

## INVESTIGATION OF PLASMA-SLAG PROCESSES IN SPECIAL AND LADLE ELECTROMETALLURGY OF STEEL

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Theoretical analysis made it possible to consider advantages of the progressive technology of steelmaking and determine specificity of main processes of the plasma-slag metallurgy, calculate partial pressure of the components and composition of the vapor-gaseous phase above slag of the fluorite–lime and lime–alumina--silicate systems. Experimental investigations allowed specifying some peculiarities of pyrometallurgical reactions in the gas (plasma)–slag–metal system in melting of ball bearing and stainless steels, low carbon iron, high carbon ferrochromium, and alloying of iron by chromium in case of application of the plasma-arc models as heat sources.

**Keywords:** thermodynamic calculations, fluxes (slag), vapor-gaseous phase, plasma-slag processes, plasma remelting furnaces, plasma ladles-furnaces, out-of-furnace refining of steel from non-metal inclusions and sulfur, top level technology

Ferrous metallurgy of Ukraine within many years developed extensively under conditions of chronic deficit of centralized capital investments, wasteful attitude to mineral-raw material resources, and orientation at unpromising equipment, which was used for decades and exhausted its possibilities. Often many scientific-technical problems were not solved at the proper level within necessary terms, too low rate of mastering of high-tech technologies and as a result sharp disproportion between fundamental-applied investigations and their practical application in production were registered, and in a number of developments serious shortcomings and strategic miscalculations occurred.

Because of these reasons a significant portion of fixed metallurgical assets has high degree of wear, and technological level of certain process stages and production lines is much lower than in the industrially developed countries. Steelmaking production, which determines technical level and results of work of the branch, occupied in 2006 eighth position in the world in regard to gross melting of steel, its structure included huge volume of openhearth and insufficient development of oxygen-converter and especially electric steel melting conversions, and specific weight of continuous steel casting (CSC) was the lowest among ten countries ---main producers of steel (Table 1) [1].

These are the reasons of insufficiently high quality of Ukrainian metal products, low level of their certification, limited assortment, high production cost, and low competitiveness both on internal and on external markets. This stipulates urgent need of scientific-technical re-equipment of ferrous metallurgy, modernization and restructuring of its enterprises, and drastic improvement of structure of steelmaking production by gradual liquidation of open-hearth production and its replacement by oxygen-converter and electric steelmaking production in combination with out-of-furnace processing (OFP) and CSC.

In steelmaking production of industrially developed countries concept of the technology of the top level steel production has formed [2]. Its essence consists in application of arc steel furnaces (ASF), oxygen converters or open-hearth furnaces as high-productivity units for melting of semi-manufactured products and maximal ensuring of conditions of operation of the furnaces exclusively for accelerating process of melting of the metal scrap, cast iron and additives. The only element of metallurgical technology, carried out simultaneously with melting of the charge, remains dephosphorization, while all other processes are imposed on OFP (adjustment of the steel chemical composition with regulation of the content of separate

**Table 1.** Volume of steel production and share of melting methods among first ten countries-main producers in 2006

Country	Total volume of production, mln t	Share of method of melting and casting, %				
		Converter	ASF	Open- hearth	CSC	
China	422.7	87.0	13.0		94.8	
Japan	116.2	74.0	26.0		97.7	
USA	98.6	43.1	56.9		96.7	
Russia	70.8	61.6	18.4	20.0	53.8	
South Korea	48.5	54.3	45.7		98.0	
Germany	47.2	68.9	31.1		96.3	
India	44.0	47.3	50.5	2.3	65.9	
Ukraine	40.9	56.4	9.8	33.8	44.7	
Italy	31.6	37.4	62.6		95.5	
Brasilia	30.9	73.9	24.4	$1.7^{*}$	92.3	
All together in the world	1244.2	65.5	32.0	2.4	90.5	

Another method of melting (in energy-optimized furnaces).

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elements and the melt temperature; refining of the metal from harmful impurities, gases, and non-metal inclusions; microalloying and modification of steel and control of morphology of non-metal inclusions; improvement of technical-economic parameters of metal production both in steelmaking conversion and within the whole cycle of steel production).

Significant shift in development of plasma technology in 1970--1980s stimulated development of plasma metallurgy as one of promising directions of scientific-technical progress.

