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BORON DISTRIBUTION BETWEEN METAL AND SLAG DURING MELTING OF METALIZED SIDERITE CONCENTRATE IN ELECTRIC FURNACE

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Abstract. The Ural metallurgical enterprises compensate for the deficit of iron ore raw materials by supplying materials from Central Russia, the Kola Peninsula and Kazakhstan. Carbonate iron ores (siderites) of the Bakalskoye deposit are poor ores and, despite large reserves (about 1 billion tons), are not in great demand among metallurgists due to their low quality (low iron content and high magnesium content). The prospects for developing the Bakalskoye deposit depend on the availability of new technologies for processing siderites. There is a technology for processing poor iron ores using the coke-free metallurgy method, including reducing roasting in a rotary kiln, grinding and magnetic separation to obtain a highly metallized product suitable for steelmaking. Laboratory studies and industrial tests confirmed its suitability for processing siderites. A modernized technology for processing siderites is proposed, in which the operations of grinding and magnetic separation are excluded, and the product of reducing roasting in a rotary kiln is loaded hot into an electric furnace for separating melting. The process is carried out in the presence of colemanite containing boric anhydride to obtain liquid slag. During melting, part of the boron passes into the metal melt. By means of thermodynamic modeling, an assessment of the distribution of boron between the metal and slag as a result of separating melting is carried out. With a content of 5 and 10 % of colemanite in the charge, depending on the proportion of carbon, up to 60 % of boron passes into the metal. Such metal can be used as a ligature for obtaining boron-containing steel or cast iron. When bubbling the metal melt with oxygen, boron content can be reduced to 0.0001 %.

Keywords: siderites, metallization, separation melting, colemanite, boron, distribution

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РАСПРЕДЕЛЕНИЕ БОРА МЕЖДУ МЕТАЛЛОМ И ШЛАКОМ В ПРОЦЕССЕ ПЛАВКИ МЕТАЛЛИЗОВАННОГО СИДЕРИТОВОГО КОНЦЕНТРАТА В ЭЛЕКТРОПЕЧИ

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Аннотация. Металлургические предприятия Урала компенсируют дефицит железорудного сырья поставкой материалов из Центральной России, Кольского полуострова и Казахстана. Карбонатные железные руды (сидериты) Бакальского месторождения относятся к бедным рудам и, несмотря на большие запасы (около 1 млрд т), не пользуются широким спросом у металлургов ввиду низкого качества (низкое содержание железа и высокое – магния). Перспективы освоения Бакальского месторождения зависят от наличия новых технологий переработки сидеритов. Существует технология переработки бедных железных руд методом бескоксовой металлургии, включающая восстановительный обжиг во вращающейся печи, измельчение и магнитную сепарацию с получением высокометаллизированного продукта, пригодного для сталеплавления производства. Лабораторные исследования и промышленные испытания подтвердили ее пригодность для переработки сидеритов. Предложена модернизированная технология переработки сидеритов, в которой операции измельчения и магнитной сепарации исключены, а продукт восстановительного обжига во вращающейся печи в горячем виде загружается в электропечь для проведения разделительной плавки. Процесс осуществляют в присутствии колеманита, содержащего борный ангидрид, для получения жидкого шлака. В ходе плавки часть бора переходит в металлический расплав. Посредством термодинамического моделирования проведена оценка распределения бора между металлом и шлаком в результате разделительной плавки. Показано, что при содержании в шихте 5 и 10 % колеманита в зависимости от доли углерода в металл переходит до 60 % бора. Такой металл может быть использован в качестве лигатуры для получения борсодержащей стали или чугуна. При барботаже металлического расплава кислородом содержание бора может быть снижено до 0,0001 %.

Ключевые слова: сидериты, металлизация, разделительная плавка, колеманит, бор, распределение

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INTRODUCTION

The reserves of carbonate iron (siderite) ores at the Bakalskoye deposit located near the town of Bakal in the Chelyabinsk Region are estimated at about 1 billion tons [1; 2]. However, the Ural metallurgical enterprises compensate for the deficit of iron ore raw materials by supplying materials from Central Russia (Mikhailovsky Mining and Processing Plant, Lebedinsky Mining and Processing Plant), the Kola Peninsula (Kostomuksha Mining and Processing Plant), and Kazakhstan (Sokolov–Sarbai Mining and Processing Plant) [3; 4]. Bakal siderites are classified as poor iron ores, containing less than 35 % iron and a large proportion of magnesium oxide. The low quality of the ore limits its demand; therefore, the production of siderite ore is much lower than the potential allowed by the mining and geological conditions. For example, in 2006, ore production amounted to 1.7 million tons, or less than 3 % of the total output in the Ural Federal District [5]. The only industrially implemented method for processing Bakal siderites is blast-furnace melting [1; 2]. Existing beneficia-

tion methods [6; 7] make it possible to increase the iron content in the obtained concentrate. However, the entire amount of magnesium oxide remains in the concentrate, which restricts its use in blast-furnace smelting to the role of a charge additive and only in quantities ensuring that the magnesium oxide content in the final slag does not exceed 20 %. Therefore, the prospects for developing the Bakalskoye deposit depend on the availability of new technologies for processing siderites.

