

## RESEARCH PAPER

## THERMODYNAMIC AND EXPERIMENTAL STUDIES ON THE INFLUENCE OF HIGH-ASH COAL IN THE PRODUCTION OF HIGH-CARBON FERROCHROME

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## ABSTRACT

This study presents a thermodynamic analysis of carbon ferrochrome smelting using the "HSC Chemistry" software and laboratory experiments in a Tamman furnace. Optimal conditions for replacing metallurgical coke with high-ash coal were established, including an oxide-to-solid carbon ratio of 1.07–1.74, ensuring complete chromium reduction. The slag phase and the conditions for reduction reactions were analyzed. It was demonstrated that high-ash coal positively affects the technological parameters of the smelting process due to its silicon oxide content and high specific electrical resistance. Large-scale laboratory experiments were conducted in a 200 kVA submerged arc furnace to validate the results, resulting in a grade experimental ferrochrome sample. The study identifies optimal technological requirements for carbon ferrochrome smelting, facilitating further industrial application.

**Keywords:** Carbon ferrochrome; thermodynamic modeling; experimental modeling

## INTRODUCTION

Due to the growing scarcity and high cost of blast furnace coke, along with the challenges in supplying ferroalloy enterprises with nut coke, alternative natural carbon reductants capable of meeting the technological requirements for ferroalloy production while simultaneously reducing costs due to the low price of raw materials are needed. This need is particularly relevant given the expansion of the ferroalloy range, including chrome alloys [1].

The production of high-carbon ferrochrome is strictly regulated and depends on the stability of chrome ore raw materials. Key aspects of the technology include maintaining the "ore" layer in the bath of the ore-thermal furnace and optimizing the composition of the final slag through technological measures to improve production efficiency.

Nut coke, traditionally used in ferroalloy production, has high porosity, allowing the necessary thermokinetic conditions for the reactions of coke carbon with the charge components. However, its high cost and scarcity necessitate the search for other reductants. Therefore, carbonaceous reductants that ensure the production of ferroalloy grades with minimal content of harmful elements, such as sulfur and phosphorus, are employed in production.

One direction in the search for reductants is using low-ash coal, which exhibits a broader spectrum of properties and is cheaper than coke. Numerous attempts have been made to use these for ferroalloy production. Some researchers have suggested including low-sulfur lean coals and semi-cokes in charge, but these reductants also remain scarce [2, 3].

The experience of using high-ash coals in the production of complex alloys by a slag-free method has led to the selection of fractionated coal from the "Saryardyr" deposit as the subject of research, considered a rational replacement for part of the nut coke in the production of high-carbon ferrochrome. Various high-ash coals are frequently used as reductants in ferroalloy production. The most suitable are coals with a stable chemical composition of ash and open-pit mining, which is the most economical method of mineral extraction. Since the cost of high-ash coals is significantly lower than that of coke, their use in ferroalloy production can substantially reduce production costs and provide economic benefits [4-6].

Regarding the relatively high ash content of the coal from the "Saryardyr" deposit, it can be noted that the ash from coke should contain a minimal amount of harmful and undesirable impurities for specific metallurgical processes. It is known that the oxides of elements in the mineral part of the carbonaceous reductant can either have an activating (Fe, Ca, Mn, alkali

elements) or a passive (Si, Al) effect on the reactivity of carbonaceous materials concerning oxygen and carbon dioxide. Mineral impurities also influence the electrical conductivity, structural changes, and porosity of carbonaceous reductants, as well as the composition and properties of melts in the electric furnace, where the oxides of ash participate directly. Ash should not be considered as ballast introduced by the reductant, as it defines the physicochemical nature of the process [7].

There are strict requirements for the fractional composition of charge materials to produce high-carbon ferrochrome. The fractionation of materials in the charge is a crucial condition for the efficient production of high-carbon ferrochrome. Therefore, carbonaceous reductants, including coal from the "Saryardyr" deposit, should be used in a fractionated form, which helps reduce the sulfur content in the coal, as it primarily concentrates in fine fractions.

