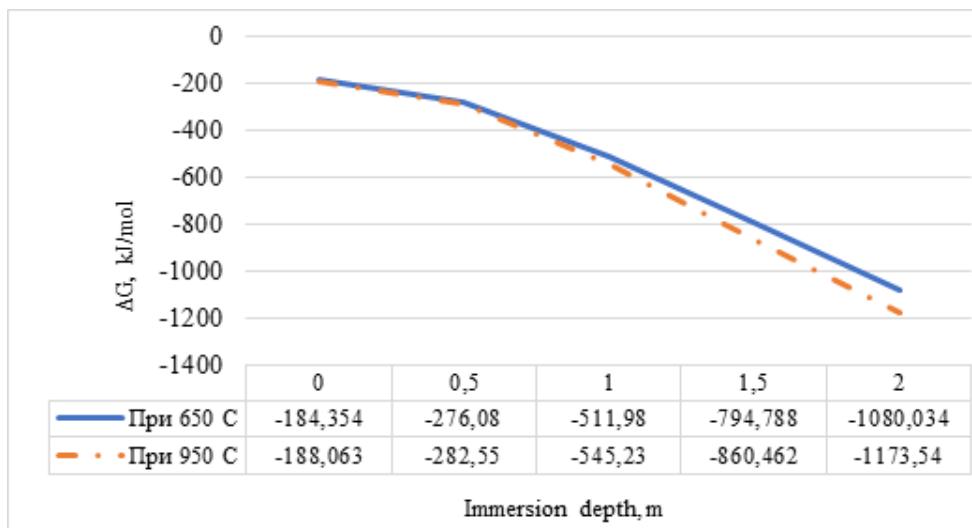


Figure 6 – Graph of the difference in changes  $\Delta G$  for the chemical reaction  $\text{AlB}_2 + \text{V} = \text{VB}_2 + \text{Al}$  on the depth of immersion of a sample of boron-based flux into the aluminum melt as the temperature changes

This observation emphasizes that chemical reactions between primary aluminum and impurities, including vanadium with boron with the formation of intermetallic compounds, can proceed in a certain direction in the considered range of temperatures and pressures (Figure 6).

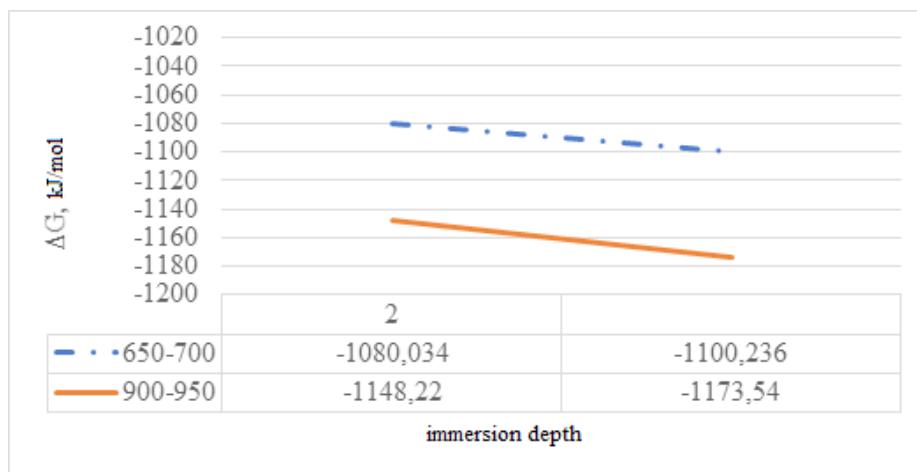


Figure 7 – Dependence of the difference in changes in the isobaric-isothermal potential of the reaction on the immersion depth of the sample

As can be seen from Figures 1–7, in the temperature range from 650 to 950 °C and pressures from 102.34 kPa to 149.88 kPa, vanadium boride phases are more stable compared to aluminum borides, and, therefore, they will be formed predominantly in this temperature range. The order of stability also suggests that vanadium can be easily removed from aluminum melts by adding boron.

### Conclusions

Thermodynamic analysis in HSC Chemistry 9.0. in the Al-V-B system in the studied range of temperatures and pressures showed that the values of the Gibbs energy ( $\Delta G$ ) over the entire range of operating temperatures of the process of electrolysis and casting of primary aluminum for VB<sub>2</sub> are significantly lower than AlB<sub>2</sub>, therefore, they will be formed predominantly in this temperature range. The order of stability also suggests that vanadium can be easily removed from aluminum melts by adding boron.

