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RATIONAL APPLICATION OF HIGH QUALITY MANGANESE CONCENTRATE

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Abstract. The article presents the results of theoretical and experimental studies of manganese reduction processes from oxides of high-quality manganese concentrate obtained by hydrometallurgical enrichment of ferromanganese ores, as well as, from marokite (product of thermal synthesis of concentrate and dolomite) with carbon and silicon. The method of thermodynamic modeling with TERRA software complex determined the optimal temperatures and consumption of reducing agents that ensure the complete reduction of manganese. It was found that any of the above-mentioned reducing agents, or a combination thereof in certain ratios, can be utilized as a reducing agent when using oxide manganese-containing materials for steel treatment. The results of experimental studies made it possible to develop technology for the production of marokite-manganite concentrate and monophase synthetic material (CaMnO_3). They can be obtained using the technology developed by the authors, which includes mechanical and thermal treatment of a mixture of high-quality manganese concentrate and calcined dolomite or lime. Marokite-manganite concentrate is useful for alloying steel with manganese when it is smelted in an electric furnace or in a ladle furnace unit, and a monophasic synthetic material is efficient for the production of metal manganese. Based on the results of thermodynamic calculations and experimental studies, technological parameters for processing steel with marokite-manganite concentrate in an electric furnace and a ladle furnace unit are proposed. Monophasic synthetic material CaMnO_3 should be used as the charge component for the production of metal manganese by the out-of-furnace aluminum thermal treatment, which will increase the thermality of the process, as well as the extraction of manganese at the level of 90 %. The results of experimental studies were obtained using modern research methods with laboratory and analytical equipment, as well as statistical processing methods.

Keywords: high-quality manganese concentrate, marokite, monophase synthetic material, thermochemical synthesis, thermodynamic reduction, reducing agents, steel treatment, metal manganese

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РАЦИОНАЛЬНЫЕ ВАРИАНТЫ ПРИМЕНЕНИЯ ВЫСОКОКАЧЕСТВЕННОГО МАРГАНЦЕВОГО КОНЦЕНТРАТА

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Аннотация. В работе представлены результаты теоретических и экспериментальных исследований процессов восстановления марганца из оксидов высококачественного марганцевого концентрата, полученного в результате гидрометаллургического обогащения железомарганцевых руд, а также из марокита (продукта термического синтеза концентрата) и доломита углеродом и кремнием. Методом термодинамического моделирования с использованием программного комплекса Терра определены оптимальные температуры и расходы восстановителей, обеспечивающие полное восстановление марганца. В качестве восстановителя при использовании оксидных марганецсодержащих материалов для обработки стали можно использовать любой из рассмотренных восстановителей или их комбинацию в определенных соотношениях. Результаты экспериментальных исследований позволили разработать технологию получения марокит-манганитового концентрата и монофазного синтетического материала (CaMnO_3). Эти материалы можно получать по технологии, которая включает механическую и термическую обработки смеси высококачественного марганцевого концентрата и обожженного доломита или извести. Марокит-манганитовый концентрат применим для легирования стали марганцем при выплавке ее в электропечи и в агрегате ковш-печь, а монофазный синтетический материал – для производства металлического марганца. На основании результатов термоди-

наимических расчетов и экспериментальных исследований предложены технологические параметры обработки стали марокит-мангани-
товым концентратом в электропечи и агрегате ковш-печь. Для получения металлического марганца внепечным алуминотермическим
процессом следует использовать в качестве шихтовой составляющей монофазный синтетический материал CaMnO_3 , что позволит повы-
сить термичность процесса, а также извлечение марганца на уровне 90 %. Результаты экспериментальных исследований были получены
при использовании современных методов исследования с применением лабораторного и аналитического оборудования, а также методов
статистической обработки результатов.

