

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property  
Organization

International Bureau

(43) International Publication Date  
27 February 2025 (27.02.2025)



(10) International Publication Number  
**WO 2025/042299 A1**

(51) International Patent Classification:

C22C 28/00 (2006.01) C22B 1/00 (2006.01)  
C22B 5/00 (2006.01)

Published:

— with international search report (Art. 21(3))

(21) International Application Number:

PCT/PL2023/050068

(22) International Filing Date:

18 August 2023 (18.08.2023)

(25) Filing Language:

Polish

(26) Publication Language:

English

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(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MU, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SC, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

(54) Title: COMPLEX MULTI-COMPONENT ALLOY OF IRON, SILICON AND CHROMIUM

(57) Abstract: A complex multi-component alloy of iron, silicon and chromium comprising, as a weight percentage of the total weight of the alloy: not less than 21.0 chromium, not less than 61.0 silicon, 0.5 to 4.0 aluminium, 0.31 to 2.0 calcium, not more than 0.02 carbon, the rest being iron and traces of impurities, with a chromium-iron ratio of not less than 2.0. This type of alloy is used in ferroalloy metallurgy, especially as a reductant of Cr<sub>2</sub>O<sub>3</sub> comprised in chromium ore in the production of low-carbon ferrochrome. The alloy according to the invention allows increasing the chromium yield from chromium ore to low carbon ferrochrome, reducing the mass content of Cr<sub>2</sub>O<sub>3</sub> in the waste slag by increasing the reduction potential with respect to Cr<sub>2</sub>O<sub>3</sub> in chromium ore.



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### Complex multi-component alloy of iron, silicon and chromium

The invention relates to a complex multi-component alloy of iron, silicon and chromium comprising aluminium, calcium and carbon in an amount not exceeding 0.02 % wt. relative to the total weight of the alloy. The chromium to iron ratio in the alloy of the invention is at least 2.0. The alloy is produced using the carbothermic reduction method of  $\text{SiO}_2$ ,  $\text{CaO}$ ,  $\text{FeO}$ ,  $\text{Al}_2\text{O}_3$  in the presence of chromium and iron derived from high-carbon ferrochrome in a resistance-arc furnace, and then subjected to ladle decarburisation refining. This type of alloy is used in ferroalloy metallurgy, especially as a reductant of  $\text{Cr}_2\text{O}_3$  comprised in chromium ore in the production of low-carbon ferrochrome (FeCr LC). The alloy according to the invention allows increasing the chromium yield from chromium ore to low carbon ferrochrome, reducing the mass content of  $\text{Cr}_2\text{O}_3$  in the waste slag by increasing the reduction potential with respect to  $\text{Cr}_2\text{O}_3$  in chromium ore.

Various types of alloys of iron, silicon and chromium are known in the state of the art, including those described in the international standard ISO 5449:1980 (*Ferrosilicochromium - Specification and conditions of delivery*), which describes an alloy of iron, chromium and silicon obtained by reduction or fusion, comprising chromium in an amount of 20.0 to 65.0 % wt. and silicon in an amount of 10.0 to 60.0 % wt.

On the other hand, international patent application WO2019240589 A1 presents a silicon-based alloy comprising 45-95 % wt. silicon, not more than 0.05 % wt. carbon, 0.4-30 % wt. chromium, 0.01-10 % wt. aluminium, 0.01-0.3 % wt. calcium, not more than 0.10 % wt. titanium, not more than 25 % wt. manganese, 0.005-0.07 % wt. phosphorus, 0.001-0.02% wt. sulphur, and the rest is iron and trace amounts of phosphorus and sulphur. The alloy described in this international patent application differs from that described in this patent application in its calcium content.

European patent application EP3075869 A1 presents a method for producing FeSiAl alloys in which carbon rock is mixed with quartzite, iron-bearing material and wood chips, if necessary, carbon with high volatile content, in a specified proportions, and the homogenised batch material is loaded into a melting furnace to produce FeSiAl alloy. The batch mix comprises 1.5-4.5%  $\text{Fe}_2\text{O}_3$ , 55-65%  $\text{SiO}_2$ , 25-35%  $\text{Al}_2\text{O}_3$ , 32-34%  $\text{CaO}$ , 0.3-3%  $\text{MgO}$  0.3-2%  $\text{TiO}_2$  and trace

amounts of phosphorus and sulphur. The alloy obtained in the described process comprises (% wt.): 40-85% silicon, 1-40% aluminium, 0.001-1.0% carbon, up to 2% titanium, up to 1.0% calcium and traces of phosphorus and sulphur, the rest is iron.

