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Optimization of the electrothermal production of ferrosilicon from the leaching tailings of the oxidized copper ore of Almaly

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Abstract. The increase in the integrated use of oxidized and mixed copper ores largely depends on the creation of a technology that allows not only to extract copper, but also to obtain marketable products from non-metallic components. The article presents the results of experimental studies of electric melting of leaching tailings of copper oxidized ore of the Almaly deposit containing by weight, %: 70.9 SiO_2, 15.9 Al_2O_3, 2.1 MaO, 2.4 CaO, 3.6 K_2O, 2.2 Na_2O, 2.0 Fe_2O_3, 0.4 TiO_2, 0.2 BaO, 0.2 CuO to obtain ferrosilicon. The studies were carried out using a single-electrode single-phase arc furnace with adjustable power, as well as the method of rotatable second-order research (Box-Hunter plan). The influence of the amount of coke and steel chips on the degree of silicon extraction into the alloy and its grade was determined. It was found that with a constant amount of coke, an increase in the charge of steel chips increases the extraction of silicon into the ferroalloy and reduces the concentration of Si in it. The influence of coke on the behavior of silicon has a lesser effect. Based on the obtained volumetric and planar images of changes in the extraction of silicon into the alloy and its composition, geometric optimization of the process was carried out. It was found that ferrosilicon grade FeSi25 with the extraction of 70-75% silicon in it is formed in the presence of 30-31.3% coke, 31.3-32.0% steel chips. To obtain ferrosilicon of the FeSi45 brand, with the extraction of 70-76% silicon, the amount of coke should be 32.2-36.3%, and steel chips 25.6-28.3% of the mass of the brags; in the presence of 24-25.2% of steel chips, 30.7-36.3% of coke, ferrosilicon of the FeSi50 brand should be smelted from the tailings.

Keywords: *oxidized copper ore, leaching tailings, rotatable planning, electric smelting, optimization, ferrosilicon*.

1. Introduction

Most of the copper in the world is produced according to the scheme: ore-enrichment – matte – cathode copper – electrolysis [1]. Along with this category of ores, part of copper is oxidized and mixed ores, in which the proportion of oxidized copper ≥ 10% [2]. Dry copper is extracted from chloride sublimates by cementation method. This technology has been used on oxide copper-bearing ores from the Aktogay, Sayak, Maldybay, Kounrad, Zhezkazgan, and Bozshakol deposits.

According to this technology, it was found that the degree of copper extraction in chloride sublimations ranges from 89 to 95%. At the same time, the iron is almost completely 93-95% transferred to the stub. Silicon is not chlorinated. The stubs contain from 61 to 77% SiO_2. The degree of silicon extraction into ferroalloy of at least 70% was achieved from these stubs in production conditions at NPF Kazhiminvest LLP (Taraz) during electric melting. At the same time, a ferroalloy with a content of 41.80% Si meets the FeSi45 brand. The only serious disadvantage is the presence of chlorine-containing substances in the technological scheme during firing, therefore, when implementing this technology, it is necessary to use special anti-corrosion equipment.

There are several methods of processing oxide copper ores. This is sulfiding followed by flotation to obtain a concentrate and then, according to a well-known scheme, obtaining cathode copper.

Sulfidation is carried out with sulfur, hydrogen sulfide, sodium sulfides, barium, ammonium, sodium polysulfides, sodium thiosulfate. Lately, studies have been carried out on the sulfidation at a temperature of 503-703 K of copper from oxide ore (0.61% oxide copper, 0.34% sulfide) with gases of autogenous processes with the extraction of 73.2% of copper from the ore. For the processing of oxide copper ore (0.95% Cu with a share of chrysocolla 38%), a method of sulfoagglomeration in the presence of pyrite-containing tailings and coke has been proposed. After agglomeration carried out at a temperature of 1473-1673 K, a copper concentrate was obtained with a content of 18.5% Cu.