That's why in future a special place in steelmaking production plasma metallurgy should occupy, which favorably distinguishes itself not just by high quality of metal, but also by better in comparison with the open arc melting conditions of work and significant environmental advantages. It allows significant increasing of technical level of production; improving quality of the metal (first of all for special-purpose branches of industry ---- aviation, ship building, power-plant engineering, chemical industry, etc.); developing new methods of production of ferroalloys and pure metals with application of lean raw materials with high level of the base component recovery; utilizing slag, slurries and other off-grade wastes of metallurgical production.

Significant contribution into development of powerful plasmatrons was made by the E.O. Paton Electric Welding Institute of the NAS of Ukraine (PWI), Institute of Metallurgy and Materials Science of RAS, TsNIICHERMET (both Russia), companies «Krupp» (Germany), «Plasma energy» (USA), etc. While PWI developed plasmatron mainly for remelting furnaces with water-cooled moulds and tested some of them in furnaces with a ceramic crucible, the «Krupp» company first planned to use such plasmatrons in furnaces with a ceramic crucible, and in future in ladlesfurnaces and intermediary ladles of CSC machines. Having at their disposal plasmatrons, operating on alternating current up to 4 kA, this company bought in 1983 from PWI plasmatrons with plasma electrodes, designed for 6 kA current. Early in 1990s this company offered 2--8 MW plasma ladles-furnaces (PLF) of 20--150 t capacity with rate of the metal preheating up to 4  $^{\circ}C/\min$ , equipped with two or three plasmatrons of up to 12 kA current [3].

In 1986 the «Nippon Steel» company (Japan) bought from PWI license for the alternating current plasma heating module (PHM) of up to 5 MW power, which included three plasmatrons of direct action, connected according to the three-phase scheme, sources of their power supply and equipment for controlling electrical and gas conditions. Joint industrial tests of this module on PLF of 100 t capacity confirmed possibility of compensation of heat losses and melt preheating at the rate 0.82 °C/min.

In development and mastering in 1996 of PHM for equipment of the unit for complex out-of-furnace processing of steel of 30 t capacity at «Bummash» Ltd (Russia) the authors used results of theoretical and experimental investigations of PWI, industrial tests of the «Nippon Steel» company and rich experience, received in development of metallurgical alternating current plasmatrons with plasma electrodes and plasma-arc furnaces of different types, and in production of high-quality steels and alloys by methods of remelting, melting and out-of-furnace processing [4].

Plasma metallurgical processes, carried out with supply on surface of a metal pool of slag or with formation of the latter as a result of chemical reactions in the melts and characterized by significant and specific advantages, received name of the plasma-slag ones.

Plasma-arc remelting, in comparison with other methods of producing especially pure metals and allows, allows regulating within wider range atmosphere of the furnace, controlling rates of the billet melting and the ingot solidification, and makes it possible to select different refining agents [5].

Of big scientific and practical interest is investigation of one of the versions of plasma-arc remelting ---- plasma-slag remelting (PSR) [6].

Under PSR conditions, like in electroslag remelting, slag on basis of fluoric calcium, oxides of calcium, aluminium, magnesium, etc. is used. Such slag is continuously or periodically supplied in the form of powders on surface of the metal pool. Consumption of slag per a melt does not exceed 2 % of the produced ingot mass.

Slag for PSR should have a set of necessary properties: high assimilation capacity in relation to oxide and nitride non-metal inclusions; low pressure of vapors; high disulfation capacity; low adhesion relative steels and alloys being remelted; low viscosity within wide temperature range; low oxidation capacity; sufficiently high electric conductivity; stable composition under action of plasma plumes on slag, etc.

Under action of heat of plasma jets and molten metal slag easily melts, spreads over the pool surface, and partially transits into the vapor-gaseous phase. A certain share of the evaporated slag condenses on end of the molten ingot, while its bulk remains on the pool surface and, as a rule, is used for formation of skull between the ingot and the mould wall.

In process of PSR so high temperatures are achieved, at which evaporation of slag is inevitably accompanied by dissociation of its components. Products of slag evaporation, similar to the submerged-arc welding [7], may participate in the metallurgical reactions. For PSR it is necessary to know composition of the vapor-gaseous phase above the used slag. Let us take double  $CaF_2$ -CaO and triple CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> systems as an example.

