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Diagram of the phase composition of the Fe - Si - Al system and its isothermal sections

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The diagram of the phase composition of the Fe – Si – Al system and its isothermal sections within the temperature range of 300-3000 K are plotted. Thermodynamic data of Fe_3Si, Fe_2Si, Fe_5Si_3, FeSi, $FeSi_2$ silicides, FeAl, $FeAl_2$, $FeAl_3$, Fe_2Al_5 aluminides and Al_2Fe_2Si, Al_3FeSi, Al_2FeSi, $Al_{11}Fe_3Si_6$, $Al_{14}Fe_3Si_3$ ternary compounds are used. The diagram consists of 20 elementary triangles of coexisting phases, the number of which decreases to 6 at the temperature 1900 K and more. At the temperatures above 2900 K, the equilibrium of the condensed phases is disturbed due to the transition of such metal components of the low-melting aluminum corner of the diagram as Al, Al_2, Fe, Si and Si_2, leading to losses of valuable elements, to the gas phase.
Crystallization fields are assessed by calculating the squares of elementary triangles, as well as the prevalence of each compound in the phase space by taking into account its participation in the construction of adjacent triangles. Using our own method, the mathematical model of the diagram was created in the form of equations for dependence of the amount of the formed phase on the chemical composition of examined metal. The computer program was developed based on the established equations. The examples of calculation of the phase composition of two grades of ferrosilicon FeSi65 and FeSi75, which are the most common in metallurgy, as well as two grades of ferrosilicoaluminum FeSi45Al15 and FeSi55Al20 are presented. Correspondence of the calculated and instrumentally found phase composition of the industrial metal has been revealed. Presence of mathematical models makes it possible to accelerate selection of the metal with the required composition and properties from the set of planned for smelting.
Key words: iron, silicon, aluminum, diagram, phases, thermodynamics, mathematical model.
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Introduction

The phase equilibrium diagram of the Fe – Si – Al system is a subject of permanent attention of researchers. This interest is connected with its necessity for analysis and improvement of technological processes in production of several metals. First of all, ferrosilicon can be mentioned here, which is widely used in metallurgy and is described be Fe – Si binary system in its main components. More strict requirements to steel quality set the task to produce ferrosilicon with extra low content of impurities (especially Al) [1]. The ternary diagram Fe – Si – Al is used for this purpose. It was also the base for creation of the complex ferroalloy ferrosilicoaluminum (FSA) [2, 3].

Fe – Al binary system arises especial interest, and not only because of ferroaluminium production on its base [4], but also due to possibility of Fe aluminides synthesis with special properties [5-7]. Si – Al system is considered as simple eutectic one, but it is also examined because it is often included in multi-component systems [8].

Technical literature describes the phase equilibrium diagrams Fe – Si – Al, but they varied noticeably one from another. Some sources does not take them into account at all [9]. The other ones suggest presence of such phases as Al_8Fe_2Si, Al_5FeSi, Al_4FeSi [10], $Al_3Fe_3Si_2$, $Al_{12}Fe_6Si_5$, $A_9Fe_5Si_5$, Al_3FeSi_2, $Al_{15}Fe_6Si_5$, Al_4FeSi [11], as well as Al_2Fe_2Si, Al_3FeSi, Al_2FeSi, $Al_{14}Fe_3Si_3$, $Al_{11}Fe_3Si_6$ [12]. It was planned to search them using the bases of thermodynamic data, which are conventional in the scientific community, not using separate investigations.

Determination of the kind and quality of forming phases for metal with required properties is considered as one of multiple destinations of the diagrams. Usually it is executed using the lever rule. However, analysis of metals variety is accompanied with simple but labour-intensive calculations. The mathematical model of the diagram was created in this work using the self method [13]; corresponding software was developed as well. When chemical composition is preset from the remote control panel, the computer provides metal phase composition in mass. %.

To examine the temperature relationship of phase composition, isothermal diagram sections were built within the temperature range 300–3000 K. It is noted that at the temperature 2900 K and higher, equilibrium of condensed phases is violated due to transition of metal components of the low-melting aluminum corner of the diagram in the gas phase. Thereby it is recommended not to overheat the melt and to operate with closed dry top throat for catching fumes and their return in the process.