A method for processing sulfide-oxide copper ores of the Chiney deposit (3.1% copper, including oxide - 0.65%), consisting in dry crushing and subsequent crushing of the ore to a size of no more than 0.074 mm, bulk flotation in the presence of Na_2S sulfide and oxide copper minerals into a collective flotation concentrate, leaching of the concentrate while stirring in an aqueous solution of sulfuric acid concentration of 10.0-80.0 g/dm^3 (with the participation of an oxygen-containing environmentally friendly oxidizer with a solid content phases 10-50%) at a temperature of 293-343 K, concentrations of trivalent iron ions from 2.0-15.0 g/dm^3, dehydration and washing of the cake leaching concentrate, combining the liquid phase of concentrate leaching with the leaching cake washing waters, releasing the combined solution from solid suspensions, electroextraction of copper from solutions to obtain cathode copper. Extraction of copper into the cathode from the ore is 90.8%.

Known combined electrothermal chloride method [2]. This method is based on two sequential processes. At the first stage, at a temperature of 1200°C oxidative chloride-firing of ore is carried out, with the production of chloride copper-containing sublimates and a stub containing a significant amount of silicon oxide. At the second stage, electric melting of the stub is carried out to obtain ferrosilicon grades from FeSi20 to FeSi45.

In the Russian Federation, for more than ten years, the oxidized copper ores of the Gumeshevsky deposit have been successfully processed by underground leaching, with the processing of solutions according to the SX-EW scheme. A number of facilities are being commissioned or planned to be put into operation.

On the territory of Georgia there are also a number of deposits of oxidized copper ores, for the processing of which a leaching scheme may be promising to obtain high-quality commercial products - cathode copper [3]. Mainly this category of ores is processed by sulfuric acid heap or underground leaching [4-8]. To obtain high-purity cathode copper from solutions, the "liquid extraction - electrolysis (SX-EW)" scheme is becoming increasingly widespread. So, in 2016, according to the SX-EW scheme, 16% of copper was obtained in the world, and in 2020 this share increased to 19% (5000 thousand tons of 25900 thousand tons of copper produced in the world) [9-12].

At present, copper heap leaching technology is widely used in many countries of the world, and in recent years, the amount of ore subjected to heap leaching has been continuously increasing. Extraction of copper from oxidized ores was carried out by cementation on iron shavings, sponge iron, detinned tinplate and other cementators.

In Kazakhstan, which has at least 1 million tons of oxidized and mixed ores, a technology for producing copper according to the heap leaching scheme - SX-EW has been developed and implemented [13]. According to this scheme, in 2019, Kazakhstan received 50 thousand tons of copper (12% of the total amount produced from copper) [13]. Despite the expansion trends of the scheme: heap leaching - SX-EW, it has an obvious drawback: formation: large volumes of tailings leaching. Bearing in mind that most of the oxidized and mixed ores contain from 45 to 73% SiO_2 [13], we have proposed a technology for producing siliceous ferroalloys from copper ore leaching tailings.

The article presents the results of ferrosilicon smelting from the tailings of heap leaching (HL) of copper containing oxidized ore of the Almaly deposit.

2. Materials and methods

The electric melting of heap leaching tails was carried out at the installation shown in Figure 1. The installation consisted of a single-electrode single-phase electric furnace, a furnace transformer TDFZh-1002 with adjustable power, a short network and current and voltage monitoring devices.

The lining of the electric furnace is chromomagnesite, the base is carbon graphite. Graphite electrode with a diameter of - 7 cm. The melting was carried out in a graphite crucible (with an internal diameter of 9 cm and a height of 12 cm). The crucible was installed on a graphite plate. The space between the crucible and the lining was filled with graphite dust. The cap of the furnace, made of refractory material, was sliding.

