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Thermodynamic Modeling of the Vanadium Alloy Smelting Process Using a Silicon-aluminum Reducing Agent

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Abstract. *The article is devoted to the study of the theoretical aspect of obtaining a vanadium alloy by aluminosilicothermic method. Vanadium pentoxide and converter vanadium slag were studied as charge materials. Silicon-aluminum ferroalloys, such as ferroaluminosilicocalcium, ferrosilicoaluminum and aluminum-silicon-manganese with calcium, will be used as a reducing agent. The chemical composition of the initial vanadium-containing materials and silicon-aluminum reducing agents were studied by the spectral method of analysis. To analyze the aluminosilicothermal reduction of vanadium in the V-Fe-Si-Al-Ca-O system, the method of complete thermodynamic modeling of metallurgical processes was used. Full thermodynamic modeling was implemented in a computer system using the HSC Chemistry software package. As initial data for determining the parameters of thermodynamic equilibrium for the actual composition of the charge for the smelting of vanadium alloys with a stoichiometric consumption of the reducing agent. To analyze the coexistence of phases in a multicomponent system during the aluminosilicothermal reduction of vanadium, a triple phase diagram was constructed for the V_2O_5-Al-Si system at different isotherms using the «Triangle» program. As a result, the processes of aluminosilicothermal reduction of vanadium in the process of smelting a vanadium alloy using silicon-aluminum reducing agents were studied. The optimal temperature range for vanadium alloy smelting was set. The range is 1650-1850 K. Also, the results of thermodynamic studies made it possible to predict the final phase composition of the alloy, in the form of V_5Si_3, VSi_2 and free silicon Si.*

Keywords: *vanadium, ferrosilicoaluminum, silicon-aluminum reducing agent, metallothermy, process modeling, vanadium ligature.*

Introduction

By data from 2016, the world production and consumption of ferrovanadium is about to 86 thousand tons per year. The global demand for vanadium is constantly growing and in 2020 it has already amounted to 102 thousand tons. In this connection, there is a constant increase in the production of this alloy and it is predicted that by 2020 this figure will reach 140 thousand tons [1]-[3]. The average annual growth rate of demand for vanadium in the most developed industrial countries is 5-7%. This is due to its use [4] in the production of low-alloy steels used for the manufacture of pipes for oil and gas pipelines, bridge structures, high-rise buildings, large-span structures, the production of railway rails, etc. It should be noted that in addition to the metallurgical and chemical industries, vanadium and its compounds are widely used in atomic hydrogen energy and in the production of vanadium batteries [5]-[9].

In recent years, due to the increase in the production of various steel grades, the demand for vanadium is constantly growing. The increase in demand for vanadium and vanadium ferroalloys raises the question of providing domestic ferroalloy plants with high-quality raw materials in the future.

Almost 60% of the world's ferrovanadium is produced from vanadium slag, 30% from primary ore, and 10% from secondary vanadium-containing raw materials. In terms of vanadium reserves, a significant part of which is concentrated on the territory of the Kyzylorda region, Kazakhstan occupies one of the first places in the world. One of the largest vanadium deposits (with reserves of more than 2 million tons) is the vanadium-bearing basin of the Big Karatau, the average content of vanadium pentoxide in which is 0,8-1,3%. The Big Karatau basin includes the Bala-Sauksandyk, Dzhabagly and Kurumsak [10].

At the same time, the industry of Kazakhstan

consumes and imports a huge amount of metallurgical products containing vanadium, in the form of railway rails and pipe products for gas and oil pipelines. Therefore, there is a need to develop technologies for the primary processing of local vanadium raw materials to obtain products with a higher added value.

In this connection, the current paper proposes the production of a vanadium-containing alloy by an unconventional method, where it is proposed to use a cheap complex silicon-aluminum ferroalloy with a high content of silicon and aluminum in its composition – ferrosilicoaluminum, instead of traditional aluminum powder and ferrosilicon, as a reducing agent.