So, the updated diagram of phase composition of the Fe – Si – Al system was built in this work, its isothermal sections are presented, mathematical model of the diagram

was created and corresponding software for numerical calculations was developed as well.

Methods and materials

Theoretical (calculating) methods of diagrams building was used in this work. The computer («Triangle») and thermodynamic-diagram methods, which have widely prevalence in researching practice [14-16], were applied. The self-developed balance method [13] was used for mathematical description of the phase composition diagram. Use of the described approach allows to delete restrictions for number of components under examination, because the mathematical model operates not only in 3D space (where diagrams are usually reflected), but also multidimensional space.

Obtained results and their analysis

The ternary system Fe – Si – Al consists of three binary partial systems: Fe – Si, Fe – Al and Si – Al. Presence of such silicides as Fe_3Si, Fe_5Si_3, FeSi, $FeSi_2$ and Fe_2Si was taken into account in the system Fe – Si. The system Fe – Al includes four Fe aluminides [17]: FeAl, $FeAl_2$, $FeAl_3$, Fe_2Al_5, which are accepted for diagram building. No binary compounds were found in the system Si – Al. The problem of presence of ternary compounds is especial one for the examined system; they are not taken into account in general in several researches [9]. Other authors suggested presence of such phases as Al_8Fe_2Si, Al_5FeSi, Al_4FeSi [10] or $Al_3Fe_3Si_2$, $Al_{12}Fe_6Si_5$, $A_9Fe_5Si_5$, Al_3FeSi_2, $Al_{15}Fe_6Si_5$, Al_4FeSi [11]. Information about five other compounds Al_2Fe_2Si, Al_3FeSi, Al_2FeSi, $Al_{14}Fe_3Si_3$, $Al_{11}Fe_3Si_6$ is presented in the data base of SGT complex [12]. As soon as these data were obtained by efforts of scientific teams from several countries and they are mutually agreed, they were used for diagram building.

It is recommended to show on the triangle of compositions (**Fig. 1**) all compounds which were found in the technical literature, when using thermodynamic-diagram method. It will provide general picture of phase building of the system.

Analysis of phase chemical composition helps to reveal that $FeSi_2$ and $Al_{11}Fe_3Si_6$ have equal Fe to Si relation (0.994), and ternary compound $Al_{11}Fe_3Si_6$ can be considered as the product of chemical reaction

$$3FeSi_2 + 11Al = Al_{11}Fe_3Si_6 \quad (1)$$

where $\Delta G_{298,15K}$ = - 52.44 KJ/mol. It allows to make the line $FeSi_2$ – Al on the diagram. The line FeSi – Al includes three ternary compounds: Al_2FeSi, Al_3FeSi and $Al_{14}Fe_3Si_3$.

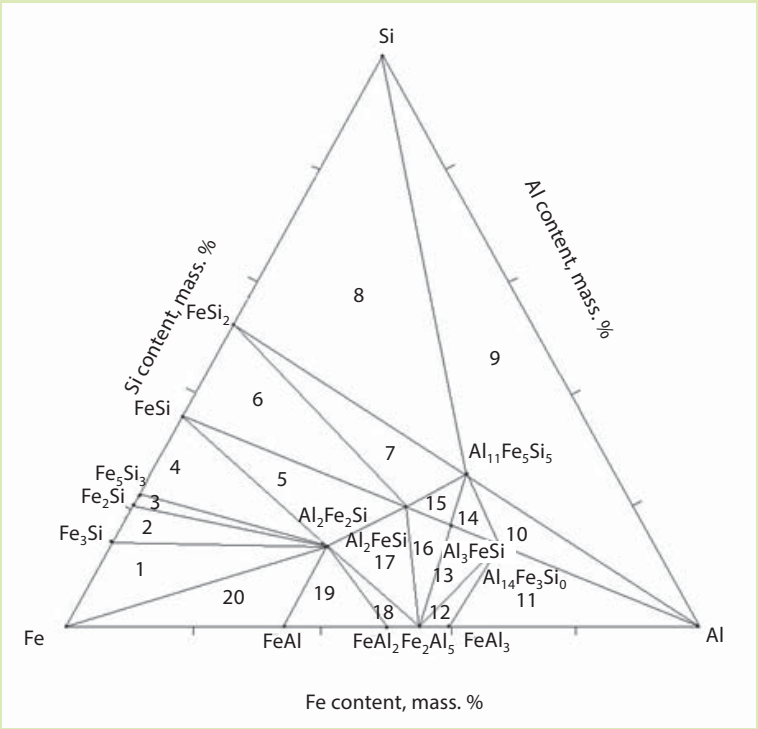

Fig. 1. Diagram of phase composition of Fe – Si – Al system

They and FeSi have also equal Fe to Si relation (1.988), what stipulates their presence on the line FeSi – Al.