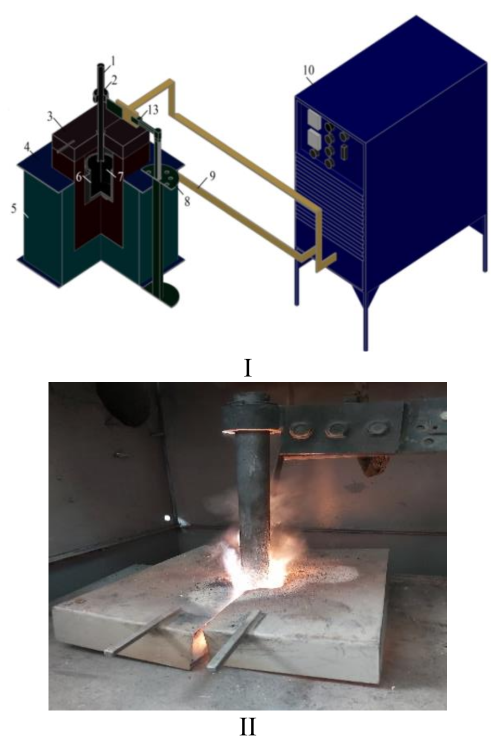

Figure 1. Installation for electric melting of copper oxide ore leaching tailings: I-installation sketch: 1 - electrode; 2- clip; 3-cap; 4 - lining; 5 - furnace casing; 6 - coke; 7 - crucible;8-the mechanism of moving the electrode, 9 - the tire; 10- transformer; II-photo of an electric furnace

The vertical movement of the electrode was carried out by a mechanical screw mechanism. The maximum movement of the electrode was 50 cm. The short network-the section from the furnace to the transformer, was made of aluminum tires (1.5 x 4.5 cm). The lower tire was connected to the carbon-graphite block using three copper studs. The upper tire was connected to the graphite electrode using a flexible copper cable. The rated power of the furnace transformer is 45 kW. The voltage and current on the low side were controlled by a voltmeter and an ammeter of the brands respectively TENGEN 42L6 GB/T7676-1998, CHNT 4226 (China) (accuracy class 1.5). Before melting, the leaching tails were granulated with 2-3% bentonite clay, dried at 150-160°C, and then fired at 600-700°C. The fired pellets had a diameter of 1-1.5 cm. Coke was also crushed to a fraction of 1-1.5 cm. The size of the steel chips did not exceed 1 cm. The furnace was heated by an arc at a current of 400-500 A and a voltage of 40-50 V.The furnace was heated for 1-1.5 hours. Then the first batch of charge (600-700 g) was loaded and melted for 5-7 minutes. After that, 2 more portions of the charge were loaded with an interval of 6-8 minutes. The total duration of the melting was 40-45 minutes. During melting, the current strength was 500-600 A, the voltage was 20-30 V. After melting, the crucible was cooled in the furnace for 3-4 hours, removed from the furnace, cooled for another 3-3.5 hours in the air. Then the crucible was broken on the cutting platform. The contents of the crucible were sorted into alloy and residue.

The elemental composition of the alloy was determined by SEM analysis using a scanning electron microscope of the brand JSM-6490LV (Japan). The degree of extraction of silicon and aluminum in the alloy was determined by the ratio of the mass of the metal in the alloy to the mass of the metal in the charge.

The research was carried out by the method of planning experiments using rotatable second-order plans (Box-Hunter plans) [16]. To obtain regression equations, the method described in [17] was used, and to construct volumetric and planar images of technological parameters, the method described in [18] was used. The tails of the Almaly ore leaching, which after calcination contained wt %: 70.9 SiO_2, 15.9 Al_2O_3, 2.1 MaO, 2.4 CaO, 3.6 K_2O, 2.2 Na_2O, 2.0 Fe_2O_3, 0.4 TiO_2, 0.2 BaO, 0.2 CuO.

In accordance with the theory of second-order rotatable planning, the number of necessary experiments with two factors is 13 [17].

Similarly, the core of the plan is characterized by five necessary experiments. The value of the "star" shoulder also depends on the number of independent factors in our case, the amount of coke (C) and steel shavings (Sh, %) from the weight of the tailings. In accordance with the theory of the rotatable planning order of the second order, the value of the "Star" shoulder (γ) is determined from the expression.

$$\gamma = 2^{k/4} \qquad (3)$$

where, K is the number of independent factors.