Ключевые слова: высококачественный марганцевый концентрат, марокит, монофазный синтетический материал, термохимический синтез,
термодинамическое восстановление, восстановители, обработка стали, металлический марганец

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RELEVANCE

To supply the metallurgical industry with manga-
nese, a strategically important raw material, state-of-the-
art technological developments should be employed for
the industrial mining and enrichment of manganese ores
from Russian deposits. Russia has significant reserves
of manganese ores (more than 290 million tons), but
most are of low quality and classified as refractory. These
ores have a low manganese content (18 – 24 %), a high
specific phosphorus content ($\text{P/Mn ratio} > 0.006$), and
increased iron and silicon content [1 – 3].

Significant reserves of ferromanganese ores are
concentrated in the Selezenskoye and Kaigadatskoye
(32.7 million tons) deposits. Currently, these ores are not
utilized in metallurgical production because metallurgi-
cal enrichment cannot be applied to them [1; 3; 4].

Over the last two decades, to ensure cost-effec-
tive resource use, Russian [5 – 8] and foreign resear-
chers [9 – 12] have been actively exploring new chemical
and hydrometallurgical methods for enriching low-grade
manganese ores, slags, and sludges [13 – 15]. Nowa-
days, environmental safety has been added to the exis-
ting requirements related to the economic efficiency
of the processes [16 – 18].

We conducted thermodynamic calculations and exper-
imental studies on the enrichment of ferromanganese ores
in the Kemerovo region (Kuzbass). The findings allowed
us to determine the main technological parameters for
extracting manganese and iron, and to develop an enrich-
ment process diagram. This enables the production
of high-quality concentrates of manganese and iron, with
manganese recovery reaching 90 – 92 % and iron reco-
very amounting to 86 – 90 % [19].

The relevant objectives include studying the pro-
cesses of manganese reduction from oxides in high-
quality manganese concentrate, choosing reducing agents
that can significantly enhance manganese extraction, and
developing effective technologies for preparing and using
high-quality manganese concentrate.

MATERIALS AND METHODS

We determined the phase and chemical compositions
of high-quality manganese concentrate using chemical
and X-ray phase analysis methods.

The research [1] showed that the metallothermic
reduction of manganese from oxides is significantly
accelerated with the presence of marokite $(\text{Ca,Mg})\text{Mn}_2\text{O}_4$
and calcium and magnesium manganites $(\text{Ca,Mg})\text{MnO}_3$,
which can be obtained from the high-quality manganese
concentrate. A constant amount of marokite and calcium
and magnesium manganites is required in the initial man-
ganese-containing material for stable reduction of man-
ganese. Marokite-manganite concentrate and monophase
synthetic manganese material (CaMnO_3) can be obtained
using a technology that includes the mechanical and ther-
mal treatment of a mixture of high-quality manganese
concentrate and calcined dolomite or lime.

Marokite-manganite concentrate can be used for
alloying steel with manganese when it is smelted in an
electric furnace or ladle furnace unit, while monophase
synthetic manganese material is efficient for the produc-
tion of metallic manganese.

For thermodynamic modeling of manganese reduc-
tion from oxides of high-quality manganese concentrate
and marokite-manganite concentrate, we used the Terra
software package. This software, based on the maximum
entropy principle, finds the equilibrium composition
of a multicomponent, heterogeneous thermodynamic sys-
tem under high-temperature conditions [20].

To determine the technological parameters of the mix-
tures for steel treatment in an electrical steel-melting
furnace, briquettes were fabricated in the ladle furnace
unit from marokite-manganite concentrate and powder
of spontaneously scattered alloy FS45Mn25 (25 % Mn
and 45 % Si) [1]. The binder consisted of 23.2 % ashes
from combined heat and power plants (8.88 % Al_2O_3 ;
23.98 % SiO_2 ; 0.56 % TiO_2 ; 45.85 % CaO ; 4.98 % MgO ;
6.32 % FeO ; 8.18 % Fe_2O_3 ; 1.82 % losses on ignition
and water (the rest).

The briquettes were melted in a Tamman furnace at a temperature of 1773 – 1823 K. After holding for 5 min, the metal and slag were drained and analyzed.

Using monophase synthetic material CaMnO_3 to obtain metallic manganese through aluminothermic treatment enhances the process's thermality (since manganese in this compound is in a higher oxidation state), allows smelting to be conducted using an out-of-furnace process, and increases manganese recovery.