Based on the above requirements, using high-ash coals with a wide range of physicochemical and mechanical properties appears to be the most promising. For a preliminary assessment of the metallurgical suitability of high-ash coal from the "Saryardyr" deposit as a reductant in carbon ferrochrome production, experimental and theoretical studies on its use need to be conducted. Data from laboratory studies are essential for assessing the potential for certain chemical interactions to occur. The mixture of raw materials for the production of high-carbon ferrochrome is multicomponent (ore, quartzite, coke, and coal). To establish the mechanism of joint carbothermic reduction of chromium, iron, silicon, and aluminum in the alloy and to investigate the slag formation process, it is necessary to study the main reactions in the system Cr-Fe-Si-Al-Mg-C-O.

## MATERIAL AND METHODS

For the analysis of the carbothermic reduction of chromium, the method of complete thermodynamic modeling (CTM) of metallurgical processes was used, implemented in a computer system via the HSC Chemistry 10 software package. The applied method is based on the principle of maximum entropy, valid according to the second law of thermodynamics for an equilibrium system, regardless of the path the system takes to reach equilibrium. The comprehensive HSC Chemistry 10 program database is based on and updated by the Scientific Group Thermodata Europe database. The calculation error in the HSC Chemistry software package does not exceed 5%, considered an acceptable margin [8-15]. For the thermodynamic modeling of the process, the

Equilibrium Composition module was used. This module is designed to calculate the equilibrium compositions of interacting phases involved in technological processes. Indeed, any technological process in metallurgy can be considered as the formation process of at least two phases—the target product and the by-product. In real processes, the number of phases may be more than two; for example, in the smelting of chromium ores with a carbonaceous reductant, the target product is high-carbon ferrochrome, and the by-products are slag, dust, and gases. The distribution of metal (or multiple metals) between products is determined by the equilibrium conditions of reversible chemical reactions occurring between substances in the interacting phases. These reactions can include oxidation, reduction, and others.

A complete thermodynamic analysis was conducted for three real batch compositions for high-carbon ferrochrome smelting per 1 ton of alloy, aiming to determine the optimal conditions for the carbothermic process (Table 1).

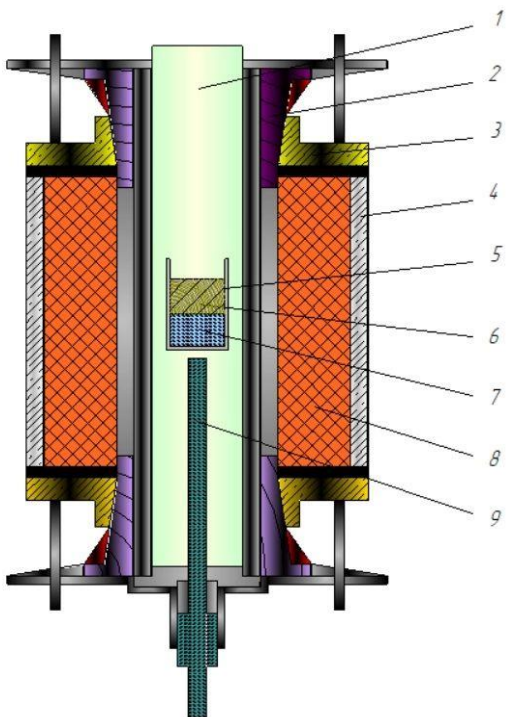
**Table 1** Composition of the Charge Mixture for High-Carbon Ferrochrome Smelting

Variants of Charge Mixture Composition	Material, kg			
	Chrome ore	Coke	Quartzite	Coal
1	2300	510	100	60
2	2120	560	70	130
3	2120	380	-	330

Based on the composition of the three variants of the charge mixture, working bodies for the smelting of carbon ferrochrome were calculated using high-ash coal from the "Saryadyr" deposit to determine the parameters of thermodynamic equilibrium (Table 2).

**Table 2** Composition of the Working Body for Carbon Ferrochrome Smelting

Variant	Cr <sub>2</sub> O <sub>3</sub>	FeO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	MgO	CaO	Fe <sub>2</sub> O <sub>3</sub>	C
1	1153,4	249,00	315,33	194,70	447,22	21,26	80,19	459,25
2	1063,1	229,87	291,11	191,73	412,16	19,97	74,26	529,93
3	1063,1	230,61	253,15	208,50	411,93	18,91	75,11	456,36



**Fig. 1** High-Temperature Tamman Furnace

1 – Graphite tube; 2 – Water-cooled copper jaws; 3 and 4 – Water-cooled body; 5 – Graphite crucible; 6 – Slag; 7 – Metal; 8 – Refractory body; 9 – Thermocouple