The developed technology for smelting vanadium-containing alloys using a silicon-aluminum alloy makes it possible to use cheap raw materials and obtain a low-cost ferroalloy. When reducing vanadium oxides in the production of vanadium master alloys, silicon is partially replaced by a stronger reducing agent, such as aluminum from a silicon-aluminum alloy, which makes it possible to significantly increase the degree of vanadium extraction. At the same time, the cost of the proposed complex reducing agent is significantly lower than traditional ferrosilicon, since it is smelted from cheaper raw materials: high-ash coal, without the use of expensive coke nuts. In addition, there is an annual demand of the industry of Kazakhstan for metal products (in the form of railway rails and large diameter pipes) containing vanadium, in quantities from 60 to 300 tons.

Technological processes for smelting new ferroalloys are a set of chemical reactions accompanied by thermal effects, during which new phases are formed. In one of these phases, which is the target product, they strive to extract the metal from the feedstock as completely as possible, and to transfer the accompanying unnecessary components into others. In order to say with certainty whether certain chemical reactions are possible between the components of the charge and what the chemical composition of the finished alloy is, thermodynamic calculations are required. Accordingly, the purpose of this work is to carry out a complete thermodynamic simulation of the smelting process of a vanadium-containing alloy using software packages.

Materials and methods of research

In this work, it is planned to carry out a complete thermodynamic analysis in order to simulate the process of smelting a vanadium-containing alloy from vanadium pentoxide and converter vanadium slag, where silicon-aluminum ferroalloys, such as ferroaluminosilicocalcium, ferrosilicoaluminum, and alumosilicomanganese with calcium will be used as a reducing agent.

The chemical composition of the initial vanadium-containing materials and silicon-aluminum reducing agents were studied by the spectral method of analysis on a vacuum wave-dispersive X-ray fluorescence spectrometer MAKS-GVM, located in the Zh. Abisheva Chemical-metallurgical institute. The principle of operation of an X-ray spectrometer is based on irradiating the sample with the primary radiation of an X-ray tube, measuring the intensity of the secondary fluorescent radiation from the sample at wavelengths corresponding to the elements being determined, and then calculating the mass fraction of these elements using the constructed calibration characteristic. The results of the spectral analysis are shown in tables 1 and 2.

To analyze the aluminosilicothermal reduction of vanadium, the method of full thermodynamic modeling of metallurgical processes implemented in a computer system using the HSC (enthalpy (H), entropy (S) and heat capacity (C)) Chemistry software package was used. The HSC Chemistry integrated program database is based and updated by SGTE (Scientific Group Thermodata Europe). The error in calculations on the HSC Chemistry software package is no more than 4-6%, which is quite acceptable.

Thermodynamic modeling of arbitrary multicomponent systems consists in determining all equi-

Table 1 – Chemical composition of initial vanadium-containing materials

Material	Content, %								
	V_{total}	Mn_{total}	Fe_{total}	TiO_2	Al_2O_3	CaO	C	S	P
Vanadium pentoxide	55,00	0,02	0,05	-	-	-	-	-	-
Vanadium slag	15,01	9,6	20,9	10,07	3,7	1,89	0,41	0,02	0,023

Table 2 – Chemical composition of silicon-aluminum reducing agents

Material	Content, %					
	Si	Al	Ca	Mn	P	Fe
Ferroaluminosilicocalcium	45-55	20-25	10-18	-	0,021	rest
Alumosilicomanganese with calcium	45-50	25-35	4-10	10-17	0,019	rest
Ferrosilicoaluminum	55-65	15-25	-	-	0,024	rest

librium parameters, thermodynamic properties, as well as the chemical and phase composition of the resulting components. With an increase in temperature, when any changes in state are accompanied by phase, polymorphic and chemical transformations, this problem is immeasurably more difficult than in the formulation of classical thermodynamics, where calculations are performed for normal conditions. However, due to the fact that the fundamental thermodynamic laws remain valid for any systems, their correct application allows us to solve the problem of calculating thermodynamic equilibrium in the general case. Consideration within the framework of a unified approach of significantly different processes and states is possible only with a known formalization of the model description of the objects under study. Any considered thermodynamic system will be characterized by the relative and absolute content of chemical elements in it (mol/kg). By condition, it remains unchanged when equilibrium is established from an arbitrary state and is sufficient to describe the system as a material object.