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## ШЕЛЕКТЕГІ БАСТАПҚЫ АЛЮМИНИЙДІ АҒЫНМЕН ТАЗАРТУДЫ ТЕРМОДИНАМИКАЛЫҚ МОДЕЛЬДЕУ

Бұл мақалада шелектегі бор қышқылымен бастапқы алюминийді өңдеу процесіне термодинамикалық талдау жасалды. Есептеу үшін Outotec Technologies компаниясының HSC Chemistry 9.0 бағдарламалық құралы пайдаланылды. Есептеулерде зерттелетін температура диапазоны 650–950 °C шегінде «Қазақстандық электролиз зауыты» АҚ өндірістік шарттарына сәйкес келді. Қысым диапазоны жұмыс биіктігі 2 м үлестірмелі шелектегі алюминийді ағынмен өңдеудің технологиялық жағдайлары негізінде қабылданды және 101,33–148,99 кПа аралығында болды. Қатты фазаның болуы және процестердің төмен кинетикасы көрсетілген температура диапазонының төменгі шегімен сипатталады. Электролиздегі нақты жұмыс жағдайларына ең жақын температура шегі көрсетіледі. Металл айнасындағы қысым қысым диапазонындағы төменгі шек болып көрінеді, жогары жағы 2 м эквивалентті ағынның батыру тереңдігіндегі қысым болып табылады. Есептеулердегі суспензияның батыру тереңдігі 0,5–2 м диапазонында өзгерді.

Зерттеулер Қазақстан Республикасы Фылым және жыныс министрлігі Фылым комитетінің «Жас ғалым» конкурсы бойынша ЖРН AP19175493 «белсенді беті бар сүзгілермен бастапқы алюминийді тазарту технологиясын өзірлеу» жобасы бойынша гранттық қаржыландыру шеңберінде жүргізілді.

Зерттелген температура мен қысым диапазонындағы термодинамикалық талдау ванадий боридінің фазалары алюминий боридтерімен салыстырғанда тұрақтырақ екенін анық көрсетеді, сондықтан олар негізінен берілген температура диапазонында түзіледі. Тұрақтылық тәртібі сонымен қатар ванадийді бор қосу арқылы алюминий балқымаларынан оңай алып тастауга болатындығын көрсетеді.

Кілтті сөздер: алюминий, термодинамикалық модельдеу, ағын, бор қышқылы, тазарту.

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## ТЕРМОДИНАМИЧЕСКОЕ МОДЕЛИРОВАНИЕ ФЛЮСОВОГО РАФИНИРОВАНИЯ ПЕРВИЧНОГО АЛЮМИНИЯ В КОВШЕ

В данной статье проведен термодинамический анализ процесса обработки первичного алюминия борной кислотой в ковше. Для расчетов использовалось программное обеспечение HSC Chemistry 9.0 компании Outotec Technologies. В расчетах исследуемый температурный диапазон соответствовал производственным условиям АО «Казахстанский электролизный завод» в пределах 650–50 °C. Диапазон давлений принимался исходя из технологических условий флюсовой обработки алюминия в раздаточном ковше с рабочей высотой 2 м и находился в пределах 101,33–148,99 кПа. Наличие твердой фазы и низкая кинетика процессов характеризуется нижним пределом указанного диапазона температур. Наиболее приближенным к реальным рабочим условиям при электролизе демонстрирует верхний предел температур. Давление у зеркала металла представляется нижним пределом в диапазоне давлений, верхним представлено давление на глубине погружения флюса эквивалентного 2 м. Глубина погружения навески в расчетахарьировалась в диапазоне 0,5–2 м.

Исследования проводились в рамках грантового финансирования Комитета науки Министерства науки и высшего образования Республики Казахстан по конкурсу «Жас Галым» по проекту ИРН AP19175493 «Разработка технологии рафинирования первичного алюминия фильтрами с активной поверхностью».

Термодинамический анализ в исследованном диапазоне температур и давлений однозначно свидетельствует о том, что фазы борида ванадия более стабильны по сравнению с боридами алюминия, и, следовательно, они будут образовываться преимущественно в данном температурном диапазоне. Порядок стабильности также предполагает, что ванадий может быть легко удален из расплавов алюминия путем добавления бора.

**Ключевые слова:** алюминий, термодинамическое моделирование, флюс, борная кислота, рафингование.

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