Experimental studies of the carbon ferrochrome smelting process using high-ash coal were conducted in a high-temperature Tamman laboratory furnace (Fig. 1). This resistance furnace is a research setup for simulating metallurgical processes at high temperatures. The high-temperature system is equipped with a heater, where the working space is a graphite tube. The temperature in the furnace is smoothly regulated by a thyristor voltage regulator connected to the primary winding of a power transformer, allowing for the output current to reach several thousand amperes at low voltage (from 0.5 to 15 V). The temperature was measured using a tungsten-rhenium thermocouple WR-5/20, whose hot junction, placed in a reinforced corundum sheath, was brought to the bottom of the crucible.

For laboratory studies, chromite ores, coke, quartzite, and high-ash coal samples were collected and prepared to study their physicochemical characteristics. Coke and its mixture with high-ash coal from the "Saryadyr" deposit were used as reductants for ferrochrome smelting. The chemical compositions of all charge materials and the technical compositions of coke and high-ash coal were determined. All charge materials were averaged and subjected to chemical analysis. The quality characteristics of the charge materials are presented in Tables 3 and 4. Calculating the charge material composition for high-carbon ferrochrome smelting involved determining the ratio of components in the charge to obtain ferrochrome of the specified grade composition, namely FeCr800. Quartzite was used as a fluxing material to regulate the high-carbon ferrochrome smelting process.

**Table 3** Chemical Composition of Initial Charge Materials for High-Carbon Ferrochrome Smelting

Material	Components, %								
	Cr <sub>2</sub> O <sub>3</sub>	FeO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	MgO	CaO	Fe <sub>2</sub> O <sub>3</sub>	S	P
Chrome ore	50,15	10,82	7,38	7,28	19,41	0,78	3,44	0,018	0,01
Coke Ash	-	-	48,9	20,17	-	3,37	-	0,088	0,006
Quartzite	-	-	96,72	0,84	0,77	0,89	0,67	-	-
Coal Ash	-	-	63,6	34,0	0,03	0,04	2,0	0,19	0,005

**Table 4** Technical Composition of Carbonaceous Reductants

Material	A <sup>c</sup> , %	W <sup>p</sup> , %	V <sup>p</sup> , %	C <sub>split</sub> , %
Saryadyr coal	42,3	2,04	17,37	38,2
Coke	15,92	2,50	0,19	81,36

The calculation was based on the complete reduction of the oxide part of the charge mixture, while also considering the oxidizing atmosphere of the environment. An excess of solid carbon was calculated to be 5-10% more than the stoichiometric amount. The ore mixture was ground to a 3-5 mm particle size. This grinding of the reductant was aimed at bringing the specific surface area values closer together and determining the effect of the reductant's chemical activity on the ore reduction processes. The mixed charge was loaded into a graphite crucible and placed in the Tamman furnace. When calculating the charge's composition, the alloy elements' distribution was assumed according to Table 5.

**Table 5** Distribution of Elements During Ferrochrome Smelting

Smelting Products	Element Distribution, %				
	Cr	Si	Fe	P	S
Alloy	94	5	97	80	10
Slag	6	95	3	10	30
Fly away	0	0	0	10	60

## RESULTS AND DISCUSSION

The thermodynamic modelling method is effectively applied to simulate technological processes during the high-temperature treatment of charge materials. This method allows for calculating equilibrium parameters, thermodynamic properties, and components' chemical and phase compositions in multicomponent systems. As the temperature increases, when phase, polymorphic, and chemical transformations occur, the problem becomes more

complex compared to classical thermodynamic conditions, where calculations are performed under standard conditions. Nevertheless, since the fundamental laws of thermodynamics apply to any system, their correct application allows for solving equilibrium calculations in general cases.

Considering various processes and states within a unified approach is only possible with a clear formalization of the model for the objects studied. Any thermodynamic system is characterized by chemical elements' relative and absolute content (mol/kg). These parameters remain unchanged when equilibrium is reached from any initial state and are sufficient to describe the system as a material object.