In our case, the value of the "Star" shoulder will be

$$\gamma = 2^{2/4} = 1.414$$

the number of experiments with "Star" shoulders is 4.0.

Table 1. Planning matrix and research results on electric melting of leaching tailings of oxidized ore from Almaly deposit

№	Variables				$\alpha_{si(alloy)}$, %		$C_{si(alloy)}$, %	
	Encoded		Natural					
	X_1	X_2	K, %	Ст, %	Exp	Calculat.	Exp	Calculat.
1	-1	-1	31.5	25.2	62.3	60.6	44.4	45.3
2	+1	-1	38.5	25.2	70.1	67.4	47.1	48.0
3	-1	+1	31.5	30.8	74.0	74.6	39.7	32.7
4	+1	+1	38.5	30.8	78.4	78.0	33.4	33.4
5	1.414	0	40	28	69.3	71.1	40.0	39.9
6	-1.414	0	30	28	63.6	63.9	37.5	37.6
7	0	1.414	35	32	82.2	81.6	30.0	31.3
8	0	-1.414	35	24	61.4	64.5	51.8	50.5
9	0	0	35	28	75.4	75.9	41.6	41.7
10	0	0	35	28	76.2	75.9	41.0	41.7
11	0	0	35	28	75.0	75.9	41.3	41.7
12	0	0	35	28	77.0	75.9	42.0	41.7
13	0	0	35	28	75.8	75.9	42.5	41.7

Table 1 shows the planning matrix and the results of experimental studies on the smelting of ferroalloys from the leaching tailings of oxidized copper ore from the Almaly deposit. Independent factors in planning are the amount of coke (C) and steel shavings (Sh), % of the tailings weight. Optimization parameters – The degree of silicon extraction into the alloy (αsi(alloy), %) and the silicon content in the alloy (Csi(alloy), %).

Using the data from Table 1 on the computer methodology [17], it was determined that the regression equations αsi(alloy) = f(C, Sh) and Csi(alloy)= f(C, Sh) have the following form:

$$\alpha si(alloy) = -667.806+21.13\cdot C+16.098\cdot Sh-0.342\cdot K2-0.1938\cdot Sh2-0.0867\cdot C\cdot Sh \qquad (1)$$

$$Csi(alloy) = -135.44+10.086\cdot C+2.164\cdot Sh-0.119\cdot C2-0.048\cdot Sh2-0.0535\cdot C\cdot Sh \qquad (2)$$

Table 1 shows that the difference between the experimental and calculated values of αsi(alloy) and Csi(alloy) insignificant. In particular, the maximum error for αsi(alloy), is observed in the second alloy and is only 3.89%, and for Csi(alloy) in the seventh alloy is 4.15%.

Equations (1, 2) are adequate, since Fischer's tabular criterion (6.59) is greater in both cases than the calculated one. All coefficients of regression equations in accordance with the Student's test are significant. Using equations (1, 2) according to the method [16, 17], three-dimensional and planar images of the influence of temperature and the amount of steel shavings on the concentration of silicon were constructed.

Figure 2 shows samples of some smelted alloys.

I

II

Figure 2. Photos of alloys: I – Experience Alloy 7; II – Experience Alloy 11

Using the methodology [18] and equations 1, 2, volumetric and planar images of the dependence αsi(alloy) - f(C, Sh) andCsi(alloy) = f(C, Sh). Figure 3 shows these images.

It can be seen that in the considered area the amounts of coke and steel shavings αsi(alloy) varies from 52 to 82.5 (points X and Y in Figure 3 (I)) and Csi(alloy) – from 28% to 50.6% (points f and t in Figure 3 (II). At a constant amount of steel chips, the dependence of the change in αsi(alloy) has an extreme character. For example, at 24% steel shavings, an increase in coke changes αsi(alloy) from coke as follows (table 2).

The decrease in αsi(alloy) with a constant amount of shavings is due to the fact that with an excess of coke, the electrical conductivity of the bath increases, as a result, the age current.