To present a more complete picture of the redox processes of the aluminosilicothermal reduction of vanadium, thermodynamic calculations were also carried out using the Terra and Triangle programs.

The program «Terra» is intended for calculation of arbitrary systems with chemical and phase transformations. It allows you to simulate limiting equilibrium states and implements the method and algorithm of calculations that was created at the N.E. Bauman Moscow State Technical University. The program is associated with an extensive database of properties of individual substances, which makes it suitable for the study of compositions of arbitrary chemical composition.

Results and its discussion

A complete thermodynamic analysis of the V-Fe-Si-Al-Ca-O system was carried out for the composition of the charge with the normal course of the vanadium ligature melting mode in order to determine the optimal mode of the aluminosilicothermic process. As initial data for determining the parameters of thermodynamic equilibrium for the actual composition of the charge for smelting vanadium alloys with a stoichiometric consumption of the reducing agent, the following content of the working fluid was used: V_2O_5 – 20,29; Fe – 25,64; Si – 19,61; Al – 4,99; SiO_2 – 1,0; Fe_2O_3 – 1,06; Al_2O_3 – 0,67; CaO – 26,73; Ar – 0,01%, which was obtained from the calculations.

As a result of modeling, it was revealed that when smelting an alloy of vanadium master alloy by aluminosilicothermal method up to 2700 K, the formation and some changes of elements are observed with their transition to the gas and condensed phase (Figure 1).

An analysis of the curves presented in Figure 1 shows that during the reduction of vanadium pentoxide in the temperature range of 700-2400 K, the formation of a condensed VSi_2 phase is observed. In the temperature range of 700-1300 K, its content is 23.8%. Further, in the range of 1400-1800 K, its content decreases to 11,38%, in parallel with the formation of another condensed phase V_5Si_3 with a content of 7,87%. Further, with an increase in temperature to 1900 K and above, the V_5Si_3 phase completely disappears, and the VSi_2 compound again reaches 23.81%. The content of the iron phase (Fe) in the temperature range of 700-2200 K remains unchanged – 28.04%, starting to gradually decrease as it approaches the boiling point.