For the thermodynamic modeling of the carbon ferrochrome smelting process with an increased share of high-ash coal in the charge mixture, principles were formulated that formed the basis of thermodynamic modeling for the Cr-Fe-Si-Al-Mg-C-O system:

1. Temperature. Thermodynamic analysis was conducted within the temperature range of 298 to 2700 K. The lower limit characterizes the standard state, and up to 600 K, changes are insignificant. The upper limit is the final state-the melting temperature of components and the formation of final reaction products, i.e., the system's initial and final equilibrium states.

2. Pressure. In all calculations, the pressure was set at 0.1 MPa, approximately corresponding to 1 physical atmosphere, typical for most metallurgical processes, including solid-phase carbothermic interactions.

3. Volume. The thermodynamic state of the system determines the volume.

4. The system is closed, with no exchange with the external environment.

The modeling was carried out using the "Equilibrium Composition" module in the 600-2700 K temperature range with 200 K increments. For the calculation of the carbothermic reduction of chromium with carbon from coke and high-ash coal, the following phases were considered: Gaseous phases:  $\text{CO}_{2(g)}$ ,  $\text{SiO}_{(g)}$ ,  $\text{Al}_{(g)}$ ,  $\text{CrO}_{3(g)}$ ,  $\text{MgO}_{(g)}$ ,  $\text{CrO}_{(g)}$ , Condensed phases:  $\text{MgCr}_2\text{O}_4$ , Cr, C,  $\text{Cr}_3\text{C}_2$ ,  $\text{MgO}$ , Fe, SiC, FeO,  $\text{Cr}_5\text{Si}_3$ ,  $\text{MgSiO}_3$ , FeSi,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}\cdot\text{Al}_2\text{O}_3$ ,  $\text{Mg}_2\text{SiO}_4$ ,  $\text{Fe}_3\text{O}_4$ ,  $\text{SiO}_2$ , CrSi,  $\text{Cr}_4\text{C}$ ,  $2\text{FeO}\cdot\text{SiO}_2$ ,  $(\text{CaMg})_{0.5}\text{SiO}_3$ ,  $\text{CrSi}_2$ ,  $\text{Fe}_3\text{C}$ ,  $\text{CaSiO}_3$ ,  $\text{CaO}\cdot\text{MgO}\cdot\text{SiO}_2$ ,  $\text{CaMgSiO}_4$ ,  $\text{MgFe}_2\text{O}_4$ ,  $\text{Fe}_2\text{MgO}_4$ ,  $\text{CaMg}(\text{CO}_3)_2$ ,  $\text{CaFe}(\text{SiO}_3)_2$ ,  $\text{FeSi}_2$ ,  $2\text{CaO}\cdot\text{MgO}\cdot 2\text{SiO}_2$ ,  $2\text{CaO}\cdot\text{SiO}_2$ , CaO,  $\text{CaFeSiO}_4$ ,  $\text{FeO}\cdot\text{SiO}_2$ ,  $\text{Ca}_3\text{Si}_2\text{O}_7$ ,  $\text{Al}_4\text{SiC}_4$ ,  $3\text{CaO}\cdot 2\text{SiO}_2$ ,  $\text{CaO}\cdot\text{MgO}$ ,  $\text{CaAl}_2\text{SiO}_6$ ,  $\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$ ,  $\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{SiO}_2$ ,  $\text{Ca}_2\text{MgSi}_2\text{O}_7$ ,  $3\text{CaO}\cdot\text{MgO}\cdot 2\text{SiO}_2$ ,  $\text{CaO}\cdot\text{Al}_2\text{O}_3$ ,  $\text{CaO}\cdot\text{Fe}_2\text{O}_3$ ,  $\text{Ca}_3\text{Fe}_2\text{Si}_3\text{O}_{12}$ ,  $3\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$ ,  $\text{CaO}\cdot 2\text{Al}_2\text{O}_3$ ,  $\text{Cr}_2\text{MgO}_4$ ,  $\text{Fe}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ ,  $\text{MgCr}_2\text{O}_4$ ,  $\text{CaFe}_5\text{O}_7$ ,  $2\text{CaO}\cdot\text{Fe}_2\text{O}_3$ ,  $\text{Mg}_7\text{Al}_9\text{O}_4\cdot\text{Al}_9\text{Si}_3\text{O}_{36}$ ,  $4\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{Fe}_2\text{O}_3$ .