In turn, European patent EP2295614 B1 discloses the composition of an alloy for deoxidation and introduction of alloying additives comprising 45.0-63.0 % wt. silicon, 10.0-25.0 % wt. aluminium, 1.0-10.0 % wt. calcium, 1.0-10.0 % wt. barium, 0.3-0.5 % wt. vanadium, 1.0-10.0 % wt. titanium, 0.1-1.0 % wt. carbon, the rest is iron.

The object of the invention was to develop a new complex multi-component alloy of iron, silicon and chromium with a high reduction potential for the metallothermic production process of FeCr LC. A critical qualitative feature of iron-silicon-chromium alloys from the point of view of their use as a reductants in FeCr LC production processes and a deoxidising agents in the production of high-chromium ferritic and austenitic steels is their reduction potential, which depends on the content of elements with a higher affinity for oxygen than chromium. Due to its high reduction potential, this alloy can be used to increase the chromium yield from chromite ore and reduce the residual mass of  $\text{Cr}_2\text{O}_3$  in the waste slag. An additional purpose of the invention is to reduce the content of harmful admixtures in FeCr LC. Impurities comprised in chromite ore reductants are the main source of impurities in FeCr LC, from where, in the process of obtaining high chromium steels, for example, austenitic stainless steels, they pass into the finished product, significantly reducing its strength parameters. The biggest contaminant is carbon, which has a significant chemical affinity for chromium and forms carbides  $\text{Cr}_{23}\text{C}_6$  with it, which diffuse toward grain boundaries, which in turn contributes to selective corrosion in chromium-depleted zones, leading to embrittlement of such steel. Another very important feature of the new alloy is its energy potential related to its high content of elements with a high affinity for oxygen, such as silicon, calcium and aluminium, whose exothermic oxidation processes will make a significant energy contribution to the ladle treatment processes of high-chromium slags carried out in the production of FeCr LC. Another object of the invention is to expand the raw material base for the production of FeCr LC. Currently, the depletion of the rich chromium ores and the resulting need to use poor chromium ores with increased MgO content (up to 22%) and reduced  $\text{Cr}_2\text{O}_3$  content in ferroalloys metallurgy can be observed, resulting in increased electricity consumption rate and raw materials per ton of alloy, increased losses associated with incomplete reduction of chromium oxides, and increased slag-to-metal ratio. Since chromium spinels are among the hard-fusible compounds, which worsens the possibility of chromium-to-metal transition, some Cr retains in the slag in the form of metallic inclusions and in the form of oxides of CrO and  $\text{Cr}_2\text{O}_3$ . The slags

formed in the smelting of FeCr LC comprise 8 to 12 % wt.  $\text{Cr}_2\text{O}_3$  and are characterised by significant viscosity. By using the process of the invention, the final yield of Cr to the alloy will be more than 95%, and the residual content of  $\text{Cr}_2\text{O}_3$  in the waste slag of less than 3 % wt.

The essence of the invention is a complex multi-component alloy of iron, silicon and chromium comprising, as a percentage by weight of the total weight of the alloy: not less than 21.0 chromium, not less than 61.0 silicon, 0.5 to 4.0 aluminium, 0.3 to 2.1 calcium, not more than 0.02 carbon, the rest being iron and traces of impurities, wherein a chromium-to-iron ratio of not less than 2.0.

Preferably, when the multi-component alloy of iron, silicon and chromium comprises chromium in an amount of 21.0 to 28.0 % wt. relative to the total weight of the alloy, advantageously 21.5 to 25.5 % wt.. A lower Cr content worsens the efficiency of the FeCr LC production process, while a higher Cr content lowers the silicon content below 61 % wt.