It is possible to form compounds of vanadium

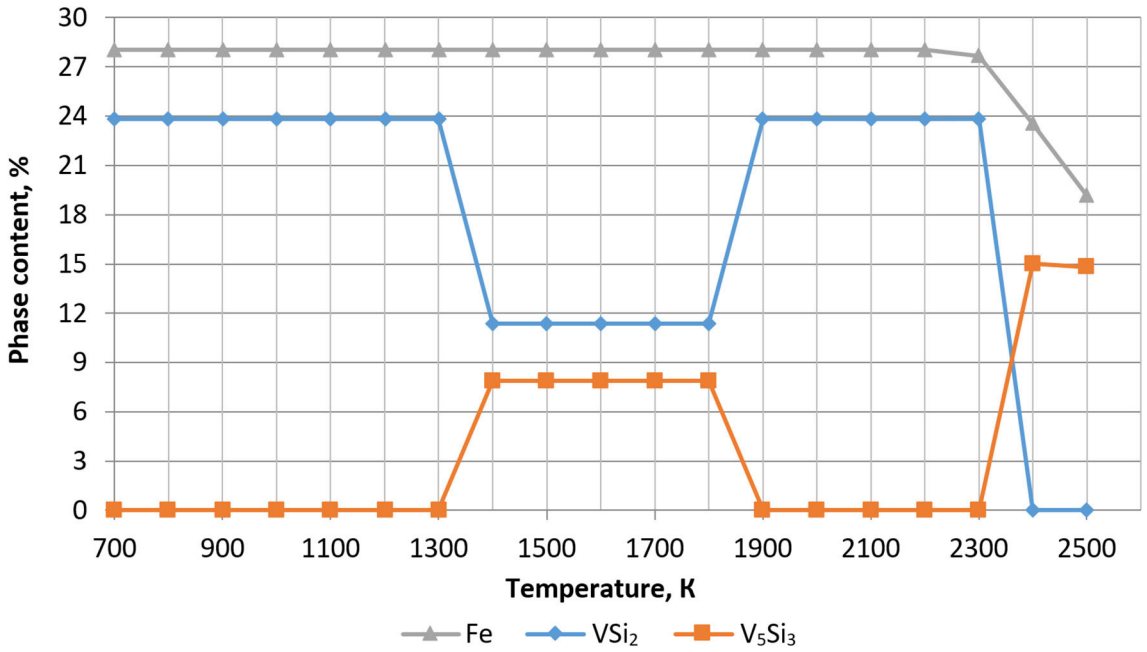

Figure 1 – Dependence of the transition of the main vanadium-containing phases on temperature

with silicon – vanadium silicides V_3Si, V_5Si_3 and VSi_2, of which the most refractory is V_5Si_3, melting at a temperature of 2147 K. Up to 35% Al can be dissolved in vanadium. Chemical compounds V_6Al_8, VAl_3, etc. are incongruent, i.e. melting with decomposition. In relation to iron, vanadium is characterized by complete mutual solubility, both in liquid and solid states.

To analyze the coexistence of phases in a multi-component system during the aluminosilicothermal reduction of vanadium, a triple phase diagram was constructed for the V_2O_5-Al-Si system (in the temperature range from 400 to 1773 K) using the Triangle program. The «Triangle» program allows you to determine the coexisting condensed and gas phases, the results of which are shown in Figures 2-5 and in tables 3-6.