Modeling of high-carbon ferrochrome smelting using high-ash coal from the "Saryadyr" deposit revealed that up to a temperature of 2800 K, the formation and some changes in elements with transitions between the gaseous and condensed phases were observed. Fig. 2-4 presents data on the changes in individual major phases' content as a temperature function.

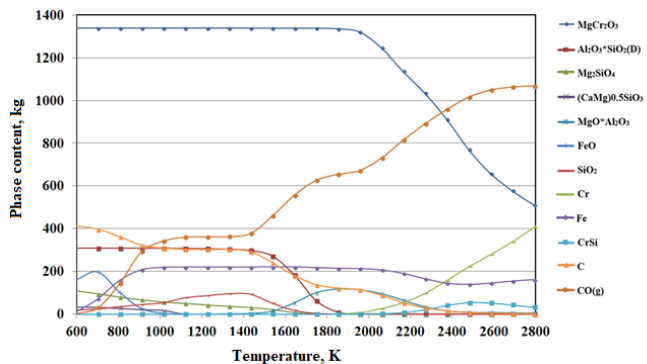


Fig. 2 Dependence of Changes in the Content of Major Phases on the Temperature of Charge Mixture No. 1

The results of thermodynamic modeling of the high-carbon ferrochrome smelting process are presented as the dependence of phase content on temperature. All graphs can be conventionally divided into three regions: below 1600 K, from 1600-1800 K, and above 2200 K.

Up to a temperature of 1500-1600 K, no sharp changes in phase content are observed, with phases such as magnesium chromite ( $\text{MgCr}_2\text{O}_4$ ), aluminum silicate ( $\text{Al}_2\text{O}_3\cdot\text{SiO}_2$ ), forsterite ( $\text{Mg}_2\text{SiO}_4$ ), magnesium-calcium metasilicate ( $(\text{CaMg})_{0.5}\text{SiO}_3$ ),  $\text{MgO}\cdot\text{Al}_2\text{O}_3$ , chromium (Cr), iron monoxide (FeO), carbon

monoxide ( $\text{CO}_{(g)}$ ), and solid carbon ( $\text{C}_{(s)}$ ) coexisting. Starting from 1400 K, the content of solid carbon and oxide compounds significantly decreases, reaching minimum values at 2000 K. As the solid carbon decreases, carbon monoxide ( $\text{CO}_{(g)}$ ) content increases. In the 1700-1800 K temperature range, a sharp reduction in  $\text{MgCr}_2\text{O}_4$  is observed, indicating chromium reduction processes, while magnesium oxide transitions to the slag phase.

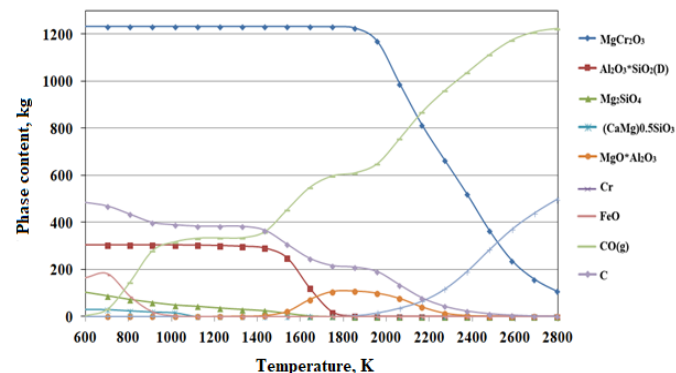


Fig. 3 Dependence of Changes in the Content of Major Phases on the Temperature of Charge Mixture No. 2

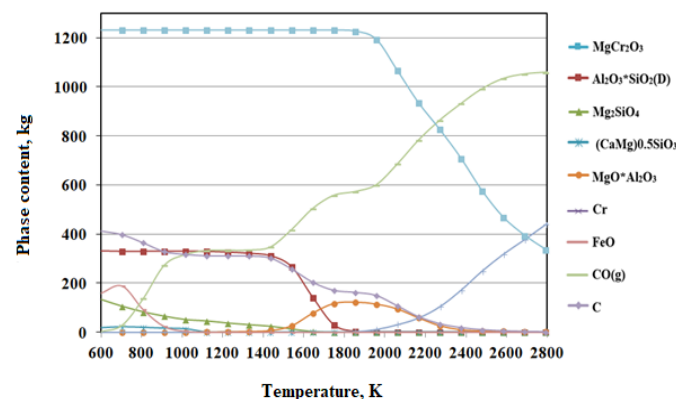


Figure 4 Dependence of Changes in the Content of Major Phases on the Temperature of Charge Mixture No. 3