Composition of the vapor-gaseous phase above the melt at different temperatures may be determined by data of dissociation of pure components and their activity in the melt [8].

Thermodynamic calculation of the vapor-gaseous phase composition was carried out at pressure in the



system 0.1 MPa and temperature 1773 K (at overheating of molten  $CaF_2$  by 100 degrees) and at 2773 and 3773 K (boiling temperature of  $CaF_2$  and CaO respectively).

Vapor-gaseous phase above slag of the first system consists mainly of  $CaF_2$ . So, at 1773 K partial pressure of  $CaF_2$  exceeds level of pressure of the rest components by 5--15 orders. Increase of temperature causes reduction of this difference: at 2772 K it constitutes 4--10 and at 3773 K ---- 3--8 orders.

Dilution of  $CaF_2$  by the CaO additive enables certain reduction of partial pressure of evaporation products of  $CaF_2$  and increase of those of CaO.

In the vapor-gaseous phase above  $CaF_2$ --CaO melts partial pressure of  $CaF_2$  is significantly higher than of CaF: at 1773 K ---- by 9, at 2773 K ---- by 6 orders, and at 3773 K by 4 orders.

Results of calculation of general pressure of vapors in the CaO--Al<sub>2</sub>O<sub>3</sub>--SiO<sub>2</sub> system and composition of the vapor phase indicate at the fact that the slag component, which determines its oxidation potential, is silica. As activity of silica grows, oxidation potential of the slag, which changes slower than the activity, increases. In the considered section activity of silica increases almost 9 times, while oxidation potential of the slag ---- only 2.25 times.

Growth of the slag oxidation potential is accompanied by increase of the silicon oxide partial pressure. Stipulated by thermal dissociation of the components oxidation potential of the slag is considered its own potential. Partial pressure of the reduction agents ---calcium, aluminium, magnesium, etc. ---- increases by means of consumption of oxygen in vapor.

Considered methodology may be used for thermodynamic analysis of specific slag of multicomponent systems, which lead to the CaO--Al<sub>2</sub>O<sub>3</sub>--SiO<sub>2</sub> quasitriple system. For steel refining in PLF the slag, containing the following components, is used, wt.%: 25--49 CaO; 7--25 MgO; 6--23 Al<sub>2</sub>O<sub>3</sub>; 14--30 SiO<sub>2</sub>; FeO, MnO ---- not more than 2.0 and 2.5 of each. Because of service conditions of the PLF lining and harmful gas release into environment, CaF<sub>2</sub> content in slag is limited by 2 wt.% [9, 10].

In state-of-the-art remelting processes three stages of metal existence in molten state are singled out: in the form of a film on the billet end being melted, drops, and a metal pool [11]. Thermodynamic analysis of composition of vapor-gaseous phase above the slag allows stating that in contrast to the electroslag remelting, at which the whole molten metal interacts with the homogeneous slag, in PSR metal in the form of a film at the billet end and in the form of drops contacts with  $CaF_2$ , while on the pool surface ---- with the used oxide-fluoric slag.

Refining of steels and alloys from non-metal inclusions may proceed in two ways. If non-metal inclusions are so strong that they do not decompose under conditions of the remelting processes, they may be removed by their carrying out by macrocurrents on interface between the metal pool and the molten slag, which wets and assimilates them. But if at the remelting temperatures non-metal inclusions dissociate into the constituting them elements and the whole oxygen or nitrogen is in the solution, their removal with assistance of the slag proceeds in the form of extraction. Slag as a medium, in which such gases dissolve better than in a metal, tends to equilibrium distribution of gases and absorbs significant share of them from the metal.

Possibility of formation (or decomposition) of non-metal inclusions in steels and alloys depends upon content of constituting them elements in the metal, and temperature and chemical composition of the steel or the alloy, i.e. upon factors, which may exert significant influence on activity of the elements, which form non-metal inclusions.