Coupled co-existance of other phases was evaluated by calculation of Gibbs energy variation for corresponding reactions. So, in the quadrangle of substance s FeSi – $FeSi_2$ – Al_2FeSi – $Al_{11}Fe_3Si_6$ (Fig. 1), the reaction written along its diagonals

$$55FeSi + 10Al_{11}Fe_3Si_6 = 30FeSi_2 + 55Al_2FeSi \quad (2)$$

Passes in straight direction with $\Delta G_{298,15K}$ = - 279.31 KJ/mol. As a result, its products were assessed as co-existing ones, and they were connected by direct line on the diagram. Conduction of this procedure with other compounds allowed to make complete triangulation of the studied system with establishment of 20 elementary triangles of co-existing phases in it (Fig. 1).

Table 1 presents the numbers of all triangles, their squares (S), equations for determination of number of phases depending on chemical composition of initial metal (Fe_0, Si_0, Al_0), as well as prevalence of each phase in the field of the diagram (W) according to [16].

Triangle No. 8 $FeSi_2$ – $Al_{11}Fe_3Si_6$ – Si occupies maximal square in the examined system, while triangle No. 3 Al_2Fe_2Si – Fe_2Si – Fe_5Si_3 occupies minimal square. The phase $Al_{11}Fe_3Si_6$ has the most existence possibility (W) in the system Fe – Si – Al, and the phase Fe_2Si is characterized by minimal existence possibility.

Temperature relationship of phase composition was studied using "Triangle" program via building isothermal