Thermodynamic studies using the HSC Chemistry 10 program examined the dynamics of possible changes in the content of key condensed and gaseous phases during high-carbon ferrochrome smelting in the studied charge mixtures. Thermodynamic calculations showed no sharp technological deviations during the smelting of carbon ferrochrome using high-ash coal; the process proceeds uniformly with a complete reduction of chromium and iron. Based on the silicon oxide and aluminium content in the coal ash, it was determined that coal could successfully replace quartzite in the charge mixture. The process occurs at high temperatures and involves complex reduction reactions. Therefore, knowing the thermodynamic data and the temperature range does not provide a complete understanding of the process characteristics. Based on the theoretical data and the physicochemical properties of the charge materials studied above, a series of laboratory experiments on the Tamman furnace is required to establish the process's temperature regime and obtain a trial sample of the alloy.

The fundamental feasibility of alloy production was established based on the thermodynamic modeling of the high-carbon ferrochrome smelting process. Therefore, a series of trial experiments were conducted with different charge materials to determine the technological and temperature regimes and technical parameters close to real conditions of reduction reactions.

In addition to measuring the temperature in the furnace chamber, the heating of the charge mixture in the crucible was measured. The heating was linear, at a rate of  $10^\circ\text{C}$  per minute. During the experiment, the mass loss of the charge

mixture was continuously recorded. Due to the indirect heating method, the holding temperature was set 50–60°C higher than the theoretical temperature. Due to the technical limitations of the Tamman laboratory furnace, the temperature was limited to 1700–1800°C.

The initial gas release at 200°C corresponds to removing volatile substances, typical for high-ash coals. Carbon oxidation begins above 1000°C, where carbon oxidizes with free oxygen in the air to CO, leaving the reaction zone. This phenomenon negatively affects the reduction of the main slag and ash oxides. A mass loss was recorded up to 400°C, which may be related to the loss of volatile substances from the slag and coal.

The increase in mass in the temperature range of 400–600°C may be due to the oxidation of lower metal oxides. As the data suggests, the continuous weighing method in the study of reduction kinetics has a significant limitation: the difficulty in quantitatively assessing the chemical composition of the resulting products. During the experiment, mass loss characterizes the total weight loss into the gas phase of carbon monoxide and SiO gas. However, the rate of formation of these products and the nature of their change varies throughout the experiment, complicating the data processing for kinetic models.

At the end of the experiment, trial samples of high-carbon ferrochrome corresponding to the FeCr800 grade and slag were obtained (Table 6). The average chemical composition of the slag is presented in Table 7. The slag ratio is respectively 1.08, 1.096, and 1.108.

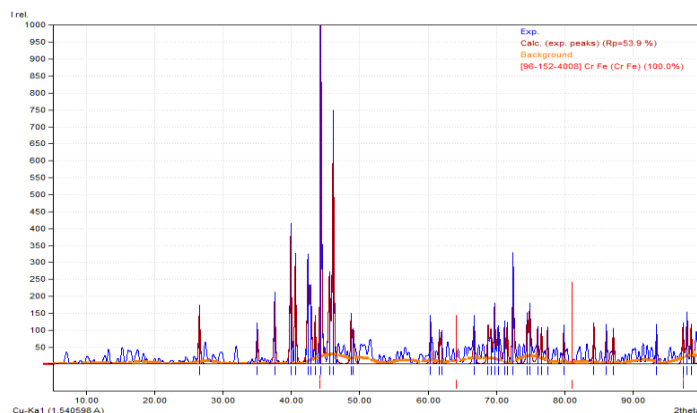
**Table 6** Chemical Composition of the Obtained High-Carbon Ferrochrome, %

Variants	Cr	Fe	Si	C	P	S
1	70,69	17,89	0,74	8,02	0,028	0,01
2	69,8	18,02	0,87	7,89	0,021	0,008
3	69,7	18,41	1,03	7,96	0,027	0,007