On basis of thermochemical data [8, 12, 13] temperature dependences of equilibrium constants of the integral reaction of formation of silica in the ShKh15 molten steel, titanium nitride in the Kh18N10T steel, and niobium nitride in the 00Kh16N15M3B steel were calculated:

$$lg K = lg (a_{Si}a_{O_2}/a_{SiO_2}) = 24800/T + 8.70;$$
  

$$lg K = lg [Ti] ([N]/0.01) = -15390/T - 0.54 lg T - -0.046 \cdot 10^{-3}T + 0.323 \cdot 10^{5}T^{-2} + 8.06;$$

lg K = lg [Nb] ([N]/0.01) = --11838/T -- 0.03 lg T --  
-- 0.036
$$\cdot 10^{-3}T -- 0.42 \cdot 10^{5}T^{-2} + 6.12.$$

In Figure 1 results of calculation of temperature dependence of oxygen content in the ShKh15 steel, equilibrium with slag A (97.25 % CaF<sub>2</sub> and 2.75 % SiO<sub>2</sub>) and slag B (70 % CaF<sub>2</sub> + 27.25 % CaO + 2.75 % SiO<sub>2</sub>). It was assumed in the calculation that in case of equilibrium between the metal and the slag the whole oxygen reacts only with silicon, and activity coefficients of silicon and oxygen in steel and activity of silicon in slags A and B were taken into account. Curve 1 (Figure 1) corresponds to equilibrium with slag A, curve 2 ---- with slag B, and lines 3 and 4



Figure 1. Dependence of oxygen content in ShKh15 steel upon temperature, and its actual content in metal

PLASMA-ARC TECHNOLOGY



**Figure 2.** Dependence of equilibrium content of nitrogen in Kh18N10T steel upon temperature and titanium content

show actual content of oxygen in the billet and in the ingot respectively. Having analyzed Figure 1, one may draw conclusion that silicon is stable in steel in equilibrium with slag A at the temperature below 1453--1530 K, and in presence of slag B silicon dissociates into constituting it elements (up to the temperature 1453--1530 K). So, one may assume that refining of the ShKh15 steel from silicate inclusions occurs mainly on melted end of the billet, and removal of oxygen from the metal pool under slag ---- by the extraction method.

In Figure 2 curves of equilibrium content of nitrogen are presented, which correspond, according to our experiments, to maximal (curve 1 - 0.50 %) and minimal (curve 2 - 0.14 %) content of titanium in the Kh18N10T steel, and curve 3, which corresponds to actual content of nitrogen. One can see that at 0.5 % content of titanium in steel, the titanium nitride dissociates at the temperature not lower than 2073 K.



Figure 3. Temperature dependence of equilibrium content of nitrogen in 00Kh16N15M36 steel at 0.045 wt.% Nb

Reduction of the titanium content down to 0.14 wt.% causes reduction of its dissociation temperature down to 2053 K. As far as at PSR in a water-cooled mould, main bulk of the metal does not reach so high temperature, titanium nitride in the Kh18N10T steel will not dissociate.

Dash lines 1 of minimal (0.019 wt.%) and 2 of maximal (0.045 wt %) concentrations of nitrogen in the 00Kh16N15M3B steel intersect with solid line 3 of its equilibrium content at the temperatures 1573 and 1653 K respectively (Figure 3). So, at PSR niobium nitride dissociates in this steel.

It is established in [14, 15] that nitrides of the transition metals, aluminium and silicon are well wetted by the lime-silica slags of the AN-29 type and somewhat worse by silica-fluoric slags of the ANF-6 type, etc. In addition, lime-silica slags have maximum solubility of nitrogen. That's why there is ground to assume that dominant part in removal of nitrides and nitrogen from stainless steels in PSR is played by the metal pool.

The most perfect OFP units are installations of the arc ladles furnaces (ALF) type, in which possibility of the molten steel heating and a set of devices for its stirring, addition of lumpy and injection of powder materials are envisaged. At present any OFP and CSC technologies and equipment, introduced at national metallurgical enterprises, are mainly of foreign production, which lags behind by 10--15 years from state-of-the-art technical and technological decisions. The only exclusion are put into operation at Enakievo Metallurgical Plant two new lines with CSC machines complete with two ladle-furnace installations, produced at Novokramotorsk Machine-Building Works [16].

Further way of improvement of ladles-furnaces is application of single-sleeve or current-conducting electrode holders, transition of the latter on direct current supply, conversion of conventional arcs into plasma ones by means of feeding argon into the hollow graphitized electrodes, replacement of the latter with plasmatrons, development of PLF, etc.