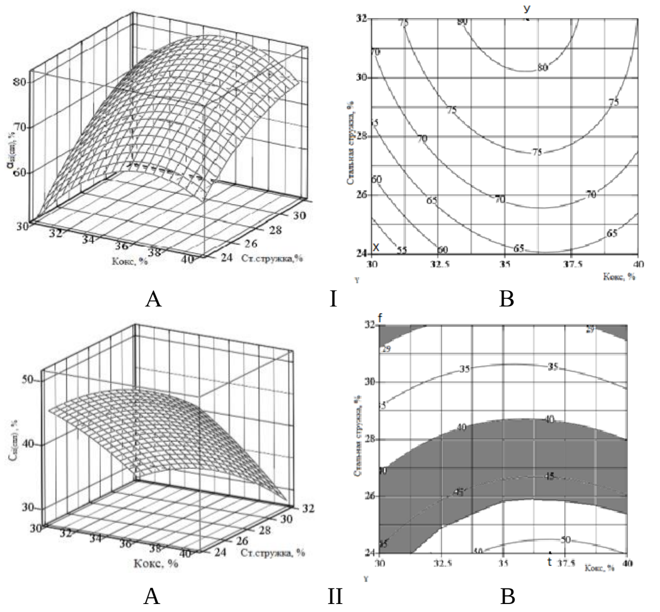

Figure 3. Influence of coke and steel shavings on Si extraction into alloy (I) and Si content in alloy (II): A – Volumetric, B – Planar

Table 2. Influence of coke and steel shavings on $\alpha_{si(alloy)}$, %

Coke, %	Steel shavings, %		
	24	28	32
30	52.0	61.5	73.2
35	64.8	74.6	81.4
40	61.4	70.6	75.4

To reduce it and maintain constant power, the electrode rises into the upper horizons of the furnace. At the same time, the filter layer of the charge decreases and the opening of the grate and the formation of fistulas is observed. In this regard, there is an increase in the transition of silicon to gaseous SiO and a decrease in the degree of silicon extraction into the alloy, for example, at 32% of steel shavings from 82% to 75%. Growth in the αsi(alloy) with an increase in the charge of steel chips is associated with an increase in the possibility of the degree of dissolution of Siin iron, thereby the equilibrium of the reaction

$SiO_2 + 2C = Si + 2CO$ shifts to the right.

To determine the conditions for obtaining branded ferrosilicon from the tailings of heap sulfuric acid leaching of copper ore with a technologically appropriate degree of silicon extraction into the alloy, we used the method of combining optimization parameters in one figure. This method has shown its effectiveness in obtaining ferroalloys from various natural and man-made raw materials [19, 20]. Figure 4 shows the combined information about αsi(alloy) and Csi(alloy), and table 3 shows the values of technological parameters at the boundary points of the formation of branded ferrosilicon with the technologically appropriate degree of silicon extraction into the alloy ≥ 65%.

From Figure 4 it can be seen that in the abc technological field, the extraction of silicon into an alloy is 75-76.4%, the amount of coke is 33.7-36.3%, and the amount of steel shavings is 27.6-28.3%. In the field of caed, the extraction of silicon into the alloy is 70-75%, the amount of coke is 32.2-36.3%, and the amount of steel shavings is 25.6-28.0%.

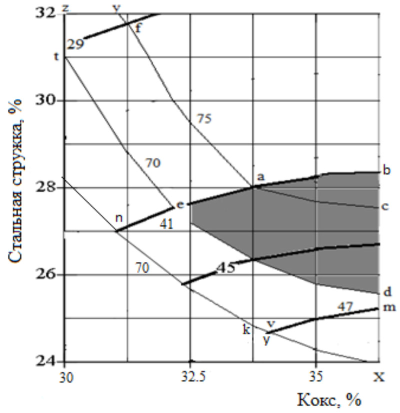

Figure 4. Combined information on the effect of coke and steel shavings on $\alpha_{si(alloy)}$ and $C_{si(alloy)}$: (—) silicon extraction, % (━━) silicon concentration in the alloy, %