Preferably, when the multi-component alloy of iron, silicon and chromium comprises silicon in an amount of 61.0 to 70.0 % wt. relative to the total weight of the alloy, preferably 63.5 to 68.0 % wt.. A lower silicon content results in a decrease in the reduction potential of the alloy, while a higher silicon content results in a lower chromium content and thus a deterioration in the efficiency of the FeCr LC production process by metallothermic reduction method.

Preferably, when the multi-component alloy of iron, silicon and chromium comprises calcium in an amount of 0.5 to 1.2 % wt. relative to the total weight of the alloy. Lower and higher calcium content causes a decrease in the reduction potential in the production of FeCr LC. In addition, the higher Ca content of the finished alloy requires a higher energy input for the calcium to be reduced from CaO in the carbothermic process of obtaining the invention in an arc-resistance furnace.

Preferably, when the multi-component alloy of iron, silicon and chromium comprises aluminium in an amount of 0.5 to 3.1 % wt. relative to the total weight of the alloy, preferably 0.5 to 1.3 percent by weight. Lower and higher aluminium content results in a decrease in the reduction potential in the production of FeCr LC. In addition, the higher Al content in the finished alloy requires a higher energy input for the aluminium to be reduced from the  $\text{Al}_2\text{O}_3$  in the carbothermic process of obtaining the invention in an arc-resistance furnace.

In another aspect, the object of the invention is also a method for producing a complex multi-component alloy of iron, silicon and chromium according to the invention, characterised in that

the oxides of  $\text{SiO}_2$ ,  $\text{CaO}$ ,  $\text{FeO}$ ,  $\text{Al}_2\text{O}_3$  are reduced carbothermally in the presence of chromium and iron derived from high-carbon ferrochrome in an arc-resistance furnace, and then subjected to ladle decarburisation refining. Such a method makes it possible to increase the yield of Si and Cr to the alloy of the invention, lowers the electricity consumption rate, simplifies the technology for obtaining the alloy, and makes it possible to achieve a very low carbon content.

The invention also includes the use of a complex multi-component alloy of iron, silicon and chromium according to any of the claims 1 to 6 to reduce  $\text{Cr}_2\text{O}_3$  comprised in chromium ore in the process of obtaining low-carbon ferrochrome.

The alloy of iron, silicon and chromium according to the embodiment was obtained by a process in which a mixture of feedstock raw materials for the production of the alloy is introduced periodically in specified proportions into the working space of the bath of a semi-closed, low-shaft, arc-resistance furnace by a method involving the simultaneous liquid transition of high-carbon ferrochrome parameters listed in Table 1, the reduction of  $\text{SiO}_2$  comprised in quartzite with the parameters listed in Table 2, reduction of  $\text{Al}_2\text{O}_3$  comprised in ceramic waste with the parameters listed in Table 3, reduction of  $\text{CaO}$  comprised in limestone with the parameters listed in Table 4, and reduction of  $\text{FeO}$  comprised in mill scale with the parameters listed in Table 5, with the use of elemental carbon comprised in hard coal with the parameters listed in Table 6 and chromium carbides comprised in high carbon ferrochrome, then dissolving the resulting Si-Al-Ca-Fe alloy in liquid ferrochrome, performing a tapping of the smelting products from the bath of the arc-resistance furnace into a tapping ladle, pouring the alloy into a refining ladle and performing decarburisation refining, and then pouring the alloy into a set of iron ingots with separating the material with higher carbon content.

**Table 1. Chemical composition of high carbon ferrochrome**

Component	Cr	Si	Fe	Mn	P	Al	C	S
Content, % by weight.	63.68	0.94	25.16	0.103	0.019	0.019	9.11	0.030

**Table 2. Chemical composition of quartzite**

Component	$\text{SiO}_2$	$\text{Al}_2\text{O}_3$	$\text{Fe}_2\text{O}_3$	$\text{CaO}$
Content, % by weight.	99.23	0.226	0.340	0.114

**Table 3. Chemical composition of ceramic waste**

Component	$\text{MgO}+\text{CaO}+\text{K}_2\text{O}+\text{Na}_2\text{O}$	$\text{Al}_2\text{O}_3$	$\text{SiO}_2$	$\text{P}_2\text{O}_5$	$\text{TiO}_2$	$\text{Fe}_2\text{O}_3$
Content, % by weight.	<3.0	55.03	40.11	<0.1	<0.1	<1.00

