

## THERMODYNAMIC SUBSTANTIATION OF PRODUCING SILICON-CONTAINING FERROALLOYS FROM PERLITE

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### ABSTRACT

*Perlite, which is quite widely used in various industries (construction, oil, chemical, food, agriculture and several other industries) due to the content of amorphous silica, which has increased reactivity, can become a reserve raw material for producing silicon-containing ferroalloys. The article presents the results of studies on the possibility of producing ferrosilicon and ferrosilicoaluminium from perlite in the presence of iron and carbon. The studies were carried out by thermodynamic modelling using the HSC-6.0 software package, as well as by methods of rotatable planning and geometric optimization. The conditions (temperature, amount of iron) have been determined that allow the extraction of 75 - 80 % of silicon into FeSi45 grade ferrosilicon (41.3 - 42.2 % of Si) and into FSA5510 grade ferrosilicoaluminium, containing 51.8 - 52.2 % of Si and 7.5 - 8.8 % of Al.*

*Keywords:* amorphous silicon - containing rock, perlite, silicon alloy, ferrosilicon, thermodynamic modelling.

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### INTRODUCTION

The main silicon - containing raw material in the ferrosilicon technology is quartzite. Its consumption per 1 ton of ferrosilicon is from 370 (FeSi20) to 1930 kg (FeSi75) [1]. To improve the economic indicators of ferrosilicon production, it is necessary to search for effective types of silica - containing raw materials with increased reactivity. This category of raw materials includes amorphous silicon - containing rocks - opoka, diatomite, tripoli [2 - 8], as well as perlite containing 68 - 75 % of SiO<sub>2</sub>, 11 - 14 % of Al<sub>2</sub>O<sub>3</sub>, 0.6 - 5 % of  $\Sigma$ CaO and MgO, 3 - 11 % of  $\Sigma$ Na<sub>2</sub>O and K<sub>2</sub>O, 2.4 - 3.8 % of  $\Sigma$ Fe<sub>2</sub>O<sub>3</sub> and FeO, loss after ignition - 0.2 - 5 %, 3 - 8 % of bound water [9]. The world reserves of perlite are 700 million tons [10, 11]. Due to its good swelling properties, perlite is used in the construction industry (asbestos - perlite cement, silicate perlite, bitumen perlite, carboperlite, ceramic perlite, gypsum perlite, basalt - perlite fiber, etc.) [12 - 18], oil industry, cryogenic engineering, food and chemical industries

[19 - 22], horticulture, and also in metallurgy (production of refractory materials, melting cast iron and steel in a ladle) [23 - 25]. There are several known methods of using perlite for producing of sorbents for extracting oil, oil products and liquid hydrocarbons from aqueous media [26 - 29], as well as hydrophobic, reinforced, lightweight hydrospheres, used not only as a sorbent, but also as a lightweight reinforcing filler in the chemical industry [30, 31].

Results of the studies of the hydrothermal - alkaline method for producing dicalcium silicates with a modulus of 1:4 by extracting up to 60 % of SiO<sub>2</sub> from perlite rock are presented in [32]. Comprehensive processing of perlite, containing 71 - 74 % of SiO<sub>2</sub>, provides the possibility of producing silicates of various metals, amorphous silica, calcined and caustic soda from it by an alkaline - acid combined method [33].

Considering the positive results of studies on the smelting of ferrosilicon from amorphous silica - containing rocks, the article presents the results of the research on the possibility of producing a silicon

ferroalloy from perlite in the presence of iron and carbon [34 - 36].

### EXPERIMENTAL

The chemical composition of the perlite used in the research was the following, mass. %: 76.9 SiO<sub>2</sub>(G), 13.0 Al<sub>2</sub>O<sub>3</sub>, 1.2 CaO, 1.0 Fe<sub>2</sub>O<sub>3</sub>, 3.6 K<sub>2</sub>O, 0.7 MgO, 4.1 Na<sub>2</sub>O, 0.5 TiO<sub>2</sub>. (The designation (G) for SiO<sub>2</sub> corresponds to the amorphous form of quartzite in the HSC database.)