Application of the alternating current plasmatrons creates the following advantages [17--20]:

• application of the inert atmosphere prevents saturation of the metal with gases (nitrogen, hydrogen, oxygen) or will make it possible to alloy the metal by nitrogen, and make easier regulation of content of the elements with high affinity to oxygen and increase their assimilation;

• absence of the need to use graphitized electrodes will allow refusing from the steel decarburization process, that will simplify and make easier process of movement of the plasmatrons;

• possibility of operation of three plasma arcs at zero point will enable efficient heating of the slag and deep desulfuration and refining of the metal;

• stability of burning of the plasma arcs makes it possible to direct them neater to the ladle center, reduce wear and increase durability of the lining.



Weight share of components, % Slags MnO TiO<sub>2</sub> CaO MgO FeO Al<sub>2</sub>O<sub>3</sub> Cr<sub>2</sub>O<sub>3</sub> SiO<sub>2</sub> CaF<sub>2</sub> S Initial VO-1 50.0 30.0 20.0 VO-2 5.0 10.5 45.0 5.0 9.5 10.0 15.0 ANF-1 1.8 0.5 96.7  $\mathbf{EZF}^*$ 0.3 44.7 17.6 4.1 30.4 Produced PO-1 80.0 19.5 PO-2 13.0 67.0 17.9 5.8 PO-3 13.4 65.718.2 3.2---PO-4 6.8 74.8 2.3 \_\_\_ \_\_\_ 15.6PF-1 13.2 6.3 3.9 5.3 42.7 29.3 PF-2 17.1 6.8 3.6 5.6 34.4 33.4 PL-1 48.5 26.422.6 0.6 1.3 0.4Is given for comparison

 Table 2. Chemical composition of initial slags and slags produced after PSR of pellets, ferrochromium and chromium steel

Specificity of the PSR processes allows considering them as physical-chemical models of plasma OFP. Thermodynamics of the PSR processes can not have differences. Mass transfer may be equalized by variation of the process rate, soaking of the metal in the crucible, and intensity of the melt mixing in OFP, when the same interaction surface areas of gaseous, slag and metal phases are achieved.

The OFP and the CSC processes as technological components of the combined systems are characterized by a rather wide range of potential possibilities for their development and improvement, which are quite within the power of Ukrainian metallurgists and machine builders, provided their efforts are consolidated [21].

Experimental investigations, simulating plasma OFP, were carried out on the OB-1501 test bed, reequipped into the plasma-arc installation for melting, soaking and casting of the metal by adding a specially developed melting-casting fitting-out. This allowed carrying out melts in a copper water-cooled or ceramic crucible. In the first case interaction of the metal and the slag melts with refractory lining was excluded, which ensured purity of the experiments, and in the second case favorable conditions were created for thermal and energy investigations and for investigation of operation of the ladle-furnace slag-line area.

In PWI a complex of laboratory installations was developed and successfully used [22, 23], on which physical-chemical properties of the slag melts, used in special electrometallurgy, were investigated [24, 25]. We investigated temperature dependences of viscosity and electric conductivity of the VO-1 and VO-2 slags for OFP of steels (Table 2). Partial replacement of calcium oxide for magnesium oxode, silicon dioxide for manganese oxide, and dialuminium trioxide for ilmenite enable significant reduction of viscosity (Figure 4) and increase of electric conductivity (Figure 5) of the slag.

Metallized pellets, produced at Oskol Electrometallurgical Plant (Russia), wastes of carbonaceous ferrochromium casting, supplied by Aksu Plant of Ferroalloys (Republic Kazakhstan), and the VO-1 and the ANF-1P slags, produced in melts carried out in Ukraine, were used as the charge material.

Plasma-arc melts were carried out in inert atmosphere after loading into the crucible about 20 kg of the charge. In first series of melts iron from the pellets was produced, in second series ---- ferrochromium from the wastes of its casting, and in third series ---- chromium steel by means of alloying of iron by ferrochromium. At the beginning of each melt of first and second series intensive removal of volatile substances and moisture from the charge were detected. This was proved by the character of voltage change of the plasmatrons ---- increasing, achieving the maximum, and then reducing down to the nominal value. After achievement of the latter the melt was subjected to soaking while the plasmatrons continued to work, for a period of, approxi-





 PLASMA-ARC TECHNOLOGY

  $\chi$ , Ohm<sup>-1.cm<sup>-1</sup></sup>

 1.0

 0.5

 0

 1400

 1500

 1600

 1700

 1800

 1800

**Figure 5.** Temperature dependence of electrical conductivity  $\chi$  of VO-1 (1) and VO-2 (2) slags

mately, 10 min for finishing of the chemical reactions and homogenization of the metal.