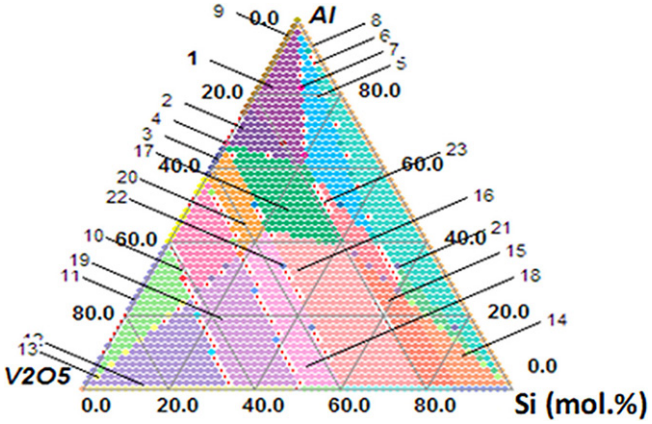

Figure 2 – Three-component phase diagram of V_2O_5-Al-Si at 400 K

Table 3 – Detected phases on the V_2O_5-Al-Si diagram at 400 K

1	Al(c), Al_2O_3(c), V(c), V_5Si_3(c)	13	Al_2SiO_5(c), V_2O_4(c), V_2O_5(c)
2	Al_2O_3(c), V(c), V_5Si_3(c), VO(c)	14	Al_2SiO_5(c), Si(c), SiO_2(c), VSi_2(c);
3	Al_2O_3(c), V_2O_3, VO(c)	15	Al_2SiO_5(c), SiO_2(c), V_5Si_3(c), VSi_2(c);
4	Al_2O_3(c), VO(c)	16	Al_2SiO_5(c), SiO_2(c), V_5Si_3(c), VO(c);
5	Al(c), Al_2O_3(c), V_5Si_3(c), VSi_2(c)	17	Al_2O_3(c), Al_2SiO_5, V_5Si_3(c), VO(c);
6	Al(c), Al_2O_3(c), Si(c), VSi_2(c)	18	Al_2SiO_5(c), SiO_2(c), V_2O_3(c), VO(c);
7	Al(c), Al_2O_3(c), V_5Si_3(c)	19	Al_2SiO_5(c), SiO_2(c), V_2O_3(c), V_2O_4(c);
8	Al(c), Si(c);	20	Al_2O_3(c), Al_2SiO_5(c), V_2O_3(c), VO(c);
9	Al(c), Al_2O_3(c), V(c);	21	Al_2O_3(c), Si(c), VSi_2(c);
10	Al_2O_3(c), Al_2SiO_5, V_2O_3(c), V_2O_4(c)	22	Al_2SiO_5(c), SiO_2(c), VO(c);
11	Al_2O_3(c), V_2O_4(c), V_2O_5(c)	23	Al_2O_3(c), Al_2SiO_5(c), V_5Si_3(c), VSi_2(c).
12	Al_2SiO_5(c), SiO_2(c), V_2O_4(c), V_2O_5(c)	-	-

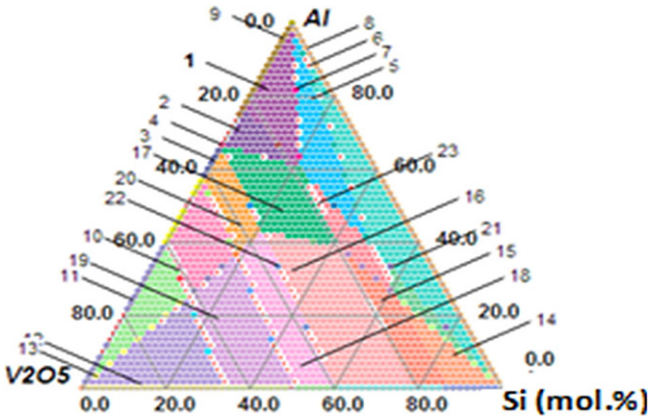

Figure 3 – Three-component phase diagram of V_2O_5-Al-Si at 800 K

Table 4 – Detected phases on the V_2O_5-Al-Si diagram at 800 K			
1	Al(c), Al_2O_3(c), V(c), V_5Si_3(c);	13	Al_2SiO_5(c), V_2O_4(c), V_2O_5(c);
2	Al_2O_3(c), V(c), V_5Si_3(c), VO(c);	14	Al_2SiO_5(c), Si(c), SiO_2(c), VSi_2(c);
3	Al_2O_3(c), V_2O_3, VO(c);	15	Al_2SiO_5(c), SiO_2(c), V_5Si_3(c), VSi_2(c);
4	Al_2O_3(c), VO(c);	16	Al_2SiO_5(c), SiO_2(c), V_5Si_3(c), VO(c);
5	Al(c), Al_2O_3(c), V_5Si_3(c), VSi_2(c);	17	Al_2O_3(c), Al_2SiO_5, V_5Si_3(c), VO(c);
6	Al(c), Al_2O_3(c), Si(c), VSi_2(c);	18	Al_2SiO_5(c), SiO_2(c), V_2O_3(c), VO(c);
7	Al(c), Al_2O_3(c), V_5Si_3(c);	19	Al_2SiO_5(c), SiO_2(c), V_2O_3(c), V_2O_4(c);
8	Al(c), Si(c);	20	Al_2O_3(c), Al_2SiO_5, V_2O_3(c), VO(c);
9	Al(c), Al_2O_3(c), V(c);	21	Al_2O_3(c), Si(c), VSi_2(c);
10	Al_2O_3(c), Al_2SiO_5, V_2O_3(c), V_2O_4(c);	22	Al_2SiO_5(c), SiO_2(c), VO(c);
11	Al_2O_3(c), V_2O_4(c), V_2O_5(c);	23	Al_2O_3(c), Al_2SiO_5(c), V_5Si_3(c), VSi_2(c).
12	Al_2SiO_5(c), SiO_2(c), V_2O_4(c), V_2O_5(c);		