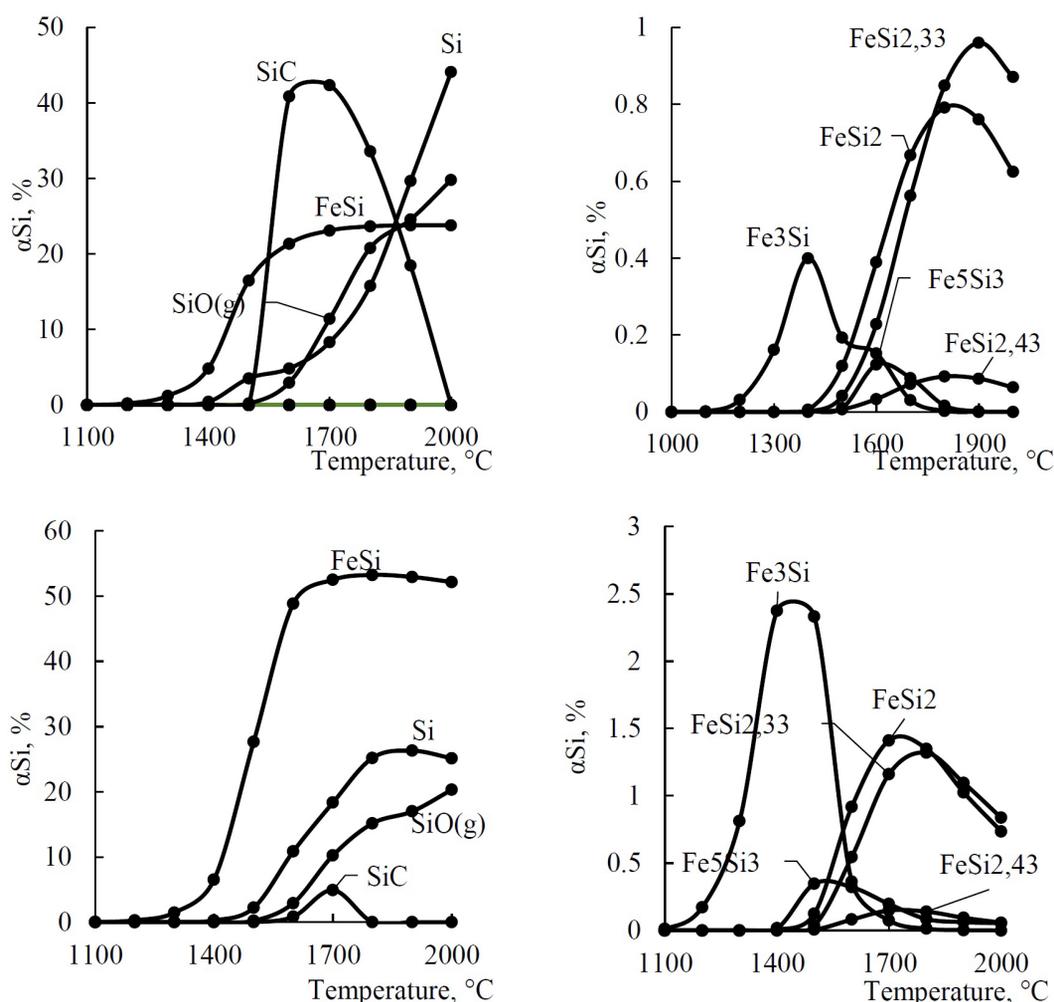
The studies were carried out by thermodynamic modelling using the HSC - 6.0 software package based on the principle of minimum Gibbs energy, the temperature range is 500 - 2000°C, the pressure is 1 bar [37]. The amount of iron was varied from 18 to 42 %

of the perlite mass (18 % iron by weight of perlite was selected to produce FeSi65 grade ferrosilicon and 42 % was selected to produce FeSi50 grade). The amount of carbon was constant in accordance with the stoichiometry for the reduction of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub>. Some studies were implemented using the second - order rotatable design (Box - Hunter plan) [38].