Table 1. **Characteristics of Fe – Si – Al system**

Triangle No.	S, sq. un.	Equations	Phases	W, %
1	0.02133	$Al_2Fe_2Si = 3.5907 \cdot Al_0$ $Fe_3Si = 6.9638 \cdot Si0 - 3.6260 \cdot Al_0$ $Fe = Fe_0 - 5.9638 \cdot Si_0 + 1.0353 \cdot Al_0$	Fe	3.34
2	0.00994	$Fe_3Si = 3.2061 \cdot Fe_0 - 13.9458 \cdot Si_0 + 0.0032 \cdot Al_0$ $Al_2Fe_2Si = 3.5906 \cdot Al_0$ $Fe_2Si = - 2.5061 \cdot Fe_0 + 14.9458 \cdot Si_0 - 2.5938 \cdot Al_0$	Si	15.54
3	0.00311	$Al_2Fe_2Si = 3.5906 \cdot Al_0$ $Fe_2Si = 7.5016 \cdot Fe_0 - 24.8608 \cdot Si_0 - 2.5847 \cdot Al_0$ $Fe_5Si_3 = - 6.5016 \cdot Fe_0 + 25.8608 \cdot Si_0 - 0.0059 \cdot Al_0$	Al	10.88
4	0.02070	$Al_2Fe_2Si = 3.5906 \cdot Al_0$ $Fe_5Si_3 = 3.2548 \cdot Fe_0 - 6.4727 \cdot Si_0 - 3.3676 \cdot Al_0$ $FeSi = - 2.2548 \cdot Fe_0 + 7.4727 \cdot Si_0 + 0.777 \cdot Al_0$	Fe_3Si	1.54
5	0.01895	$Al_2Fe_2Si = 3.4697 \cdot Fe_0 - 6.9 \cdot Si_0 + 0.0008 \cdot Al_0$ $FeSi = - 0.0003 \cdot Fe_0 + 2.989 \cdot Si_0 - 1.5558 \cdot Al_0$ $Al_2FeSi = - 2.4694 \cdot Fe_0 + 4.911 \cdot Si_0 + 2.555 \cdot Al_0$	FeSi	5.35
6	0.02878	$FeSi = 3.0047 \cdot Fe_0 - 2.9868 \cdot Si_0 - 1.5551 \cdot Al_0$ $Al_2FeSi = 2.5555 \cdot Al_0$ $FeSi_2 = - 2.0047 \cdot Fe_0 + 3.9868 \cdot Si_0 - 0.0004 \cdot Al_0$	$FeSi_2$	11.71
7	0.01938	$Al_2FeSi = 4.9397 \cdot Fe_0 - 4.9101 \cdot Si_0 - 0.001 \cdot Al_0$ $FeSi_2 = 0.1825 \cdot Fe_0 + 1.8126 \cdot Si_0 - 1.1324 \cdot Al_0$ $Al_{11}Fe_3Si_6 = -4{,}1222 \cdot Fe_0 + 4{,}0975 \cdot Si_0 + 2{,}1334 \cdot Al_0$	FeAl	2.43
8	0.10154	$FeSi_2 = 2.006 \cdot Fe_0 - 1.1328 \cdot Al_0$ $Al_{11}Fe_3Si_6 = 2.1326 \cdot Al_0$ $Si = - 1.006 \cdot Fe_0 + 1 \cdot Si_0 + 0.0002 \cdot Al_0$	Fe_2Al_5	2.45
9	0.10090	$Al_{11}Fe_3Si_6 = 3.7764 \cdot Fe_0$ $Si = - 1.0056 \cdot Fe_0 + 1 \cdot Si_0$ $Al = - 1.7708 \cdot Fe_0 + 1 \cdot Al_0$	Al_3FeSi	1.51
10	0.01382	$Al_{11}Fe_3Si_6 = - 3.7818 \cdot Fe_0 + 7.5157 \cdot Si_0$ $Al = -2.7395 \cdot Fe_0 + 0.9632 \cdot Si_0 + 1 \cdot Al_0$ $Al = -2.7395 \cdot Fe_0 + 0.9632 \cdot Si_0 + 1 \cdot Al_0$ $Al_{14}Fe_3Si_3 = 7.5213 \cdot Fe_0 - 7.4789 \cdot Si0$	Al14Fe3Si3	4.04
11	0.02516	$Al = -1.4491 \cdot Fe_0 - 1.601 \cdot Si_0 + 1 \cdot Al_0$ $Al_{14}Fe_3Si_3 = 7.4682 \cdot Si_0$ $FeAl_3 = 2.4491 \cdot Fe_0 - 4.8672 \cdot Si_0$	$Al_{11}Fe_3Si_6$	18.74
12	0.00286	$Al_{14}Fe_3Si_3 = 7.4683 \cdot Si_0$ $FeAl_3 = - 12.2371 \cdot Fe_0 - 21.0921 \cdot Si_0 + 10.1342 \cdot Al_0$ $Fe_2Al_5 = 13.2371 \cdot Fe_0 + 14.6238 \cdot Si_0 - 9.1342 \cdot Al_0$	Al_2FeSi	6.93
13	0.00655	$Al_{14}Fe_3Si_3 = - 5.6342 - 2{,}2432 \cdot Si_0 + 4.666 \cdot Al_0$ $Fe_2Al_5 = 2.2068 \cdot Fe_0 - 4.388 \cdot Si_0 + 0.0006 \cdot Al_0$ $Al_3FeSi = 4.4274 \cdot Fe_0 + 7.6312 \cdot Si_0 - 3.6666 \cdot Al_0$	Al_2Fe_2Si	9.44
14	0.00323	$Al_{11}Fe_3Si_6 = - 3.7795 \cdot Fe_0 + 7.5148 \cdot Si_0 - 0.0008 \cdot Al_0$ $Al_{14}Fe_3Si_3 = -5.2616 \cdot Fe_0 - 2.9841 \cdot Si_0 + 4.6661 \cdot Al_0$ $Al_3FeSi = 10.0411 \cdot Fe_0 - 3.5307 \cdot Si_0 - 3.6653 \cdot Al_0$	$FeAl_2$	1.03
15	0.00316	$Al_{11}Fe_3Si_6 = - 3.778 \cdot Fe_0 + 7.5156 \cdot Si_0 - 0.0022 \cdot Al_0$ $Al_3FeSi = - 0.9852 \cdot Fe_0 - 9.7843 \cdot Si_0 + 6.1131 \cdot Al_0$ $Al_2FeSi = 5.7632 \cdot Fe_0 + 3.2687 \cdot Si_0 - 5.1109 \cdot Al_0$	$FeAl_3$	2.19
16	0.00641	$Al_3FeSi = -7.3772 \cdot Fe_0 + 2.9314 \cdot Si_0 + 6.1094 \cdot Al_0$ $Al_2FeSi = 6.1712 \cdot Fe_0 + 2.4569 \cdot Si_0 - 5.1107 \cdot Al_0$ $Fe_2Al_5 = 2.206 \cdot Fe_0 - 4.3883 \cdot Si_0 + 0.0013 \cdot Al_0$	Fe_2Si	1.02
17	0.01196	$Al_2FeSi = - 2.4704 \cdot Fe_0 + 5.8909 \cdot Si_0 + 2.0458 \cdot Al_0$ $Fe_2Al_5 = 0.0002 \cdot Fe_0 - 3.5119 \cdot Si_0 + 1.8279 \cdot Al_0$ $Al_2Fe_2Si = 3.4706 \cdot Fe_0 - 1.380 \cdot Si_0 - 2.8737 \cdot Al_0$	Fe_5Si_3	1,86
18	0.00354	$Fe_2Al_5 = - 8.8381 \cdot Fe_0 + 17.5636 \cdot Si_0 + 9.1474 \cdot Al_0$ $Al_2Fe_2Si = 6.8965 \cdot Si_0$ $FeAl_2 = 9.8381 \cdot Fe_0 - 23.4601 \cdot Si_0 - 8.1474 \cdot Al_0$	-	-
19	0.00972	$Al_2Fe_2Si = 6.8965 \cdot Si_0$ $FeAl_2 = - 1.9656 \cdot Fe_0 - 0.0031 \cdot Si_0 + 4.0694 \cdot Al_0$ $FeAl = 2.9656 \cdot Fe_0 - 5.8934 \cdot Si_0 - 3.0694 \cdot Al_0$	-	-
20	0.02142	$Al_2Fe_2Si = 6.8965 \cdot Si_0$ $FeAl = - 5.8977 \cdot Si_0 + 3.0703 \cdot Al_0$ $Fe = 1 \cdot Fe_0 + 0.0012 \cdot Si_0 - 2.0703 \cdot Al_0$	-	-