A software application was developed to calculate the charge composition, smelting products, and the specific thermal effect of the aluminothermic treatment of metallic manganese, based on the stoichiometric equations of heat balance in the metallothermic process.

For experimental melting, the charge included high-quality concentrate, monophase material (CaMnO_3), and aluminum powder. The melting was carried out in a crucible with an upper opening.

RESULTS AND DISCUSSION

The averaged chemical composition of the high-quality manganese concentrate is as follows: 59.50 % Mn_{tot} ; 0.28 % Fe_{tot} ; 5.35 % CaO ; 4.00 % CaCl_2 ; less than 1.00 % SiO_2 ; and less than 0.01 % P. The results of X-ray phase analysis showed that manganese in the high-quality concentrate mainly exists in the form of Mn_3O_4 , and it also contains small quantities of α -manganese, manganosite MnO , and calcium chloride CaCl_2 .

The study of the carbonothermal reduction of manganese in the Mn_3O_4 –C system, in the absence of iron, showed that the manganese reduction begins at temperatures above 1723 K with carbon consumption exceeding 1.5 moles. At this temperature, manganese starts to evaporate. At 1723 K, with excess carbon, manganese carbide (Mn_7C_3) is present in the system but disappears with increasing temperature. Complete reduction of manganese occurs at a carbon consumption of 2 moles.

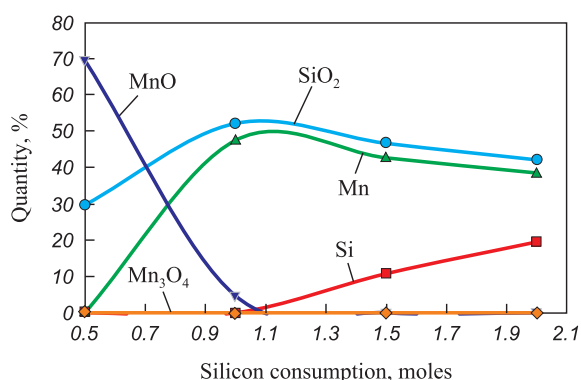


Fig. 1. Dependence of equilibrium compositions in the Mn_3O_4 – Si system on silicon consumption

Рис. 1. Зависимости равновесных составов в системе Mn_3O_4 – Si от расхода кремния

Calculations in the Mn_3O_4 –Si system showed that manganese can be reduced by silicon over the entire specified temperature range. The research results, presented in Fig. 1, indicate that complete reduction of manganese occurs at a silicon molar rate of 1 mole. This value corresponds to the maximum (47 %) manganese content in the system, which decreases with increasing consumption of the reducing agent due to dilution with excess silicon.

During thermodynamic modeling of the manganese reduction from marokite oxides, calculations were performed for 1 kg of CaMn_2O_4 , with the amount of reducing agents (carbon and silicon) ranging from 0 to 0.30 kg/kg marokite at temperatures from 1273 to 2273 K.

The results demonstrated that when carbon is used as a reducing agent, reduction begins at a temperature of 1623 K with carbon consumption exceeding 0.05 kg/kg marokite, and ends at 1723 K. When manganese is reduced from marokite oxides with silicon, the process is independent of temperature within the given range, meaning that at steelmaking temperatures, manganese reduction depends solely on the consumption of the reducing agent.

During the joint reduction of manganese from marokite with carbon and silicon at steelmaking temperatures, manganese is found in the form of metallic manganese in both condensed and gaseous phases, with no manganese carbide present.

Fig. 2 shows the dependence of the manganese recovery coefficient on the consumption of carbon and silicon at $T = 1923$ K, proving that when the consumption of reducing agents exceeds 0.2 kg/kg marokite, manganese is completely reduced.

Thus, any of the considered reducing agents or their combinations in specific proportions can be utilized as a reducing agent when manganese-containing oxide materials are used for steel treatment.