### RESULTS AND DISCUSSION

Fig. 1 shows the effect of temperature and iron on the equilibrium degree of silicon distribution in the products of perlite reduction with carbon.

It is evident that the main SiO<sub>2</sub> reduction products (which occurs at a temperature of > 1200°C) are FeSi,



Amount of iron, %: I - 18, II - 42.

Fig. 1. The effect of temperature and iron on the equilibrium degree of silicon distribution into the products of perlite reduction with carbon.

SiO(g), Si and SiC (to a lesser extent, silicon also turns into Fe<sub>3</sub>Si, Fe<sub>5</sub>Si<sub>3</sub>, FeSi<sub>2</sub>, FeSi<sub>2.33</sub> and FeSi<sub>2.43</sub>). An increase in the amount of iron leads to a significant change in the equilibrium degree of silicon extraction into the SiO<sub>2</sub> reduction products (Fig. 2). Thus, an increase in the amount of iron increases the degree of silicon conversion into all silicides, up to 1880°C - silicon into its elemental state, as well as the total silicon extraction into the resulting alloy to 81.3 % at 1800°C and reduces the silicon extraction into SiC and SiO (g) (for example, at 1800°C from 33.6 to 0 % and from 20.8 to 15.2 %, respectively).

A decrease in the degree of silicon extraction into SiC with an increase in the amount of iron is associated with its ability to destroy silicon carbide [39, 40]. For instance, ΔG° of the reaction presented in the Eq. (1):



calculated using the reaction equations module of the HSC - 6.0 software package is - 14.9 kJ at 1500°C, and - 25.8 kJ in 1800°C.

The decrease in the transition of silicon into SiO is explained by the fact that the carbon formed in reaction (1) reacts with SiO(g) according to the reaction presented in the Eq. (2):



ΔG° of this reaction is - 22.3 kJ at 1500°C, and - 33.5 kJ in 1800°C.

It should be noted that aluminium in the system begins to be reduced at a temperature above 1700°C. With an increase in the amount of iron in the charge, the degree of aluminium reduction decreases from 68.8 to 53.1 at 2000°C. At the same time, from 7.1 to 5.1 % of aluminium passes into gas.

The effect of iron on the concentration of silicon and aluminium in the alloy is shown in Fig. 3.

Increasing the iron content in the charge leads to decreasing in the silicon and aluminium concentrations in the alloy. For example, the silicon concentration at 1800 - 1900°C reduces from 44.0 - 50.3 % to 40.5 - 40.0 %, and the aluminium concentration in the alloy at 1900 - 2000°C decreases from 2.2 - 8.9 % to 1.5 - 4.5 %. The phase composition of the resulting ferroalloy is given in Table 1.

It is evident that the main silicon - containing phases

are FeSi and Si. At a small amount of iron, an increase in the temperature reduces the concentration of the FeSi phase due to an increase in the silicon and aluminium concentrations. At 42 % of iron, the concentration of FeSi decreases less noticeably due to a less significant increase in the aluminium concentration in the alloy.

To determine the optimal conditions for the possible production of high - quality ferrosilicon from pearlite, further studies were carried out using the second order rotatable design. Independent factors in the study are temperature, °C (T, °C) and the amount of iron, % (Fe, %). Optimization parameters are the degree of silicon extraction into the alloy (αSi(alloy), %) and the silicon concentration in the alloy (CSi(alloy), %). The research planning matrix and the results are presented in Table 2.

Using the results of the study adequate regression equations of αSi(alloy) = f(T, Fe) and CSi(alloy) = f(T, Fe) was obtained [41]. Then, in accordance with [42], volumetric and planar images of the change in αSi(alloy) and CSi(alloy) depending on the temperature and amount of iron were constructed.