**Table 4. Chemical composition of limestone**

Component	CaCO <sub>3</sub>	SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub> +Al <sub>2</sub> O <sub>3</sub>
Content, % by weight.	97.5	0.43	0.10

**Table 5. Chemical composition of mill scale in terms of pure elements**

Component	Fe	Cr	Mn	Ca	P	Ti
Content, % by weight.	77.9	1.00	0.22	0.25	0.020	0.018

**Table 6. Parameters and chemical composition of hard coal**

Component	Content, % by weight.
Moisture, as received basis, W <sup>r</sup>	15.1
Volatile, as received basis, V <sup>r</sup>	35.06
Ash, as received basis, A <sup>r</sup>	4.84
SiO <sub>2</sub> , as received basis	2.62
Al <sub>2</sub> O <sub>3</sub> , as received basis	0.835
Fe <sub>2</sub> O <sub>3</sub> , as received basis	0.271
P <sub>2</sub> O <sub>5</sub> , as received basis	0.012
CaO, as received basis	0.228
S, as received basis	0.283

The furnace used to produce alloy of iron, silicon and chromium according to the embodiment is equipped with a 12 MVA three-phase transformer, a round bath, three self-baking electrodes with a diameter of 900 mm, a low hood with five windows intended for the introduction of the batch mixture using buckets of loading machines and one tapping hole intended for periodic tapping of the smelting products, operating in continuous mode with an active power of 7.8 MW and an electrode current in the range of 32-35 kA.

Five alloy samples with different compositions (P1-P5) were prepared using different amounts of raw materials. Raw material and electricity consumption rates for alloy production are included in Table 7.

**Table 7. Raw material consumption rates for the production of the alloy according to the invention**

Item number	Ingredient consumption	u.m.	P1	P2	P3	P4	P5
1	High carbon ferrochrome	kg/t	380	379	397	435	438
2	Quartzite	kg/t	1650	1617	1545	1648	1574
3	Hard coal	kg/t	1352	1390	1294	1345	1320
4	Wood chips	kg/t	342	314	299	315	315
5	Mill scale	kg/t	-	8	12	5	5
6	Limestone	kg/t	100	280	190	80	120
7	Ceramic waste	kg/t	25	50	120	55	-
8	Electrode paste	kg/t	45	41	39	41	40
9	Electricity consumption	kWh/t	7341	7104	7342	7150	7103
10	Daily capacity of the furnace	t	23.5	25.1	24.4	25.4	26

The chemical composition of the samples of the resulting alloy (P1-P5) along with the comparative samples (PP1-PP5) prepared for the reduction potential test are shown in Table 8.

**Table 8. Chemical composition of the alloy from the embodiment and for comparative samples**

Component	P1	P2	P3	P4	P5	PP1	PP2	PP3	PP4	PP5
	Alloy samples according to the invention					Comparative samples				
Cr	21.52	21.69	22.79	23.16	24.02	26.34	26.51	22.86	34.1	26.58
Si	66.53	63.46	61.94	65.09	63.29	59.96	56.86	60.93	47.85	60.48
Fe	10.2	10.72	10.64	10.33	10.96	7.47	6.48	10.58	17.21	6.21
Al	0.596	1.264	3.066	1.01	0.154	6.112	5.899	1.076	0.11	4.114
Ca	0.598	2.059	1.046	0.318	0.61	0.024	4.004	4.041	0.008	2.011
C	0.018	0.015	0.008	0.011	0.019	0.006	0.022	0.008	0.011	0.006
Cr/Fe	2.11	2.02	2.14	2.24	2.28	3.53	4.09	2.16	1.98	4.28

The resulting alloy grades in the embodiment with the chemical composition indicated in Table 8 were used in the reduction of chromite ore with the chemical composition indicated in Table 9 in the production of FeCr LC.