To expand the range of siderite applications and increase their processing efficiency, a pyrometallurgical beneficiation technology has been developed [2; 8; 9]. It includes reducing roasting of siderites in a rotary kiln using a solid reducing agent, followed by magnetic separation to obtain a highly metallized product. The resulting concentrate contains less than 5 % of gangue and, when briquetted, can serve as a feed material for an electric-arc furnace.

A process was proposed to reduce costs by eliminating grinding, magnetic separation, drying, and briquetting, in which hot metallized lump concentrate is fed directly

from the rotary kiln to the electric furnace instead of using briquettes [10]. Comparative analysis of the two melting routes showed that the specific power consumption for melting metallized briquettes loaded into the furnace at 25 °C amounts to 773 kW·h per ton of metal, whereas melting hot metallized lump concentrate at 1000 °C requires 725 kW·h per ton of metal [11].

It is well known that the addition of boric anhydride to slag lowers its melting temperature [12; 13]. Since the gangue in the metallized concentrate contains a large amount of magnesium oxide, boron oxide in the form of calcined colemanite is added to the charge to produce a liquid, free-flowing slag with a low melting temperature. This provides a slag viscosity of less than 0.4 Pa·s at 1600 °C and a melting temperature of 1300 – 1500 °C, depending on the colemanite content in the charge and the SiO₂/MgO ratio in the slag [14].

When boron oxide is present in the steelmaking slag, boron can pass into the metal [15]. This study uses thermodynamic modeling to evaluate how boron partitions between metal and slag during separation melting of metallized siderite concentrate in the presence of colemanite.

MATERIALS AND METHODS

Metallurgical processes occur at high temperatures and proceed at high rates; therefore, it can be assumed with sufficient confidence that during the melting of siderites the system reaches a state close to equilibrium. Consequently, to evaluate boron distribution between the metal and slag, the thermodynamic modeling (TDM)

method was applied. This method is widely used to calculate multicomponent and multiphase systems when solving both theoretical and applied problems related to improving metallurgical technologies. Modeling of equilibrium states in the Fe–Ca–Si–Mg–Al–Mn–C–O system was performed using the IVTANTHERMO software package. Numerical values of the total enthalpy of the resulting equilibrium compositions were obtained using HSC Chemistry 6.12.

In siderite ores of the Bakal ore field, subjected to preliminary gravitational or X-ray radiometric beneficiation [16] for shale removal, the contents of most components differ only slightly. The main difference lies in the SiO₂/MgO ratio [17; 18]. Therefore, a concentrate of the following composition (wt. %) was taken as the initial material for the study: 33.08 Fe_{total}; 38.94 FeO; 2.27 Fe₂O₃; 1.41 CaO; 0.66 SiO₂; 11.22 MgO; 0.38 Al₂O₃; 0.85 Mn. The SiO₂/MgO ratio was adjusted by adding quartz. The final compositions of the model concentrates are presented in Table 1.

The masses (m_c) and compositions of the metallized siderite concentrates (MSC) with a metallization degree of $\varphi = 95$ %, obtained from 1 kg of initial concentrate, are given in Table 2.

As a fluxing additive used to liquefy the slag, calcined colemanite [19] with the following composition (wt. %) was employed: 8 SiO₂; 34 CaO; 4 MgO; and 54 B₂O₃. In practice, the solid reducing agent (coke breeze) with a particle size of 3 – 5 mm is separated from MSC lumps sized 10 – 60 mm by screening. However, some portion of the reducing agent may enter the electric furnace

Table 1. Compositions of the initial model concentrates used in thermodynamic modeling method as part of the working fluid

SiO ₂ /MgO ratio	Content, wt. %								
	Fe _{tot}	FeO	Fe ₂ O ₃	CaO	SiO ₂	MgO	Al ₂ O ₃	MnO	LOI
0.75	31.34	38.30	2.23	1.39	8.28	11.04	0.37	1.08	37.31
1.00	30.50	37.28	2.17	1.35	10.74	10.74	0.36	1.05	36.31
1.25	29.70	36.30	2.12	1.31	13.07	10.46	0.35	1.02	35.36

Table 2. Masses and compositions of MSCs with a metallization degree of 95 %

SiO ₂ /MgO ratio	m_c , kg	Content, wt. %							
		Fe _{tot}	Fe _{met}	FeO	CaO	SiO ₂	MgO	Al ₂ O ₃	MnO
0.75	539.43	58.10	55.19	3.73	2.57	15.35	20.46	0.69	2.00
1.00	551.79	55.27	52.51	3.55	2.45	19.46	19.46	0.66	1.90
1.25	563.51	52.71	50.07	3.39	2.33	23.20	18.56	0.63	1.82

together with the concentrate; therefore, it was assumed in the calculations that the electric-furnace charge contains carbon.