The obtained regression equations (Eq. (3) and Eq. (4)) have the form:

$$\alpha\text{Si}_{(\text{alloy})} = - 762.62 + 0.738T + 4.852\text{Fe} - 1.664 \cdot 10^{-4}T^2 + 8.456 \cdot 10^{-3}\text{Fe}^2 - 2.195 \cdot 10^{-3}T\text{Fe} \quad (3)$$

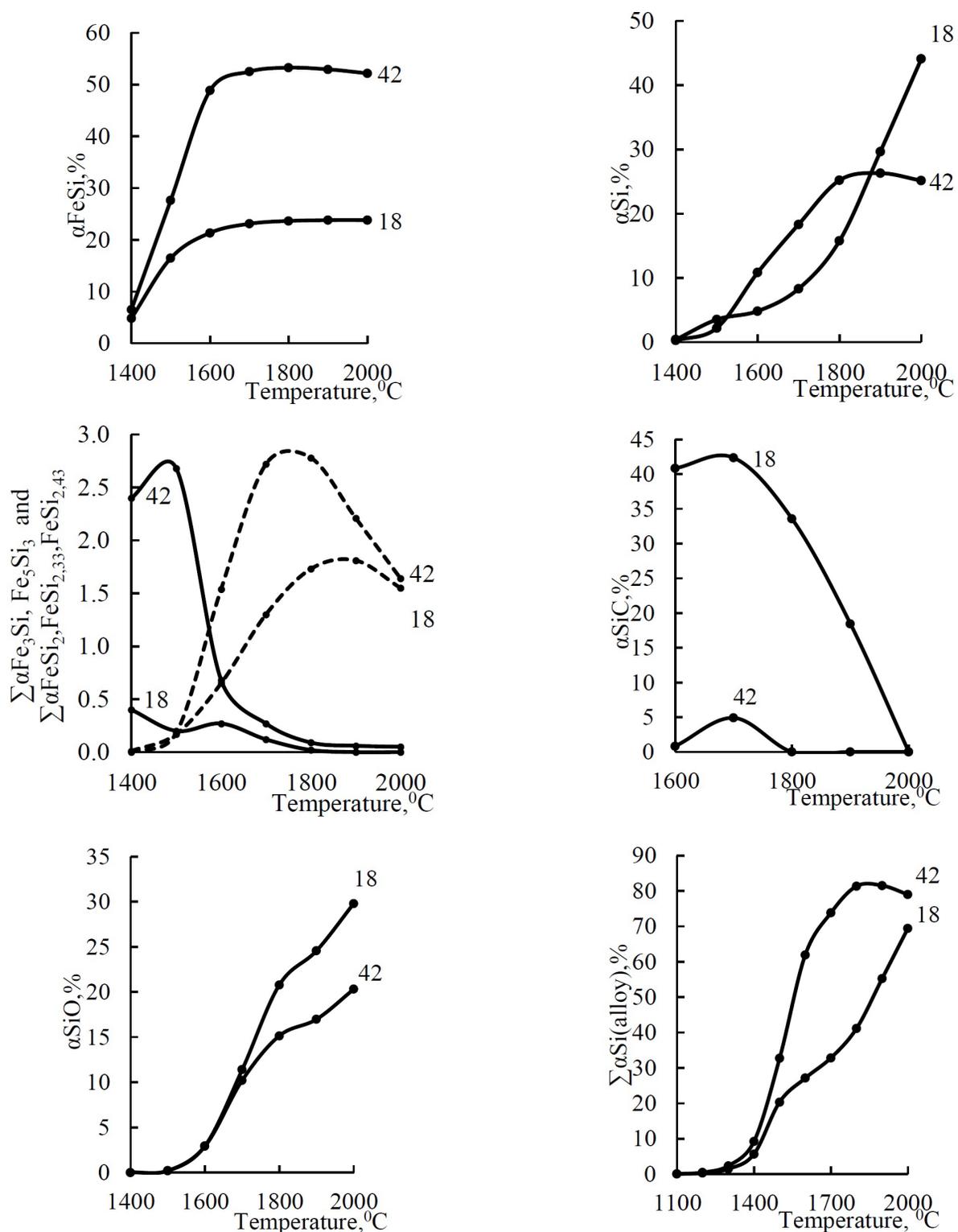
$$\text{CSi}_{(\text{alloy})} = - 299.08 - 0.33T - 1.652\text{Fe} - 7.41 \cdot 10^{-5}T^2 + 7.97 \cdot 10^{-3}\text{Fe}^2 - 1.284 \cdot 10^{-3}T\text{Fe} \quad (4)$$

The obtained equations are adequate to the research, since for the selected level of reliability (≥ 95 %) [38], the tabular value of the Fisher criterion (6.59) is greater than the calculated values of the criterion: 6.583 for Eq. (3) and 1.059 for Eq. (4).