The results of the phase analysis of marokite-manganite concentrate samples obtained by heat treatment

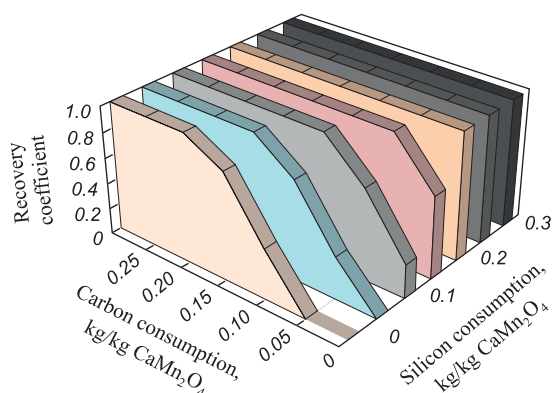


Fig. 2. Manganese recovery coefficient at combined reduction with silicon and carbon at $T = 1923$ K

Рис. 2. Коэффициент извлечения марганца при совместном восстановлении кремнием и углеродом при температуре 1923 K

of mixtures of the high-quality manganese concentrate and calcined flux metal, and their weighing after two weeks of storage in air are presented in Table 1.

The mixture of high-quality manganese concentrate and calcined dolomite, after being held for 2 h at a temperature of 1223 K, transforms into marokite-manganite concentrate, which is practically non-hygroscopic in air as it does not contain free calcium oxides.

Experimental data on the manganese reduction from briquettes with the marokite-manganite concentrate are presented in Table 2. When the marokite-manganite concentrate is obtained from the high-quality manganese concentrate and dolomite, steel treatment with manganese-containing materials becomes practically waste-free.

The ratio $(\text{CaO} + 1.4\text{MgO})/\text{Mn}$ in the marokite-manganite concentrate can range from 0.5 to 1.0. However, during smelting, the best results are obtained with a ratio of $(\text{CaO} + 1.4\text{MgO})/\text{Mn}$ in the marokite-manganite concentrate between 0.50 and 0.72. This is due to the lower consumption of the reducing agent (silicon), which in turn reduces the slag ratio. At $(\text{CaO} + 1.4\text{MgO})/\text{Mn} = 0.51$ to 1.00, all manganese is bound into calcium and magnesium manganites and marokite, ensuring its complete reduction. At $(\text{CaO} + 1.4\text{MgO})/\text{Mn} < 0.50$, free manganese oxides appear, leading to increased manga-

nese loss during reduction. Conversely, when this ratio exceeds 0.72, silicon consumption for reduction and the slag ratio increase, though the reduction value remains high if the ratio is less than 1.0.

Based on the results of thermodynamic calculations and experimental studies, the following technological parameters for steel treatment with marokite-manganite concentrate in an electric furnace and a ladle-furnace unit are proposed: the concentrate should be applied to the surface of the metal, and initially, the metal and slag should be thoroughly deoxidized to reduce the total oxidation of the metal-slag systems. The actual reducing agent that reduces manganese from the melt of the marokite-manganite concentrate is silicon. To conserve silicon, manganese should be initially reduced with carbon introduced with coke onto the surface of the manganese-containing oxide melt.

When using high-quality manganese concentrate to produce metallic manganese through thermochemical synthesis, a monophasic material is obtained, and the results of its X-ray phase analysis are presented in Table 3.

The manganese reduction by aluminum from the synthesized material is accompanied by a significant release of heat and can be represented by the equation

Table 1. Results of phase analysis of the samples

Таблица 1. Результаты фазового анализа образцов

Indicator	Indicator value as an experiment			
	1	2	3	4
Ratio $(\text{CaO} + 1.4\text{Mg})/\text{Mn}$ in the mixture	1.00	0.83	0.72	0.55
Main phase	Manganite	Manganite	Manganite	Marokite
Present in noticeable quantities	–	Marokite	Marokite	–
Traces, a few	Marokite, hausmannite	Gausmanite, manganosite	Gausmannite	Gausmannite
Change in weight after two weeks of holding	Not observed	Not observed	Not observed	Not observed

Table 2. Average results of experiments on reduction of briquettes with marokite-manganite concentrate

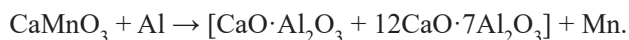
Таблица 2. Средние результаты опытов восстановления брикетов с марокит-манганитовым концентратом