Thus, the model systems used for calculating phase equilibria consisted of MSC (Table 2), colemanite, and carbon, taken in amounts of 0, 5, and 10 wt. % and 0, 2, 4, and 6 wt. %, respectively, relative to the mass of metallized concentrate. The modeling was carried out at a temperature of 1600 °C, pressure of 0.1 MPa, and under the assumption that the oxide and metallic phases behave as ideal solutions.

RESULTS AND DISCUSSION

Equilibrium calculations for the MSC–carbon system show that in the presence of carbon, the amounts of iron, manganese, and silicon oxides in the oxide phase

decrease (Fig. 1), and these elements pass into the metal together with residual carbon (Fig. 2).

At a temperature of 1600 °C, corresponding to the melting temperature, the iron-based metal exists in a liquid state. To estimate the crystallization onset temperature of the slags obtained from equilibrium calculations, two methods were applied.

The first method was based on the results reported in [14], which analyzed the effect of the SiO_2/MgO ratio and the amount of colemanite in the charge on the transition temperature of slags from their high-temperature stability region to the low-temperature region, close to the liquidus temperature. The dependence is described by the following empirical equation:

$$T = 1801.7 - 199.5x - 28.6x^2 - 7.19y + 0.03y^2 - 4.0xy,$$

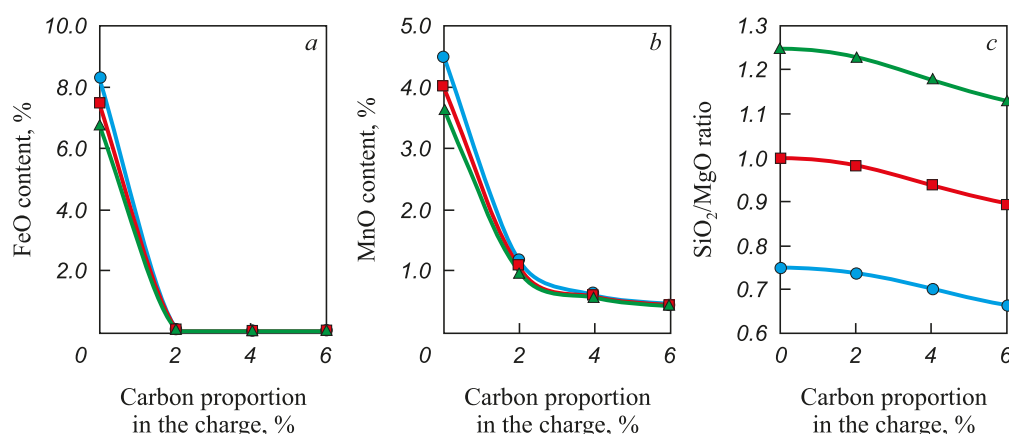


Fig. 1. Dependence of content of FeO (a), MnO (b), SiO_2/MgO ratio (c) in the oxide phase on carbon proportion in the charge: ● – 0.75; ■ – 1.00; ▲ – 1.25

Рис. 1. Зависимость содержания в оксидной фазе FeO (a), MnO (b), соотношения SiO_2/MgO (c) от доли углерода в шихте: ● – 0,75; ■ – 1,00; ▲ – 1,25

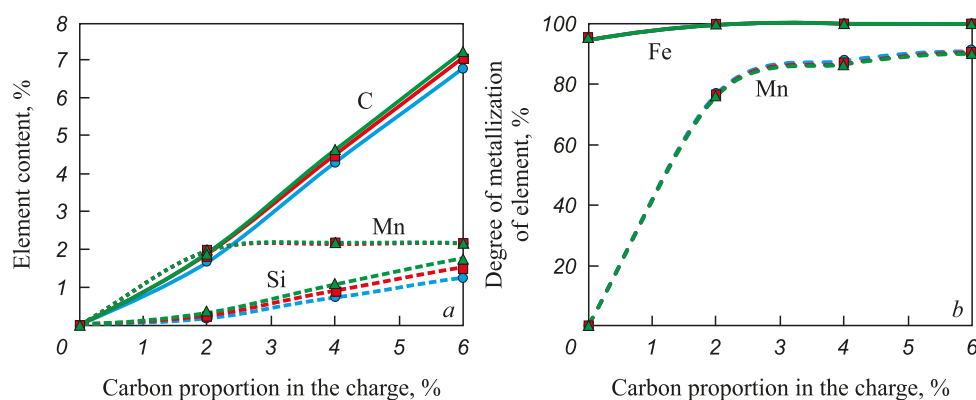


Fig. 2. Dependence of content of carbon, manganese and silicon in the metal (a) and metallization degree of iron and manganese (b) on carbon proportion in the charge: ● – 0.75; ■ – 1.00; ▲ – 1.25