The constructed images (volumetric and horizontal) are shown in Fig. 4.

Judging by the figure, the maximum αSi(alloy) (82.2 %) occurs in 1900°C in the presence of 42 % of iron, and the maximum silicon concentration in the alloy (48.7 %) is achieved in 1900°C in the presence of 18 % of iron. The FeSi45 grade of ferrosilicon (41 - 47 % of Si) [43] is formed in the shaded area of the figure, and the FeSi50 grade of ferrosilicon (47 - 48.7 % of Si) - in the xyz region.

To determine the conditions for the formation of high

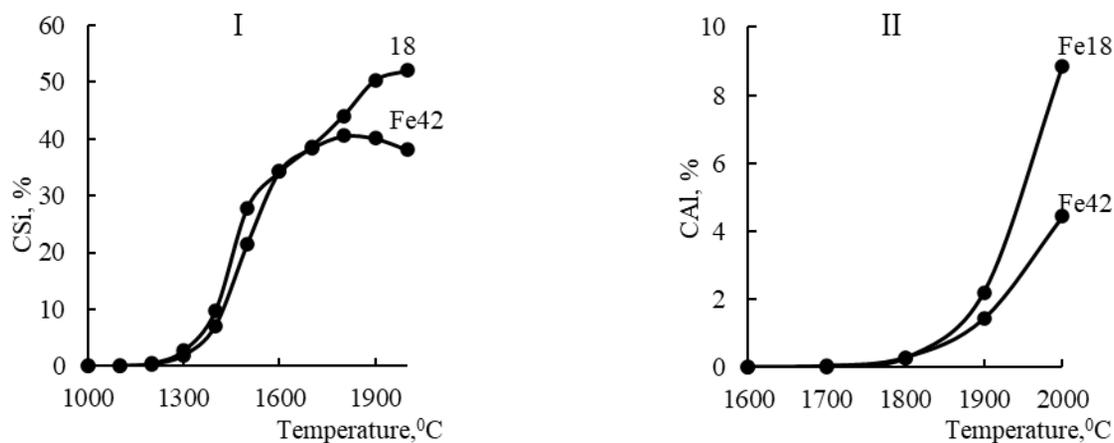


The numbers near the lines - the amount of iron, % of the perlite mass.