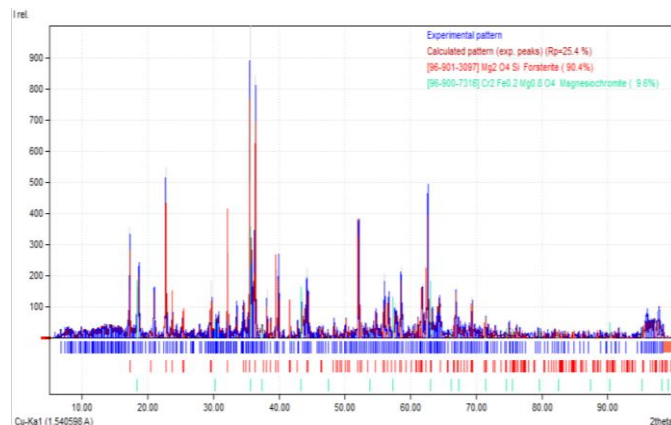
**Table 7** Average Chemical Composition of Slag, %

Variants	Cr <sub>2</sub> O <sub>3</sub>	FeO	SiO <sub>2</sub>	CaO	Al <sub>2</sub> O <sub>3</sub>	MgO	P <sub>2</sub> O <sub>5</sub>
1	6,29	0,69	35,27	1,91	15,31	40,51	0,005
2	6,21	0,68	35,28	1,87	15,9	40,02	0,006
3	6,12	0,6	36,01	1,78	16,35	39,13	0,005

X-ray phase analysis was performed using an Empyrean Malvern Panalytical diffractometer to determine the phase composition of the obtained alloy and slag. The results of the X-ray phase analysis of the obtained alloy and slag are shown in Fig. 5 and 6. The study showed that the slag contains forsterite (2MgO·SiO<sub>2</sub>) and magnesiochromite (Cr<sub>2</sub>Fe<sub>0.2</sub>Mg<sub>0.8</sub>O<sub>4</sub>). FeCr compounds were detected in the metal, and silicide compounds were absent, indicating a low silicon content in the alloy.



**Fig. 4** X-ray Phase Analysis of the Alloy



**Fig. 5** X-ray Phase Analysis of the Slag

## CONCLUSION

Results of Thermodynamic Analysis and Laboratory Smelting of Carbon Ferrochrome based on the thermodynamic analysis conducted using the "HSC Chemistry" software and laboratory experiments on smelting carbon ferrochrome in a Tamman furnace, the following key findings were established:

Replacement of Metallurgical Coke with High-Ash Coal: The optimal oxide-to-solid carbon ratio is 1.07–1.74.

After the reduction processes, the oxide phase of chromium is virtually absent; however, under real conditions, a small amount of carbon burns off in the furnace top, naturally balancing the process.

Reaction Environment and Charge Calculation: The charge was loaded into a graphite crucible, where the main reactions occurred in an oxidative environment.

Calculations revealed the need for an excess of solid carbon (1.07–1.74) since part of the carbon on the crucible surface burns off due to oxidation by atmospheric oxygen.

Mass Loss: Significant mass losses necessitate maintaining a constant charge level under the furnace top to minimize metal loss as gaseous oxides.

Intense gas evolution caused the loss of silicon and aluminum in the form of suboxides.

Temperature Regime: The working temperature of the Tamman furnace was 2000–2100 K. To ensure complete chromium reduction from complex oxides, a high concentration of heat is required to sustain temperatures around 2000 K.

Characteristics of the Slag Phase: Before the reduction processes, the slag phase was represented mainly by complex oxides (MgCr<sub>2</sub>O<sub>4</sub>, Al<sub>2</sub>O<sub>3</sub>·SiO<sub>2</sub>, Mg<sub>2</sub>SiO<sub>4</sub>, (CaMg)<sub>0.5</sub>SiO<sub>3</sub>, MgO·Al<sub>2</sub>O<sub>3</sub>).

As a result of the experiments, a grade experimental alloy sample was obtained.

Impact of High-Ash Coal: High specific electrical resistance and silicon oxide content in coal ash positively influenced the technological parameters of the ferrochrome smelting process.

Based on the obtained thermodynamic and laboratory data, optimal technological requirements for carbon ferrochrome smelting were developed for application in submerged arc furnaces. Preliminary large-scale laboratory experiments were conducted in a 200 kVA submerged arc furnace to validate the results.

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