Рис. 2. Зависимость содержания углерода, марганца и кремния в металле (a) и степени металлизации железа и марганца (b) от доли углерода в шихте: ● – 0,75; ■ – 1,00; ▲ – 1,25

where x is the SiO_2/MgO ratio (units); y is the colemanite content (wt. %).

It was established that in the absence of colemanite in the charge, the transition temperature decreases from 1640 to 1510 °C as the SiO_2/MgO ratio increases from 0.75 to 1.25.

A similar result was obtained using the second method described in [20], according to which the temperatures of the beginning (t_s) and end (t_l) of melting (solidus and liquidus, respectively) were determined from inflection points on the total entropy and enthalpy curves characterizing the system state. This approach was used as the basis for liquidus temperature calculations of the slag systems considered in this study.

The slags used as input were those whose compositions were determined from the equilibrium calculations

in the MSC–carbon system. The results showed that in slags without carbon and with $\text{SiO}_2/\text{MgO} = 0.75 - 1.25$, t_l lies in the range of 1450 – 1600 °C (Fig. 3). In slags formed as a result of the interaction between MSC and carbon, a decrease in FeO content – caused by an increase in the carbon proportion – does not lead to a further increase in t_l (Fig. 4).

Thus, the calculated crystallization onset temperatures of oxide phases formed during the interaction of MSC with carbon indicated that at 1600 °C, corresponding to the electric furnace temperature, these phases remain heterogeneous. This confirms the need for a flux additive based on boric anhydride to liquefy the slag, for which colemanite was chosen.

Equilibrium calculations for the MSC–colemanite–carbon system showed the following. The addition of carbon increases the manganese and silicon content in

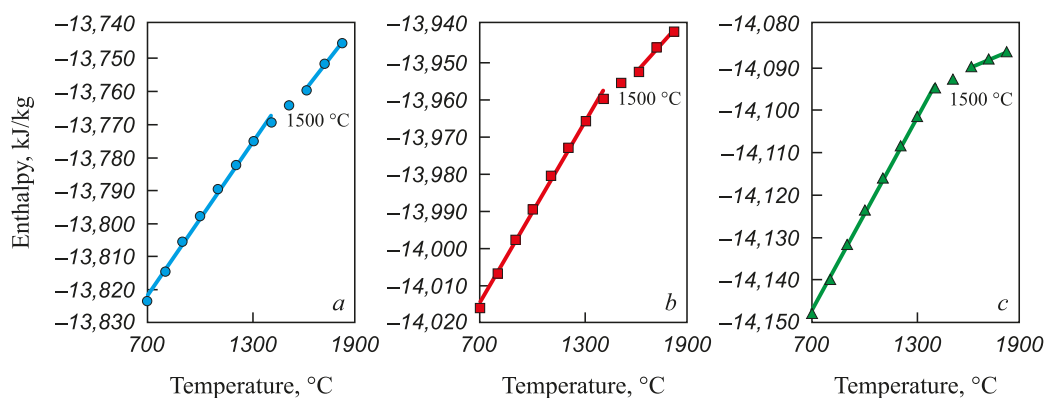


Fig. 3. Dependence of the system total enthalpy on temperature for slag systems not containing colemanite and carbon, with a change in SiO_2/MgO ratio: $a - 0.75$; $b - 1.00$; $c - 1.25$

Рис. 3. Зависимость полной энтальпии системы от температуры для шлаковых систем, не содержащих в своем составе колеманит и углерод, при изменении соотношения SiO_2/MgO : $a - 0,75$; $b - 1,00$; $c - 1,25$