**Table 9. Chemical composition of chrome ore concentrate**

Component	Cr <sub>2</sub> O <sub>3</sub>	FeO	SiO <sub>2</sub>	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	P	S	TiO <sub>2</sub>	NiO	MnO	V <sub>2</sub> O <sub>5</sub>	ZnO
% by weight.	51.78	16.41	3.38	0.92	15.77	10.01	0.002	0.008	0.143	0.17	0.17	0.13	0.043

As a result of using the alloys according to the embodiment in the reduction of chromite ore, a FeCr LC alloy was obtained with the chemical composition cited in Table 10 and a waste slag with the chemical composition presented in Table 11. In addition, Tables 10 and 11 show the results of chromite ore reduction for comparative alloys.

**Table 10. Summary results of FeCr LC chemical analysis**

Alloy grade	Chemical composition of FeCr LC [% wt.]					
	Cr	Fe	Si	Ca	Al	C
P1	68.57	29.12	1.19	0.0125	0.0135	0.012
P2	68.61	29.07	1.11	0.0165	0.0183	0.011
P3	68.01	28.95	1.6	0.0355	0.0666	0.014
P4	68.32	29.17	1.17	0.0183	0.0119	0.012
P5	67.94	29.63	1.1	0.035	0.053	0.012
PP1	65.69	31.13	1.83	0.0643	0.0207	0.015
PP2	66.08	31.57	1.07	0.0279	0.0406	0.017
PP3	67.78	29.09	1.78	0.0741	0.0327	0.014
PP4	64.77	32.75	0.63	0.0496	0.0678	0.015
PP5	67.72	29.46	1.51	0.0346	0.0515	0.018

**Table 11. Summary results of chemical analysis of FeCr LC slag**

Alloy grade	Chemical composition of FeCr LC slag [% wt.]					
	Cr <sub>2</sub> O <sub>3</sub>	FeO,	SiO <sub>2</sub>	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>
P1	1.35	0.204	27.85	36.72	8.38	24.63
P2	1.79	0.358	26.46	30.12	6.81	33.71
P3	2.03	0.382	25.89	30.1	6.52	34.32
P4	0.691	0.143	29.51	36.43	8.89	23.82
P5	1.381	0.353	29.65	38.08	8.93	20.5
PP1	7.35	0.131	31.07	32.21	8.55	19.63
PP2	8.16	0.319	27.17	29.29	6.48	26.66
PP3	2.09	0.895	32.39	36.57	6.12	21.41
PP4	14.22	0.238	21.37	27.86	6.42	27.71
PP5	3.98	1.002	27.63	36.06	8.65	21.91

Based on the performed mass balance of the FeCr LC production process, the chromium yield from the ore to FeCr LC was calculated, which, together with the residual content of Cr<sub>2</sub>O<sub>3</sub> in the slag is a determinant of the reduction potential of the obtained alloys in the metallothermic

process. Method of calculating the % chromium yield in the production of FeCr LC is defined as follows:

$$U_{\%Cr} = \frac{G_{Cr} - G_{Cr \text{ Crosimax}}}{G_{Cr \text{ chromium ore}}} \cdot 100\%$$

where:

$U_{\%Cr}$  – chromium yield by metallothermic reduction [%]

$G_{Cr}$  – mass of chromium obtained in the alloy, after the metallothermic reduction reaction [g]

$G_{Cr}$  – mass of chromium in the Crosimax alloy used as reductant [g]

$G_{Cr \text{ chromite ore}}$  – mass of chromium in chromite ore [g]

Table 12 shows the results of chromium yield (reduction potential) using different alloy grades.

**Table 12. Chromium yield (reduction potential)**

Sample No.	Cr yield [%]
P1	95.75
P2	95.73
P3	95.19
P4	96.17
P5	93.68
PP1	88.2
PP2	89.46
PP3	91.99
PP4	88.26
PP5	92.61

From the test results cited in Tables 10-12, it can be seen that when alloys comprising more than 4 % wt. aluminium and more than 3 % wt. calcium are used, the results of the reduction potential are lower than when using the contents of these elements in the range of 0.5 to 4.0 aluminium, 0.31 to 2.0 calcium. In addition, it was shown that at calcium contents of less than 0.01 wt. and aluminium contents of less than 0.5 % wt., lower reduction potential results are obtained than for alloys with compositions according to the invention. The best reduction potential results were achieved for samples P1, P2 and P4, in which the amount of calcium ranges from 0.3 to 2.1 % wt., and the amount of aluminium from 0.5 to 3.1 % wt.