Ratio $(\text{CaO} + 1.4\text{MgO})/\text{Mn}$ in the concentrate	Briquette composition, %		Melting indicators		Metal composition, %		Manganese recovery, %		Silicon consumption, kg/kg, manganese reduction	Beneficial use of silicon, %
	Alloy FS45Mn25	Concentrate	Metallic yield, %	Slag ratio, t/t	Mn	Si	from the briquette	from the concentrate		
0.55	52.6	47.4	116.5	0.61	74.14	18.55	95.8	92.80	0.441	90.3
0.72	51.7	48.3	115.7	0.64	75.26	18.90	96.0	94.27	0.422	91.6
0.83	50.5	49.5	115.5	0.71	73.30	18.15	93.1	87.90	0.483	87.0
1.00	50.2	49.8	112.2	0.75	74.50	17.45	92.3	89.48	0.540	82.3

Table 3. Results of X-ray phase analysis of synthesized monophase material

Таблица 3. Результаты рентгенофазового анализа синтезированного монофазного материала

Material	Phase composition
Lime	Large quantity of: lime CaO Present: portland Ca(OH) ₂ , calcite CaCO ₃
High quality manganese concentrate	Large quantity of: hausmannite Mn ₃ O ₄
Synthetic material	Large quantity of: CaMnO ₃ Small quantity of: marokite CaMn ₂ O ₄



The resulting Al₂O₃ reacts with CaO to form low-melting aluminates. Therefore, during reduction, manganese losses can theoretically be minimal.

Since a mixture of high-quality manganese concentrate and monophase synthetic material CaMnO₃ was used as the initial manganese-containing material, the optimal ratio of the mixture components had to be determined. Heat balances of the aluminothermic melting of metallic manganese were calculated using the developed method. The calculation results are presented in Fig. 3, *a*.

To achieve a specific thermal effect of 2500 – 2600 kJ/kg of charge required for the spontaneous process and good separation of metal and slag, the charge should contain 25 – 35 % of the synthesized monophase material

CaMnO₃ and 65 – 75 % of the high-quality manganese concentrate, as confirmed by experimental data (Fig. 3, *b*).

As a result of experimental melting, the metal was obtained with the chemical composition presented in Table 4. The results obtained clearly show that the alloy's chemical composition meets the requirements of GOST 6008 – 80.

It should be noted that metallic manganese has a low content of harmful impurities (phosphorus and sulfur), and the iron content does not exceed 1 %. The manganese recovery from the concentrate averaged approximately 90 %, and the slag ratio ranged from 2.30 to 2.65.

During the experiments, the melt temperature was about 2300 – 2373 K, with the optimal ratio of high-quality concentrate to monophase material (CaMnO₃) being 6.5 – 7.5 to 3.5 – 2.5.

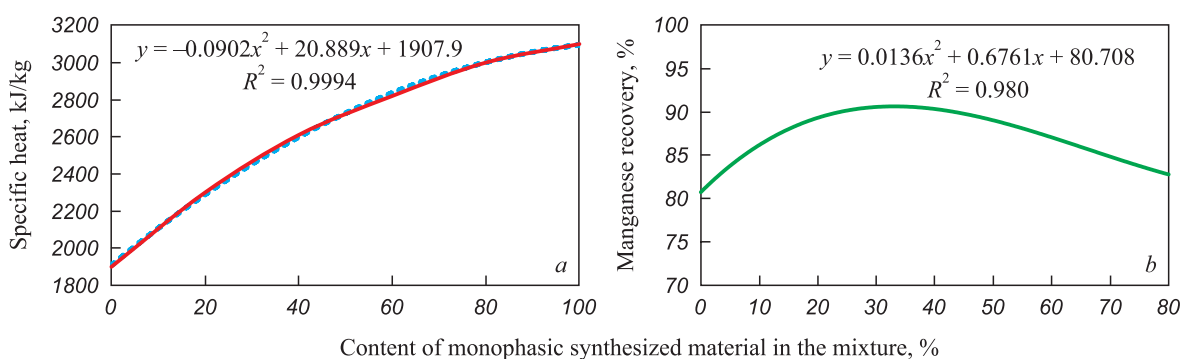
Fig. 3. Dependence of specific heat of the process (*a*) and of manganese yield (*b*) on the content of synthetic monophase materialРис. 3. Зависимость удельной теплоты процесса (*a*) и выхода марганца (*b*) от содержания синтетического монофазного материала