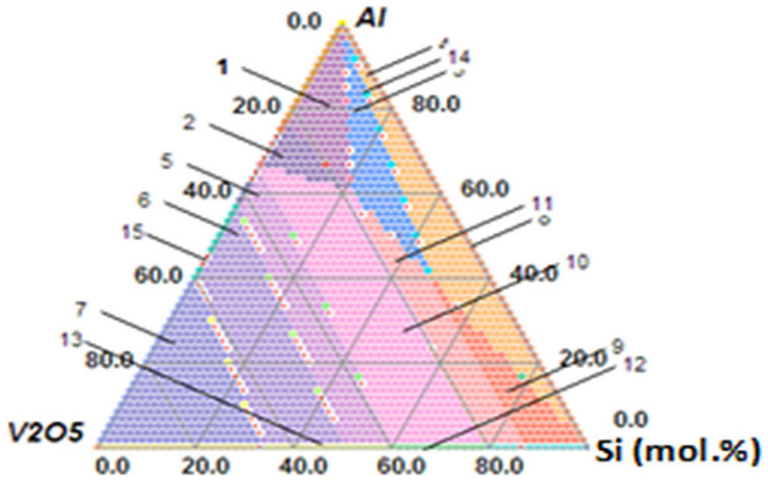

Figure 4 – Three-component phase diagram of V_2O_5-Al-Si at 1073 K

Table 5 – Detected phases on the V_2O_5-Al-Si diagram at 1073 K			
1	Al(c), Al_2O_3(c), V(c), V_5Si_3(c);	9	Al_2O_3(c), Si(c), SiO_2(c), VSi_2(c);
2	Al_2O_3(c), V(c), V_5Si_3(c), VO(c);	10	Al_2O_3(c), SiO_2(c), V_5Si_3(c), VO(c);
3	Al(c), Al_2O_3(c), V_5Si_3(c), VSi_2(c);	11	Al_2O_3(c), SiO_2(c), V_5Si_3(c), VSi_2(c);
4	Al(c), Al_2O_3(c), Si(c), VSi_2(c);	12	SiO_2(c), V_5SiO_2(c), VO(c);
5	Al_2O_3(c), SiO_2(c), V_2O_3(c), VO(c);	13	SiO_2(c), V_2O_3(c), V_2O_4(c);
6	Al_2O_3(c), SiO_2(c), V_2O_3(c), V_2O_4(c);	14	Al(c), Al_2O_3(c), VSi_2(c);
7	Al_2O_3(c), SiO_2(c), V_2O_4(c), V_2O_5(c);	15	Al_2O_3(c), V_2O_3(c), V_2O_4(c).
8	Al(c), Si(c);		

The three-component phase diagram of V_2O_5-Al-Si at different temperatures shows the constant presence of the following coexisting phases: V_5Si_3, VSi_2 and free silicon Si. Thus, the conducted thermodynamic – diagrammatic analysis gives a predictable estimate of the phase composition of the final ferroalloy, namely V_5Si_3, VSi_2 and free silicon Si.