The result of increasing the mass content of silicon in the alloy in the range of 61-68 % wt. was the increase of the reduction potential to chromium ore in the FeCr LC production process as measured by the Cr yield to the alloy and the residual mass content of  $\text{Cr}_2\text{O}_3$  in the waste slag. A comparison of the behaviour, including the reducing capacity of Fe-Si-Cr alloys, as reductants in the production of FeCr LC showed that the greatest influence on the reducing capacity of the alloys studied is the addition of silicon. The applied Ca and Al additives, at this concentration level, did not significantly change the reduction capacity of these alloys, although the increase in the content of Al to 3.1 wt.% and Ca to 1.2 wt.% had a positive effect on increasing the efficiency of the reduction process compared to other Fe-Si-Cr alloys, but not as effectively as Si. An unexpected effect was a decrease in the reduction potential of the alloy with an increase in the mass content of Al above 4% and Ca above 2%, both of which have a higher affinity for oxygen than Si.

### Claims

1. A complex multi-component alloy of iron, silicon and chromium comprising, as a percentage by weight of the total weight of the alloy: not less than 21.0 chromium, not less than 61.0 silicon, 0.5 to 4.0 aluminium, 0.3 to 2.1 calcium, not more than 0.02 carbon, the rest being iron and traces of impurities, with a ratio of chromium to iron of not less than 2.0.
2. The complex multi-component alloy of iron, silicon and chromium according to claim 1, **characterised by the fact that** it comprises chromium in an amount of 21.0 to 28.0 % wt. relative to the total weight of the alloy, preferably 21.5 to 25.5 % wt.
3. The complex multi-component alloy of iron, silicon and chromium according to claim 1, **characterised in that** it comprises silicon in an amount of 61.0 to 70.0 % wt. relative to the total weight of the alloy, preferably 63.5 to 68.0 % wt.
4. The complex multi-component alloy of iron, silicon and chromium according to claim 1, **characterised in that** it comprises calcium in an amount of 0.5 to 1.2 % wt. relative to the total weight of the alloy.
5. The complex multi-component alloy of iron, silicon and chromium according to claim 1, **characterised in that** it comprises aluminium in an amount of 0.5 to 3.1 % wt. relative to the total weight of the alloy, preferably 0.5 to 1.3 % wt.
6. A method of producing a complex multi-component iron alloy of iron, silicon and chromium according to any of the claims 1 to 5, **characterised in that** the oxides of  $\text{SiO}_2$ ,  $\text{CaO}$ ,  $\text{FeO}$ ,  $\text{Al}_2\text{O}_3$  are reduced carbothermically in the presence of chromium and iron derived from high-carbon ferrochrome in an arc-resistance furnace, and then subjected to post-furnace decarburisation refining.
7. Use of a complex multi-component alloy of iron, silicon and chromium according to any of the claims 1 to 5 for the reduction of  $\text{Cr}_2\text{O}_3$  comprised in chromium ore in the process of obtaining low carbon ferrochrome.

# INTERNATIONAL SEARCH REPORT

International application No  
**PCT/PL2023/050068**

**A. CLASSIFICATION OF SUBJECT MATTER**  
**INV. C22C28/00 C22B5/00**  
**ADD. C22B1/00**

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
**C22B C22C**

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

**EPO-Internal**

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
<b>A</b>	<b>US 11 542 578 B2 (ELKEM MATERIALS [NO])</b> <b>3 January 2023 (2023-01-03)</b> <b>col. 2-4, 9</b> -----	<b>1-7</b>
<b>A</b>	<b>WO 2018/068066 A2 (BEYLEFELD JACQUES [ZA])</b> <b>12 April 2018 (2018-04-12)</b> <b>0005, 0008, 0020, 0021, 0070</b> -----	<b>1-7</b>
<b>A</b>	<b>CN 207 973 790 U (FENGZHEN CITY XINYE METALLURGY CO LTD)</b> <b>16 October 2018 (2018-10-16)</b> <b>the whole document</b> -----	<b>1-7</b>

Further documents are listed in the continuation of Box C.

See patent family annex.

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Date of the actual completion of the international search

Date of mailing of the international search report

**20 February 2024**

**29/02/2024**

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

**PCT/PL2023/050068**

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