Table 3. Parameter values at the points of technological areas, branded ferrosilicon Figure 3

The points in Figure 3	Technological parameters			
	Coke, %	St.sh., %	$\alpha_{si(alloy)}$, %	$C_{si(alloy)}$, %
a	33.7	28.0	75.0	41.0
b	36.3	28.3	76.4	41.0
c	36.3	27.6	75.0	42.8
e	32.2	27.4	70.0	41.0
d	36.3	25.6	70.0	46.4
n	30.7	27.0	65.0	41.0
m	36.3	25.2	69.1	47.0
k	33.7	24.6	65.0	47.0
x	36.3	24.0	65.0	50.6
z	32.0	32.0	72.3	28.1
y	32.0	32.0	75.0	28.6
t	30.0	31.0	70.6	29.4
f	31.3	31.8	75.0	29.0

In the nedmk technological field, the extraction of silicon into an alloy is 65-70%, the amount of coke is 30.7-36.3%, and the amount of steel shavings is 24.6-27.4%. In this technological area, ferrosilicon of the FeSi45 brand is formed.

In the kmx region, silicon is extracted into an alloy by 65-70%, the amount of coke is 33.7-36.3%, and steel shavings are 24.0-24.6%. In the tzyf region, the extraction of silicon into the alloy reaches 60-65%, the amount of coke is 32.5-36.3%, and the amount of steel shavings is 24.0-24.6%. In this technological field, ferrosilicon of the FeSi50 brand.

In the technological field of tzyf, the extraction of silicon into an alloy is 70-75%, the amount of coke is 30-31.3%, and the amount of steel shavings is 31.2-31.8%. In this area, ferrosilicon of the FeSi25 brand is formed.

Based on the data in Table 3, Table 4 is compiled, which shows the conditions for smelting branded ferrosilicon [21] from copper ore leaching tailings.

Table 4. Conditions for the smelting of branded ferrosilicon

The technological area in Figure 3	$\alpha_{si(alloy)}$, %	Alloybrand	Amount	
			Coke, %	St. shavings, %
abc	75-76.4	FeSi45	33.7-36.3	27.6-28.3
caed	70-75	FeSi45	32.2-36.3	25.6-28.0
nedmk	65-70	FeSi45	30.7-36.3	24.6-27.4
kmx	65-70	FeSi50	33.7-36.3	24.0-25.2
kxl	60-65	FeSi50	32.5-36.3	24.0-24.6
tzyf	70-75	FeSi25	30-31.3	31.2-31.8

The eabd region (the darkened area of Figure 3) should be taken as the optimal conditions for electric tailings leaching of copper ores, which is distinguished not only by high extractions (70-76.4%) of silicon and the ferrosilicon (FeSi45) running grade, but also by non-narrow areas of charge components (coke 32.2-36.3%, steel shavings 25.6-28.3%).

3. Conclusions

Based on the results obtained on the electric melting of the leaching tailings of the oxidized ore of Almaly, the following conclusions can be drawn:

- ferrosilicon grade FeSi25 with the extraction of 70-75 silicon into it is formed in the presence of 30-31% coke and 31.2-31.8% steel shavings;
- to obtain ferrosilicon grade FeSi45, with the extraction of 70-76% silicon, the amount of coke should be 32.2-36.3%, and steel chips 25.6-28.3% of the tailings weight;
- in the presence of 24-25.2% of steel chips, 30.7-36.3% of coke, ferrosilicon grade FeSi50 is smelted from the tailings.