Conclusions

In the work, a complete thermodynamic modeling of metallurgical processes in the V-Fe-Si-Al-Ca-O system was carried out using the HSC Chemistry software package. An analysis was also made of the coexistence of phases in the multicomponent V_2O_5-Al-Si system during the aluminosilicothermal reduc-

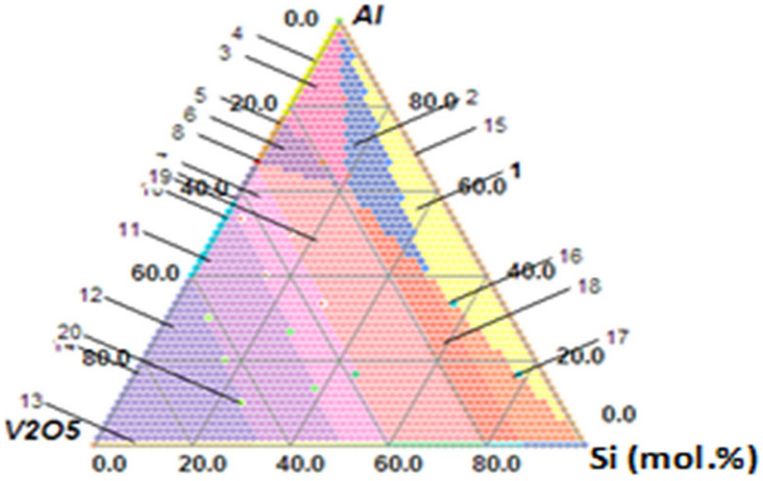

Figure 5 – Three-component phase diagram of V_2O_5-Al-Si at 1773 K

Table 6 – Detected phases on the V_2O_5-Al-Si diagram at 1773 K			
1	Al(c), Al_2O_3(c), Si(c), VSi_2(c);	11	Al_2O_3(c), SiO_2(c), V_2O_3(c), V_2O_4(c);
2	Al(c), Al_2O_3(c), V_5Si_3(c), VSi_2(c);	12	Al_2O_3(c), SiO_2(c), V_2O_4(c), V_2O_5(c);
3	Al(c), Al_2O_3(c), V(c), V_5Si_3(c);	13	SiO_2(c), V_2O_4(c), V_2O_5(c);
4	Al(c), Al_2O_3(c), V(c);	14	Al_2O_3(c), V_2O_4(c), V_2O_5(c);
5	Al_2O_3(c), V(c), VO(c);	15	Al(c), Si(c);
6	Al_2O_3(c), V(c), V_5Si_3(c), VO(c);	16	Al_2O_3(c), Si(c), VSi_2(c);
7	Al_2O_3(c), SiO_2(c), V_2O_3(c), VO(c);	17	Al_2O_3(c), Si(c), SiO_2(c), VSi_2(c);
8	Al_2O_3(c), VO(c);	18	Al_2O_3(c), SiO_2(c), V_5Si_3(c), VSi_2(c);
9	Al_2O_3(c), V_2O_3(c), VO(c);	19	Al_2O_3(c), SiO_2(c), V_5Si_3(c), VO(c);
10	Al_2O_3(c), V_2O_3(c), V_2O_4(c);	20	Al_2O_3(c), SiO_2(c), V_2O_4(c).

tion of vanadium using the Triangle program. As a result, the processes of aluminosilicothermal reduction of vanadium in the process of smelting a vanadium alloy using silicon-aluminum reducing agents were studied.

It has been established that the optimal temperature ranges for smelting a vanadium alloy is 1650-1850 K, since silicon and aluminum waste increases with increasing temperature. For high extraction of vanadium into the alloy, the slag must have a basicity of 1,75-1,95. Thus, the thermodynamic calculations carried out made it possible to fully consider all the physicochemical processes that occur during the smelting of a vanadium alloy.

Thus, the conducted studies give a predictable estimate of the phase composition of the final ferroalloy, namely V_5Si_3, VSi_2 and free silicon Si.