Table 4. Chemical composition of the experimental metal

Таблица 4. Химический состав опытного металла

Melt	Metal composition, %						MnO content in the slag, %	Mn recovery, %
	Mn	Al	Si	Fe	S	P		
1	96.89	0.81	0.45	0.88	0.004	0.006	7.24	90.70
2	97.00	0.75	0.38	0.83	0.002	0.006	6.15	90.11
3	97.12	0.73	0.40	0.74	0.003	0.005	6.01	91.16
4	96.78	0.68	0.58	0.97	0.004	0.004	8.15	87.68
5	96.84	0.83	0.64	0.91	0.004	0.006	7.35	89.89

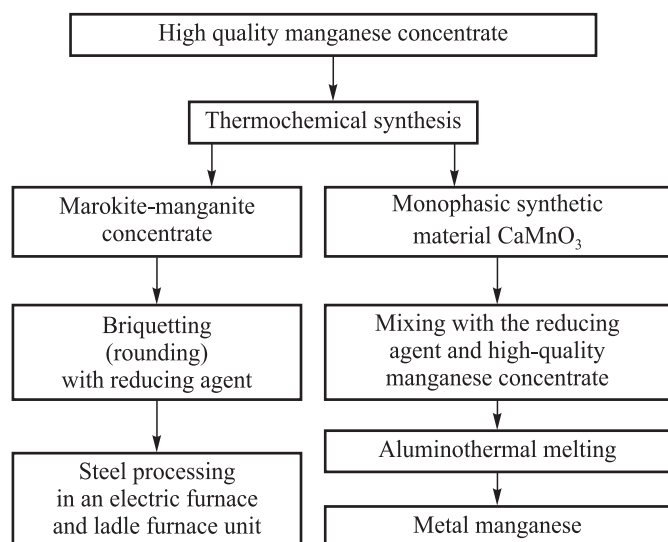


Fig. 4. Process diagram of application of high-quality manganese concentrate

Рис. 4. Технологическая схема применения высококачественного марганцевого концентрата

The recovery of manganese during smelting from the high-quality manganese concentrate reached 85.3 – 89.3 %, which significantly exceeds the manganese recovery when producing metallic manganese from peroxide manganese ores by aluminum thermal out-of-furnace treatment (69 – 72 %). The beneficial use of aluminum was 94 – 96 %.

Based on the thermodynamic and experimental studies, we developed the process diagram (Fig. 4) for using high-quality manganese concentrate obtained through hydrometallurgical enrichment of ferromanganese ores from Kuzbass.

CONCLUSIONS

The results of experimental studies enabled development of a technology for producing marokite-manganite concentrate and single-phase synthetic material (CaMnO_3), the use of which enhances manganese recovery to 90 – 92 %.

Thermodynamic modeling was used to determine the optimal temperatures and consumption of reducing agents (carbon and silicon) to ensure manganese reduction from the oxides of high-quality manganese concentrate obtained from the hydrometallurgical enrichment of ferromanganese ores, as well as from marokite-manganite concentrate. We established that any of the considered reducing agents or their combinations in specific proportions can be used as a reducing agent when oxide manganese-containing materials are used for steel treatment.

Based on the results of thermodynamic calculations and experimental studies, we proposed technological parameters for processing steel with marokite-manganite

concentrate in an electric furnace and a ladle furnace unit. To obtain metallic manganese using the out-of-furnace aluminum thermal treatment, optimal processing methods were developed involving the monophasic synthetic material (CaMnO_3) and high-quality manganese concentrate, which will increase manganese recovery to 90 %.

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M. A. Golodova – processing thermodynamic calculations, plotting and describing them, design of drawings.

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