Metallized pellets were melted according to three technological options: without addition of synthetic slags (PO-1) and with addition to 20 kg of the pellets of 1.3 kg of the ANF-1P (PO-2 and PO-3) and the VO-1 (PO-4) slag. In majority of these melts current of each plasmatron was 2 kA, while in case of melting with PO-3 it was increased up to 2.4 kA. Chromatographic analysis of the furnace atmosphere composition showed formation in the gaseous medium of hydrogen and carbon oxides, growth of their content at initial period of the melting, achievement of the maximum, and their smooth reduction down to full disappearance (Figure 6). Synchronism of the content change of these gases and voltage of the plasmatrons in the course of a melt should be noted.

In melting of the ferrochromium wastes the charge was used in the state, in which it was supplied (PF-1), or was subjected to calcination at the temperature 800 °C that caused reduction of its mass by 5 %.

It became possible to produce low-carbon iron (0.013--0.027 wt.% C) and ferrochromium, chemical composition of which meets requirements of GOST 4757-89, according to the described technology (Table 3).

Data of this Table have to be considered in greater detail. So, in plasma melting of pure pellets, 80 % of the slag constitutes iron oxide, the rest is silica. Addition of the ANF-1P slag reduces content of iron oxide down to 67 wt.%, while increase of current



Figure 6. Dynamics of content change of hydrogen and carbon oxide in gaseous medium in plasma melting of pellets

 Table 3. Chemical composition of metal produced by PSR

Slags	Weight share of elements, %							
	Fe	Cr	С	S	Р			
PO-1	Base		0.027	0.019	0.014			
PO-2	Same		0.013	0.018	0.012			
PO-3	»		0.013	0.018	0.012			
PO-4	»		0.014	0.015	0.013			
PF-1	23.5	71.6	5.400	0.080	0.031			
PF-2	20.7	73.6	5.700	0.120	0.031			
PL-1	Base	8.9	0.860	0.005	0.018			

enables deeper change of the slag composition evidently due to a fuller progress of the reaction between calcium fluoride and iron oxide with formation of the volatile iron fluoride. Absence of calcium fluoride when the VO-1 slag is added causes reduction of the iron oxide content in the final slag only up to 75 wt.%. Slag, formed in melting of ferrochromium wastes in the water-cooled crucible, differs from conventional slag of the carbothermal production by a very high content of dichrome trioxide and calcium oxide and low content of magnesium oxide and dialuminium trioxide, weight share of silicon dioxide being constant.

Products of the melts of first and second series were used as the charge for melting of chromium steels. For investigation of the desulfuration processes, about 4 kg of ferrochromium and iron of general mass 20 kg were loaded into the skull crucible, and the VO-1 slag, aluminium, and sulfurous iron were added to them, and then melting was carried out. Due to such melting significant degree of desulfuration was achieved owing to high efficiency of plasma heating and increased activity of the slag caused by high temperature. So, slag of OFP, similar to the PShP [26] and EShP [27] slags, allows ensuring achievement of high coefficients of sulfur distribution between the slag and the metal in plasma melting both in the described experiments and according to data of the work [10].

Alloying of steel by chromium, accompanied by reduction of the melt temperature because of endothermicity of the process, became the reason of reduction of the degree of overheating of water, which cools melting crucible [28].

Ladle refining of metal is an important link in production technology of steel of both special-purpose and general designation. Progress achieved within the last years in improvement of production technology of refractory materials, design of the installations, application of plasma heating, and other technological measures created premises for deepened investigation of the processes, which proceed in ladles-furnaces. This allowed significant improving quality of steel and controlling not just amount, but also chemical composition of the non-metal inclusions [29].



1. Thermodynamic analysis of composition of the vapor-gaseous medium above some slags of plasma remelting and out-of-furnace processing was carried out.

2. Carried out plasma-slag melts allowed specifying peculiarities of behavior of gases in stainless and bearing steels.

3. Plasma melts of the metallized pellets and ferrochromium wastes for production of chromium steels were carried out.

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