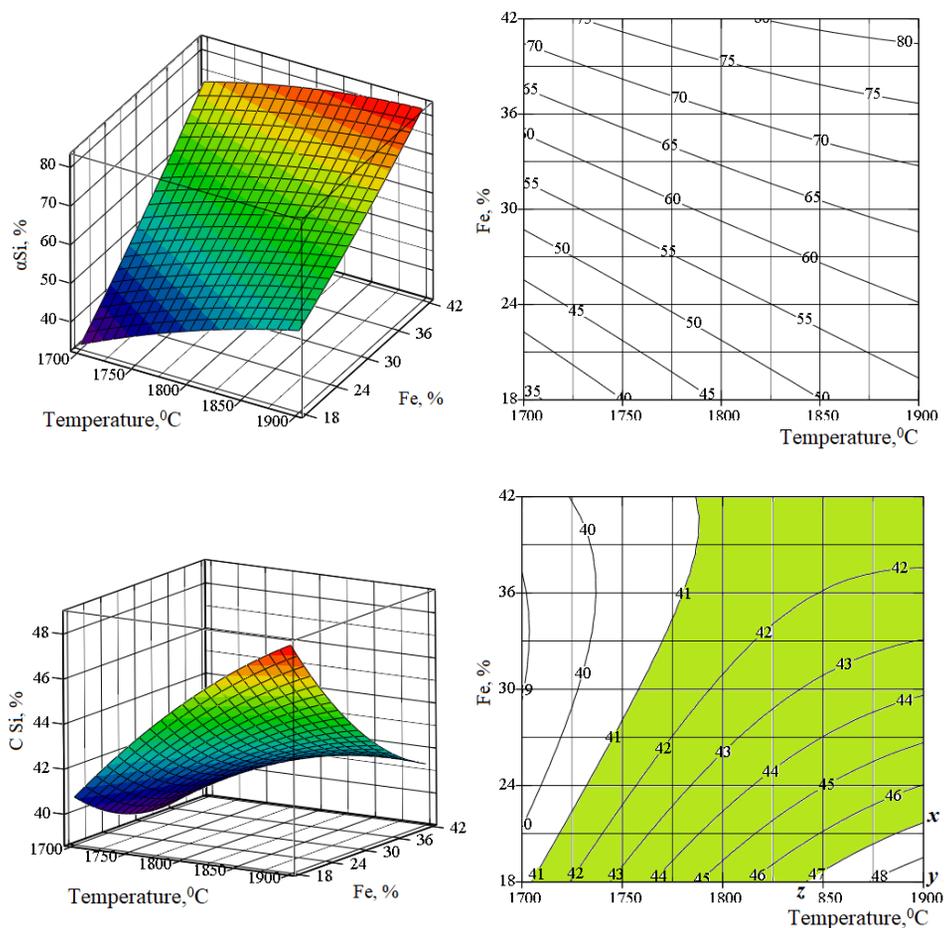
The degree of silicon distribution into: I - FeSi, II - Si, III (—) -  $\alpha\text{Si}\Sigma\text{FeSi}_2, \text{FeSi}_{2.33}, \text{FeSi}_{2.43}$ ;

(- -) -  $\Sigma\text{Fe}_3\text{Si}, \text{Fe}_5\text{Si}_3$ ; IV - SiC, V - SiO(g), VI - Si(alloy).

Fig. 2. The effect of temperature and iron on the degree of silicon distribution into the products of perlite reduction with carbon.



The numbers near the lines - the amount of iron, % of the pearlite mass.  
 Fig. 3. The effect of temperature and iron on the concentration of silicon (I) and aluminium (II) in the alloy.



Images: A - volumetric, B - planar.  
 Fig. 4. The effect of temperature and amount of iron on the degree of silicon extraction in the alloy (I) and the silicon concentration in the alloy (II).

Table 1. Phase composition of the alloy.

Fe, %	T, °C	Fe	Si	FeSi	Fe <sub>3</sub> Si	Fe <sub>5</sub> Si <sub>3</sub>	FeSi <sub>2</sub>	FeSi <sub>2,33</sub>	FeSi <sub>2,43</sub>	Al	Others*
18	1800	3.04	16.86	75.56	0.02	0.08	1.68	1.67	0.17	0.27	0.65
	2000	2.46	33.05	53.32	< 0.01	< 0.01	0.93	1.21	0.09	8.86	0.06
42	1800	4.95	12.53	79.22	0.05	0.18	1.34	1.21	0.13	0.29	0.1
	2000	6.46	12.14	75.29	< 0.01	0.11	0.71	0.75	0.05	4.44	0.04

\*)  $\sum$ Na, K, Mg, Ca

Table 2. The matrix and the research results.