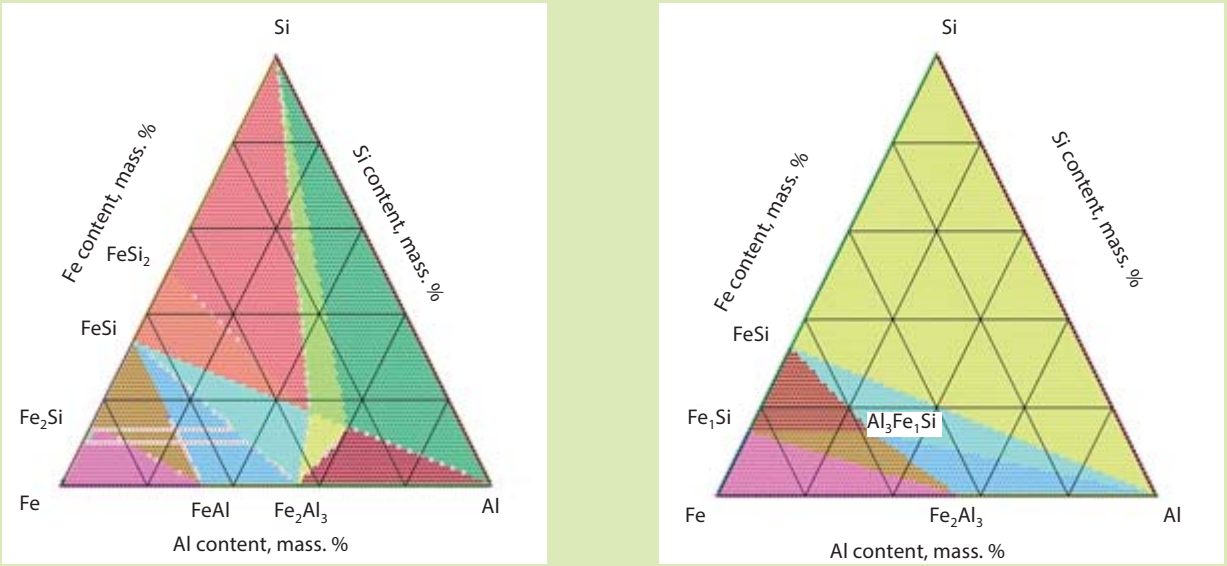

Fig. 2. Isothermal sections of the diagram of phase composition of Fe – Si – Al system at 300 K (*a*) and 1900 K (*b*)

sections of the Fe – Si – Al diagram with 200 K interval. Several obtained data are presented on the **Fig. 2**.