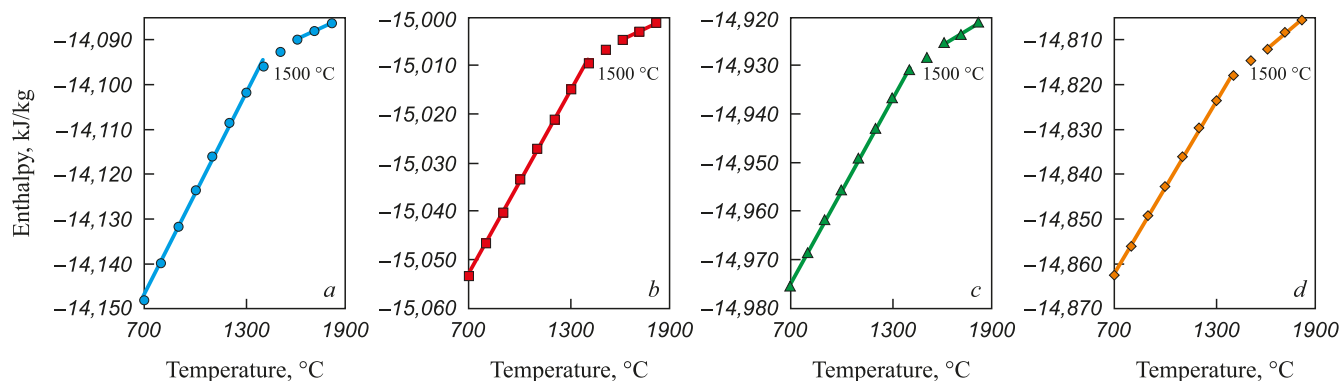


Fig. 4. Dependence of the system total enthalpy on temperature for slag systems calculated at $\text{SiO}_2/\text{MgO} = 1.25$ and not containing colemanite, with a change in carbon proportion, %: $a - 0$; $b - 2$; $c - 4$; $d - 6$

Рис. 4. Зависимость полной энтальпии системы от температуры для шлаковых систем, рассчитанных при соотношении $\text{SiO}_2/\text{MgO} = 1,25$ и не содержащих в своем составе колеманит, при изменении доли углерода, %: $a - 0$; $b - 2$; $c - 4$; $d - 6$

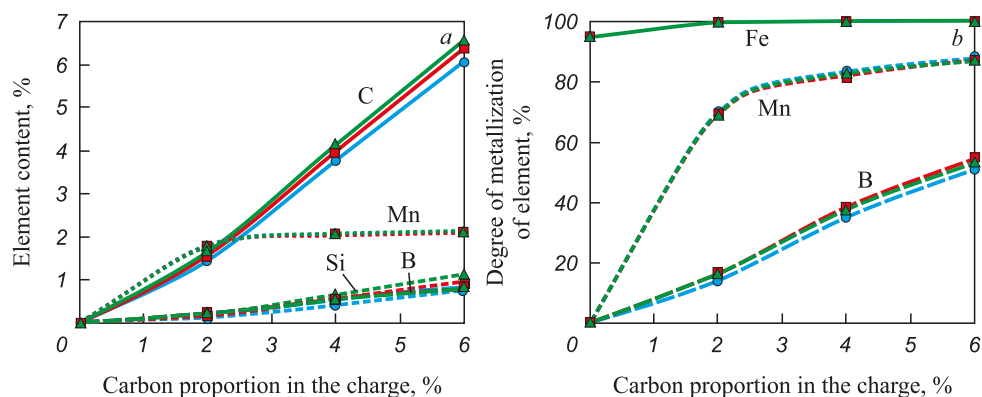


Fig. 5. Dependence of content of carbon, manganese, boron and silicon in the metal (a) and metallization degree of iron, manganese and boron (b) on carbon proportion in the charge with addition of 5 % of colemanite to the metallized siderite concentrates (MSC):

● – 0.75; ■ – 1.00; ▲ – 1.25

Рис. 5. Зависимость содержания углерода, марганца, бора и кремния в металле (a) и степени металлизации железа, марганца и бора (b) от доли углерода в шихте при добавке к МСК 5 % колеманита:

● – 0,75; ■ – 1,00; ▲ – 1,25

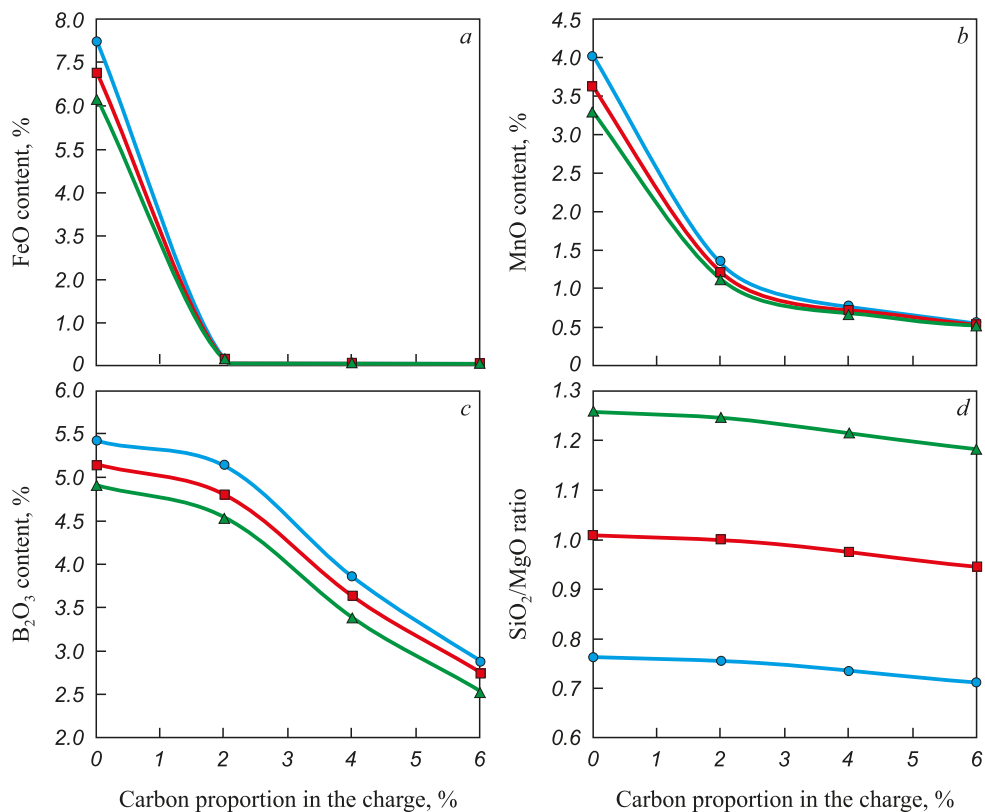


Fig. 6. Dependence of content in the oxide phase of FeO (a), MnO (b), B₂O₃ (c), and ratio SiO₂/MgO (d) on carbon proportion in the charge with addition of 5 % of colemanite to MSC:

● – 0.75; ■ – 1.00; ▲ – 1.25

Рис. 6. Зависимость содержания в оксидной фазе FeO (a), MnO (b), B₂O₃ (c) и соотношения SiO₂/MgO (d) от доли углерода в шихте при добавке к МСК 5 % колеманита:

● – 0,75; ■ – 1,00; ▲ – 1,25

the metal and decreases the amount of iron, manganese, and silicon oxides in the slag phase, similar to the MSC–carbon system (Figs. 5 – 8). In addition, boron passes into the metal, accompanied by a decrease in boron oxide content in the slag.

In the absence of carbon, the oxide phase compositions are close to those of the slags investigated in [14], where, as the SiO₂/MgO ratio increased from 0.75 to 1.25, t_1 decreased from 1520 to 1370 °C, and the viscosity of all slags was below 0.3 Pa·s.

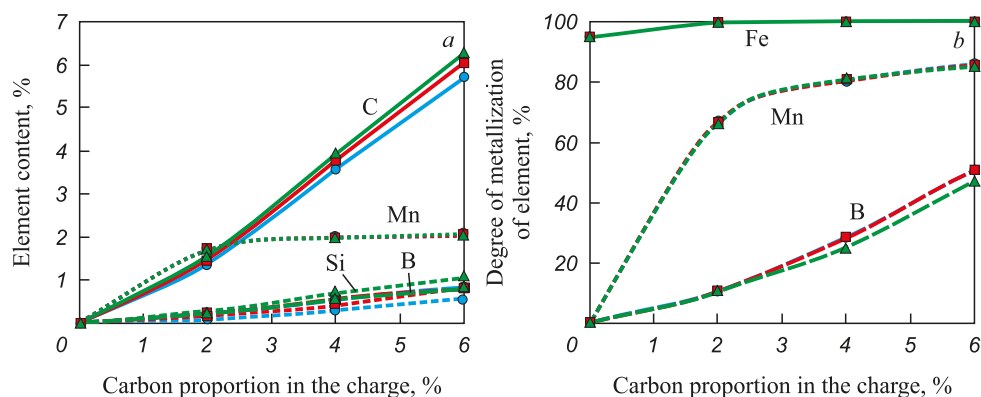


Fig. 7. Dependence of content of carbon, manganese, boron and silicon in the metal (a) and metallization degree of iron, manganese and boron (b) on carbon proportion in the charge with addition of 10 % of colemanite to MSC:

● – 0.75; ■ – 1.00; ▲ – 1.25

Рис. 7. Зависимость содержания углерода, марганца, бора и кремния в металле (a) и степени металлизации железа, марганца и бора (b) от доли углерода в шихте при добавке к МСК 10 % колеманита:

● – 0,75; ■ – 1,00; ▲ – 1,25

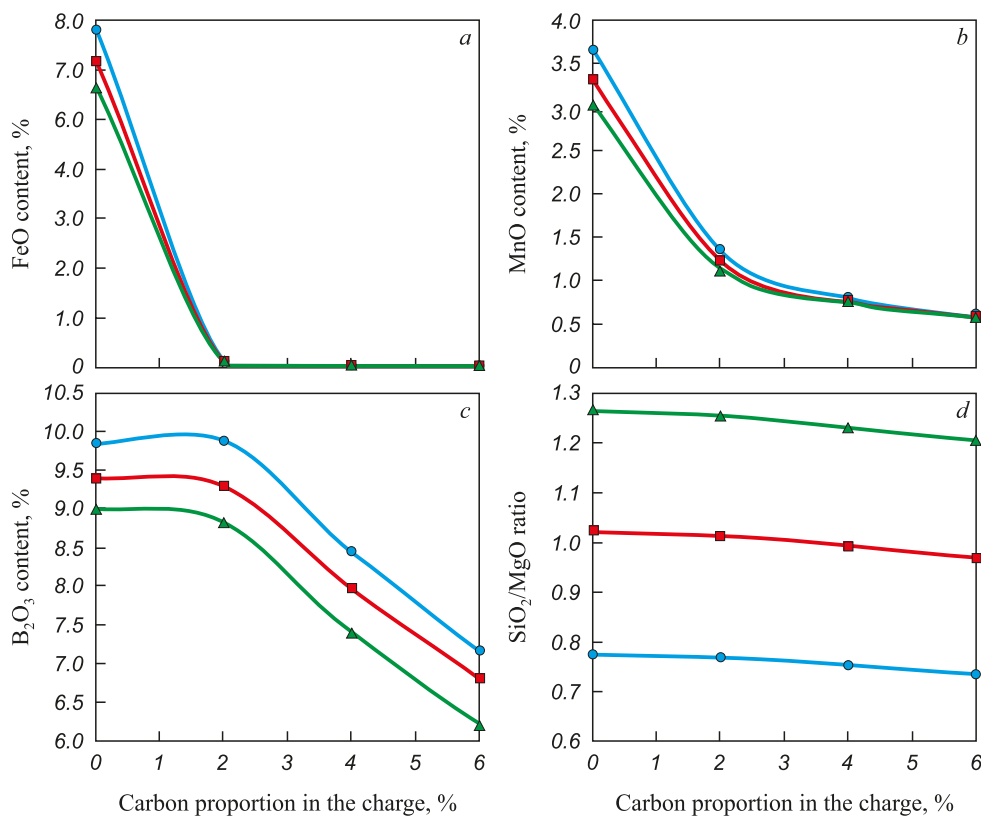


Fig. 8. Dependence of content in the oxide phase of FeO (a), MnO (b), B₂O₃ (c), and the ratio SiO₂/MgO (d) on carbon proportion in the charge with addition of 10 % of colemanite to MSC:

● – 0.75; ■ – 1.00; ▲ – 1.25

Рис. 8. Зависимость содержания в оксидной фазе FeO (a), MnO (b), B₂O₃ (c) и соотношения SiO₂/MgO (d) от доли углерода в шихте при добавке к МСК 10 % колеманита:

● – 0,75; ■ – 1,00; ▲ – 1,25

Evaluation of phase transition temperatures in the MSC–colemanite–carbon systems (Fig. 9) indicates a decrease in the liquidus temperature from 1500 to 1400 °C when 5 – 10 wt. % colemanite is added to the charge. This suggests that separation mel-

ting of metallized siderite concentrate in the presence of colemanite is feasible even when carbon is present in the charge. According to theoretical calculations, the slag in this case will have a sufficiently low melting temperature, allowing it to be removed from the furnace after

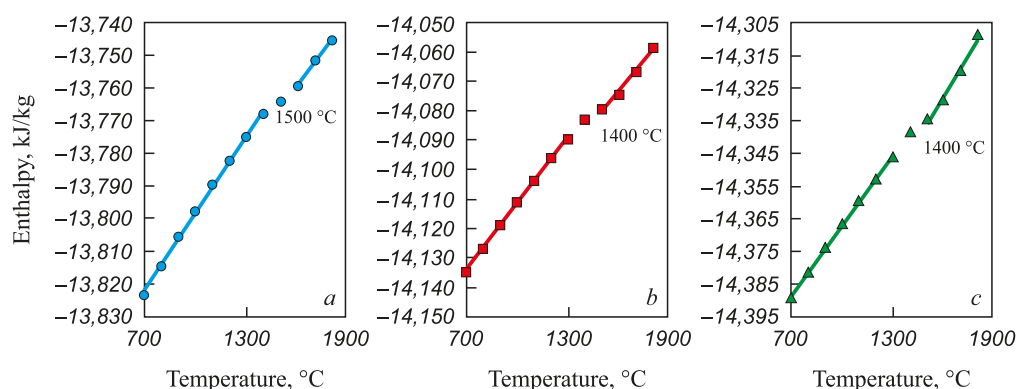


Fig. 9. Dependence of the system total enthalpy on temperature for slag systems calculated at $\text{SiO}_2/\text{MgO} = 0.75$ and not containing colemanite, with a change in carbon proportion, %:

a – 0; b – 5; c – 10

Рис. 9. Зависимость полной энтальпии системы от температуры для шлаковых систем, рассчитанных при соотношении $\text{SiO}_2/\text{MgO} = 0,75$ и не содержащих в своем составе углерод, при изменении соотношения колеманита, %:

a – 0; b – 5; c – 10

separation melting from the metal at 1600 °C. Depending on the carbon content in the charge, the metal will contain manganese, silicon, and boron.