#	Variables				Parameters			
	Coded		Natural		$\alpha\text{Si}_{(\text{alloy})}$ , % research	$\alpha\text{Si}_{(\text{alloy})}$ , % according to the equation	$C_{\text{Si}(\text{alloy})}$ , % research	$C_{\text{Si}(\text{alloy})}$ , % according to the equation
	X1	X1	T, °C	Fe, %				
1	- 1	- 1	1871	38.5	42.5	42.3	41.5	41.4
2	+ 1	- 1	1729	38.5	56.0	55.4	47.0	46.4
3	- 1	+ 1	1871	21.5	67.0	68.9	39.8	39.9
4	+ 1	+ 1	1729	21.5	75.2	76.6	42.2	41.8
5	+ 1.414	0	1900	30	67.0	66.7	43.3	43.9
6	- 1.414	0	1700	30	53.0	52.0	39.0	38.9
7	0	+ 1.414	1800	42	81.3	79.2	41.0	41.1
8	0	- 1.414	1800	18	44.5	45.3	45.1	45.5
9	0	0	1800	30	62.0	61.0	42.0	42.2
10	0	0	1800	30	61.6	61.0	42.7	42.2
11	0	0	1800	30	61.0	61.0	42.8	42.2
12	0	0	1800	30	60.6	61.0	41.8	42.2
13	0	0	1800	30	60.0	61.0	41.5	42.2

- quality ferrosilicon, a combined image of  $\alpha\text{Si}(\text{alloy})$ ,  $C_{\text{Si}(\text{alloy})} = f(T, \text{Fe})$  was constructed (Fig. 5). In this case, to determine the optimal temperature and amount of iron for formation of FeSi45, the following conditions were adopted:  $\alpha\text{Si}(\text{alloy}) \geq 75\%$ ,  $C_{\text{Si}(\text{alloy})} = 41 - 47\%$ , and for FeSi50 -  $C_{\text{Si}(\text{alloy})} \geq 47\%$ .

From Fig. 5 it is evident that, based on the accepted conditions, FeSi45 ferrosilicon is formed in the abcd region, and FeSi50 in the xyz region. The process parameters at the boundary points of the abcd and xyz regions are presented in Table 3.

According to Technical Specifications 0820-011-14513884-2013, a ferrosilicoaluminium alloy must contain at least 7.5 % of aluminium [44]. From Fig.

3 this condition is met at 2000°C and 18 % of iron. From Fig. 6 it follows that at 18 % of iron, a ferroalloy with aluminium concentration of  $\geq 7.5\%$  is formed in the temperature interval of 1985 - 2000°C. The  $C_{\text{Si}(\text{alloy})}$  in this temperature range is 51.8 - 52.1 % (Fig. 6). In terms of aluminium (7.5 - 8.8 %) and silicon (51.8 - 52 %) content, the resulting alloy is close to ferrosilicoaluminium of FS55A10 grade [44].

For the practical implementation of the technology for producing silicon - containing ferroalloys from perlite, a preliminary processing is necessary, which involves its swelling. This can be a slow heat treatment with the release of crystallization moisture to the softening temperature of perlite (i.e.  $\approx$  up to 850 - 1200°C) [45, 46].

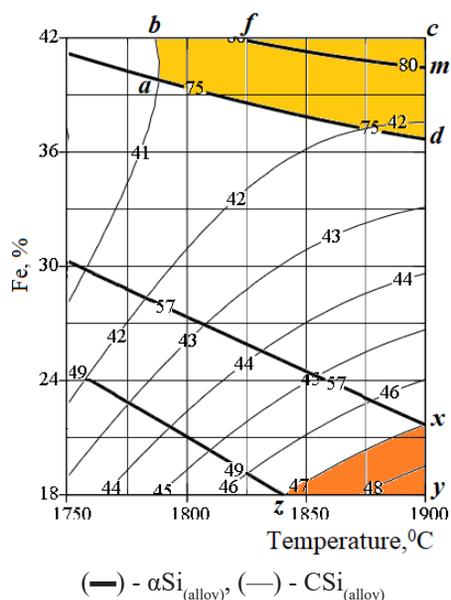


Fig. 5. Combined image of the temperature and iron effect on  $\alpha\text{Si}(\text{alloy})$  and  $\text{CSi}(\text{alloy})$ .

Table 3. Values of the process parameters at the boundary points of the FeSi45 and FeSi50 ferrosilicon formation areas.