Only stable binary compounds Fe_2Al_5, FeSi, Fe_3Si, as well as the ternary compound Al_2Fe_2Si, are saved at the increased temperatures (1900 K). Within the temperature range 1900-2700 K the phase composition of the Fe – Si – Al system stabilizes, and it includes 6 elementary triangles of co-existing phases. At the temperatures 2900 K and higher, equilibrium of condensed phases is violated due to transition of low-melting aluminium diagram corner in the gas phase of metal components.

Fig. 3 displays the temperature relationship between metal phase composition in the aluminium corner, containing 90Al, 5Si and 5Fe (mass. %), which is built on semi-logarithmic coordinates. The condensed phases are marked with "c" index, e.g. Al (c), while gas phases don't have such index.

It can be seen, that increased amount of Al, Al_2, Fe, Si and Si_2, which leads to losses of valuable elements of melting metal, can be observed outside the temperature 2800 K in the gas phase.

The concluded equations (see Table 1) allow to calculate metal phase composition in the total area of Fe – Si – Al diagram. It is shown in the **Table 2** on the example of two grades of ferrosilicon (FS75 and FS65) and two grades of ferrosilicoaluminium FeSi45Al15 and FeSi55Al20

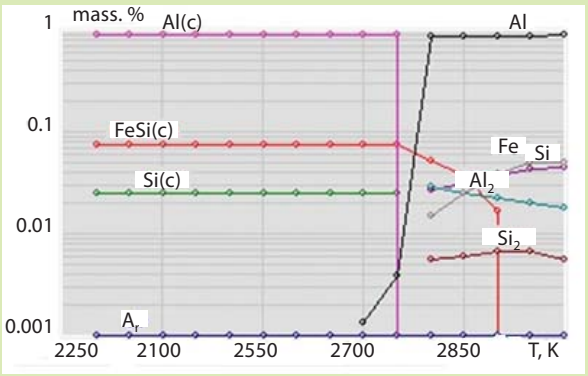

Fig. 3. Relationship between metal phase composition in the aluminium corner, containing 90Al, 5Si and 5Fe (mass. %), and temperature

Conclusions

Both grades of ferrosilicon FS75 and FS65 are located in the triangle field Al_3FeSi – $FeSi_2$ – Si with their phase composition. $FeSi_2$ and Si, which were instrumentally revealed in industrial metal ingots [18], dominate in the structure in this case. Role of loboite ($FeSi_2$) in ferrosilicon and ferrosilicoaluminium is an especial one. It is connected with decomposition (dispersion) of these metals during their storage due to transition of high-temperature form of Fe disilicide in low-temperature form, accompanied by volume increase by 17 %. In correspondence with the diagram (Fig. 1), it can be avoided via work in the triangle $Al_{11}Fe_3Si_6$ – Si – Al or below the line FeSi – Al, where this phase is absent. However, the authors of [18] noted that loboite is not considered as a trigger of ferrosilicon dispersion, while the observing process can be connected with presence of excessive phases based on aluminium, calcium, ferrum, phosphorus, which are instable in wet atmosphere. This problem needs further investigation.

Thereby, the phase composition diagrams for Fe – Si – Al system were built after calculation, their isothermal sections were presented and the mathematical model was

Table 2. **Compositions of ferroalloys**

No.	Alloy	Alloy composition, mass. %					
		Chemical			Phase		
		Fe	Si	Al	Si	$FeSi_2$	$Al_{11}Fe_3Si_6$
1	FeSi75	24	75	1	50.86	47.01	2.13
2	FeSi65	33	65	2	31.8	63.93	4.27
3	FeSi45Al15	40	45	15	4.76	63.25	31.99
4	FeSi55Al20	25	55	20	29.85	27.49	42.66

created. Applicability of this model for analysis of phases forming in silicon and silicon-aluminium ferroalloys was shown.

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