Small additions of boron are known to improve the quality of cast iron [21; 22], steel [23 – 25], and surfacing materials [26]. They significantly refine the grain structure. Strengthening of grain boundaries by borides enhances high-temperature strength, as well as increased hardness and wear resistance, and sharply improves hardenability compared to similar steels without boron. Microadditions of boron (about 0.01 % in cast irons and 0.001 – 0.003 % in steels) can replace alloying elements such as nickel, chromium, molybdenum, and manganese in amounts 300 – 400 times greater than that of boron, while maintaining metal quality.

In the proposed technology, the amount of boron transferred to the metal is significantly higher than that required for producing quality cast iron or steel. Therefore, the resulting metal can be used as a master alloy, or its boron content should be substantially reduced, for example, by bubbling oxidation with oxygen.

To assess the feasibility of bubbling oxidation, additional thermodynamic modeling was performed using the methodology previously applied for modeling bubbling reduction of metals from oxide melts [27 – 29]. The initial model system consisted of metal with the following composition (%): 96.1 Fe; 1.7 C; 0.20 Si; 1.80 Mn; and 0.26 B (1 kg). The gas phase was oxygen, supplied in an amount of 7.3 dm³ per kilogram of metal. Modeling was carried out at 1600 °C and 1 atm, assuming the melt to be an ideal solution.

The calculations were performed in the following sequence:

– input of initial data on the amount of metal melt and oxygen;

– equilibrium calculation of the system using thermodynamic modeling methods;

– recording equilibrium compositions and the amounts of components in the oxide and metallic melts, as well as in the gas phase;

– performing the next cycle, in which the composition of the metallic melt obtained in the previous step was taken as the initial state, while the slag was considered removed from the system and not accounted for; the gas composition and amount remained unchanged;

– repeating the cycles until the amount of oxidized components in the melt decreased to the specified level.

Fig. 10 shows the change in metal composition depending on oxygen consumption.

As a result of bubbling oxidation of the metal, the iron content increases to 99.86 %, the manganese content decreases to 0.13 %, silicon is completely oxidized, and boron content drops to 0.000002 %. From 1 kg of the ini-

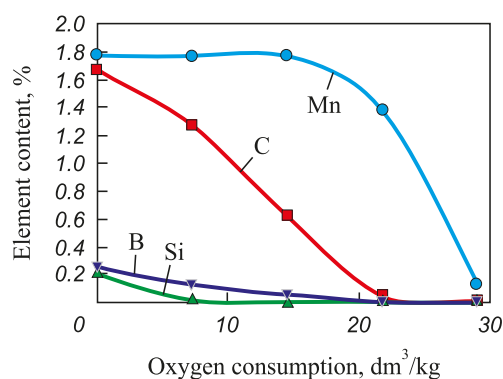


Fig. 10. Dependence of the components content in the metal on oxygen consumption

Рис. 10. Зависимость содержания компонентов в металле от расхода кислорода

tial metal, 940 g of final metal and 201 g of slag are formed, with the slag composition (wt. %) as follows: 46.7 FeO; 6.7 SiO₂; 34.3 MnO; 12.4 B₂O₃. This slag can be used together with colemanite as an additional source of boron.

CONCLUSIONS

Thermodynamic modeling of the separation melting of metallized siderite concentrate with colemanite in the presence of carbon made it possible to describe the distribution of boron between the metal and slag depending on the proportions of colemanite and carbon in the system. It is shown that adding 5 – 10 % colemanite to the charge reduces the slag melting temperature to 1400 °C, enabling efficient extraction of both metal and slag from the furnace at 1600 °C. With an increase in carbon content in the charge up to 6 %, a linear dependence of the degree of boron transfer to the metal was observed, reaching 60 %. The resulting boron-containing metal can be used as a master alloy for microalloying cast irons and steels, or as a semi-finished metal product suitable for secondary (ladle) treatment after preliminary oxygen bubbling. The slag produced during bubbling can be reused as an additional boron-bearing component of the charge. The results of the calculations can serve as a basis for planning and conducting experimental studies on the processes of separation melting of metallized siderite concentrates.

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