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Алмалы тотыққан мыс кенін шаймалау қалдықтарынан ферросилицийдің электротермиялық өндірісін оңтайландыру

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Аңдатпа. Тотыққан және аралас мыс кендерін кешенді пайдаланудың артуы көбінесе мысты ғана емес, сонымен қатар кенді емес компоненттерден тауарлық өнімді алуға мүмкіндік беретін технологияны құруға байланысты. Мақалада құрамында Алмалы кен орнының массасы бар тотыққан мыс кенін шаймалау қалдықтарын электрмен балқытудың эксперименттік зерттеулерінің нәтижелері келтірілген, %: 70.9 SiO_2, 15.9 Al_2O_3, 2.1 MaO, 2.4 CaO, 3.6 K_2O, 2.2 Na_2O, 2.0 Fe_2O_3, 0.4 TiO_2, 0.2 BaO, 0.2 CuO алынған ферросилиций. Зерттеулер бір электродты, бір фазалы, реттелетін доғалы пешті, сондай-ақ екінші ретті айналмалы зерттеу әдісін (бокс-Хантер жоспары) қолдана отырып жүргізілді. Кокс пен болат жаңқаларының мөлшері кремнийдің қорытпаға және оның маркасына қаншалықты алынатындығына әсері анықталды. Кокстың тұрақты мөлшерімен болат жаңқаларының шихтасындағы ұлғаюы кремнийдің ферроқорытпаға шығарылуын жоғарылататыны және ондағы Si концентрациясын төмендететіні анықталды. Кокстың кремнийдің мінез-құлқына әсері аз әсер етеді. Кремнийді қорытпаға шығарудың өзгеруінің және оның құрамының алынған көлемді және жазықтық кескіндерінің негізінде процесті геометриялық оңтайландыру жүргізілді. ФС25 маркалы ферросилиций оған 70-75% в кремнийин бөліп алу арқылы 30-31.3% кокс, 31.3-32.0% болат жаңқаларының қатысуымен түзілетіні анықталды. ФС45 маркалы ферросилиций алу үшін 70-76% кремний алынып, кокс мөлшері 32.2-36.3%, ал болат жаңқалары 25.6-28.3% массасынан болуы керек; 24-25.2% болат жаңқалары, 30.7-36.3% кокс болған кезде ФС50 маркалы ферросилиций қалдықтардан балқытылады.

Негізгі сөздер: *тотыққан мыс кені, сілтілеу қалдықтары, айналмалы жоспарлау, электрлі балқыту, оңтайландыру, ферросилиций.*

Оптимизация электротермического получения ферросиллиция из хвостов выщелачивания окисленной медной руды Алмалы

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Аннотация. Увеличение комплексного использования окисленных и смешанных медных руд во многом зависит от создания технологии, позволяющих извлечь не только медь, но и получать товарную продукцию из нерудных составляющих. В статье приводяться результаты экспериментальных исследований электроплавки хвостов выщелачивания медной окисленной руды месторождения Алмалы содержащей масс. %: 70.9 SiO_2, 15.9 Al_2O_3, 2.1 MaO, 2.4 CaO, 3.6 K_2O, 2.2 Na_2O, 2.0 Fe_2O_3, 0.4 TiO_2, 0.2 BaO, 0.2 CuO с получением ферросилиция. Исследования проводили с использованием одно одноэлектродной однофазной дуговой печи с регулируемой мощностью, а также метода рототабельного исследования второго порядка (план Бокса-Хантера). Определялось влияние количества кокса и стальной стружки на степень извлечения кремния в сплав и его марку. Было установлено, что при постоянном количестве кокса увеличение в шихте стальной стружки повышают извлечение кремния в ферросплав и уменьшает в нем концентрацию Si. Влияние кокса на поведение кремния оказывает влияние в меньшей мере. На основании полученных объемных и плоскостных изображений изменения извлечения кремния в сплав и его состава была проведена геометрическая оптимизация процесса. Найдено, что ферросилиций марки ФС25 с извлечением в него 70-75% кремния образуется в присутствии 30-31.3% кокса, 31.3-32.0% стальной стружки. Для получения ферросилиция марки ФС 45, с извлечением 70-76% кремния, количество кокса должно составлять 32.2-36.3%, а стальной стружки 25.6-28.3% от массы хвастов; в присутствии 24-25.2% стальной стружки, 30.7-36.3% кокса из хвостов выплавляться ферросилиций марки ФС50.

Ключевые слова: *окисленная медная руда, хвосты выщелачивания, рототабельное планирование, электроплавка, оптимизация, ферросилиций.*

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