Point in Fig. 5	T, °C	Fe, %	$\alpha\text{Si}(\text{alloy})$ , %	$\text{C}_{\text{Si}(\text{alloy})}$ , %	The alloy grade
a	1876	39.8	75.0	41.6	FeSi45
b	1873	42.0	78.4	41.4	
c	1900	42.0	82.2	41.3	
d	1900	36.7	75.0	42.2	
x	1900	22.3	57.0	46.7	FeSi50
y	1900	18.0	53.4	48.7	
z	1841	18.0	49.0	47.0	
f	1825	42.0	80.0	41.3	FeSi45
m	1900	40.8	80.0	41.5	

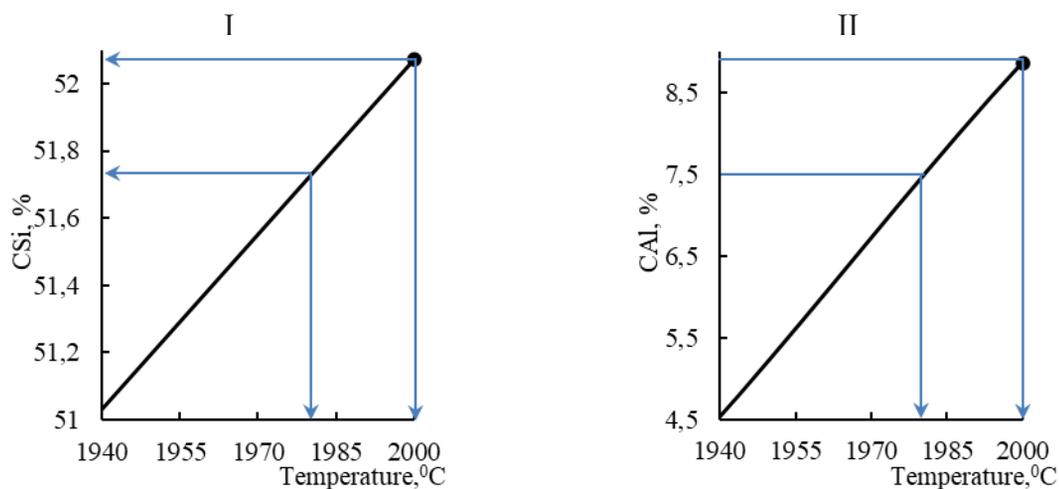


Fig. 6. The effect of temperature on the aluminium (I) and silicon (II) concentrations in the alloy in the temperature range of 1950 - 2000°C at 18 % of iron.

## CONCLUSIONS

Based on the obtained results on the equilibrium interaction of perlite with carbon and iron, the following conclusions can be made:

- products of  $\text{SiO}_2$  reduction at the temperature of  $> 1200^\circ\text{C}$  are  $\text{Fe}_3\text{Si}$ ,  $\text{FeSi}$ ,  $\text{Fe}_5\text{Si}_3$ ,  $\text{FeSi}_2$ ,  $\text{FeSi}_{2.33}$ ,  $\text{FeSi}_{2.43}$ ,  $\text{SiO}(\text{g})$ ,  $\text{Si}$  and  $\text{SiC}$ ;
- an increase in the amount of iron increases the degree of silicon transition into the alloy in the form of iron silicides and elemental silicon and reduces losses in the form of  $\text{SiC}$  and  $\text{SiO}(\text{g})$ ;
- ferrosilicon of  $\text{FeSi45}$  grade with a content of 41.3 - 42.2 % of  $\text{Si}$  (the extraction of silicon into the alloy is 75 - 80 %) can be produced in the presence of 36.7 - 42 % of steel shavings in the temperature range of 1825 - 1900 $^\circ\text{C}$ ;
- the formation of ferrosilicoaluminium of  $\text{FS55A10}$  grade containing 51.8 - 52 % of  $\text{Fe}$  and 7.5 - 8.8 % of  $\text{Al}$  and with the silicon extraction degree of 80 % takes place in the temperature interval of 1985 - 2000 $^\circ\text{C}$  in the presence of 18.0 - 22.3 % of iron.

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## Authors' contributions

*V.Sh.: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data Curation, Writing - Original Draft, Writing - Review & Editing, Visualization, Supervision.*

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