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Кремний алюминий тотықсыздандырғышын қолдана отырып, ванадий қорытпасын балқыту процесін термодинамикалық модельдеу

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***Аңдатпа.** Мақала ванадий қорытпасын алюмосиликотермиялық жолмен алудың теориялық аспектісін зерттеуге арналған. Шихта материалдары ретінде ванадий пентаоксиді және конвертер ванадий қожы зерттелді. Тотықсыздандырғыш ретінде ферроалюмосиликокальций, ферросиликоалюминий және кальций бар алюминий силикомарганец сияқты кремний алюминий ферроқорытпалары қолданылады. Құрамында ванадий бар бастапқы материалдар мен кремний алюминий тотықсыздандырғыштарының химиялық құрамы спектрлік талдау әдісімен зерттелді. Ванадийдің алюминий силикотермиялық тотықсыздануын V-Fe-Si-Al-Ca-O жүйесінде талдау үшін HSC Chemistry бағдарламалық кешені арқылы компьютерлік жүйеде жүзеге асырылатын металлургиялық процестерді толық термодинамикалық модельдеу әдісі қолданылды. Тотықсыздандырғыштың стехиометриялық ағынымен ванадий лигатурасын балқыту шихтасының нақты құрамы үшін термодинамикалық тепе-теңдік параметрлерін анықтауға арналған бастапқы деректер ретінде. Ванадийдің алюминий силикотермиялық тотықсыздануында көп компонентті жүйеде фазалардың қатар өмір сүруін талдау үшін «Triangle» бағдарламасы әртүрлі изотермияларда V_2O_5-Al-Si жүйесі үшін үш фазалық диаграмма жасады. Нәтижесінде кремний алюминий тотықсыздандырғыштарын қолдана отырып, ванадий қорытпасын балқыту процесінде ванадийдің алюминий силикотермиялық тотықсыздану процестері зерттелді. 1650-1850 К диапазонында ванадий қорытпасын балқытудың оңтайлы температуралық аралығын орнатты. Сондай-ақ, термодинамикалық зерттеулердің нәтижелері V_5Si_3, VSi_2 және бос кремний Si түрінде қорытпаның соңғы фазалық құрамын болжауға мүмкіндік берді.*

***Кілт сөздер:** ванадий, ферросиликоалюминий, кремний алюминий тотықсыздандырғыш, металлотермия, процесті модельдеу, ванадий лигатурасы.*

Термодинамическое моделирование процесса выплавки ванадиевого сплава с использованием кремнеалюминиевого восстановителя

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***Аннотация.** Статья посвящена изучению теоретического аспекта получения ванадиевого сплава алюмосиликотермическим способом. В качестве шихтовых материалов изучены пентаоксид ванадия и конвертерный ванадиевый шлак. В роли восстановителя будут использованы кремнеалюминиевые ферросплавы, такие как ферроалюмосиликокальций, ферросиликоалюминий и алюмосиликомарганец с кальцием. Химический состав исходных ванадийсодержащих материалов и кремнеалюминиевых восстановителей были изучены*

спектральным методом анализа. Для анализа в системе V-Fe-Si-Al-Ca-O алюмосиликотермического восстановления ванадия был использован метод полного термодинамического моделирования металлургических процессов, реализованного в компьютерной системе с помощью программного комплекса HSC Chemistry. В качестве исходных данных для определения параметров термодинамического равновесия для реального состава шихты выплавки ванадиевой лигатуры с стехиометрическим расходом восстановителя. Для анализа сосуществования фаз в многокомпонентной системе при алюмосиликотермическом восстановлении ванадия с помощью программы «Triangle» построили тройную фазовую диаграмму для системы V_2O_5-Al-Si при разных изотермиях. В результате были изучены процессы алюмосиликотермического восстановления ванадия в процессе выплавки ванадиевого сплава с использованием кремнеалюминиевых восстановителей. Установили оптимальный температурный интервал выплавки ванадиевого сплава в диапазоне 1650-1850 К. Также полученные результаты термодинамических исследований позволили спрогнозировать конечный фазовый состав сплава в виде V_5Si_3, VSi_2 и свободного кремния Si.

Ключевые слова: *ванадий, ферросиликоалюминий, кремниеалюминиевый восстановитель, металлотермия, моделирование процесса, ванадиевая лигатура.*

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