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Phase constituents modeling in hybrid multi-component high-boron alloy

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The present work is focused on analyzing the thermodynamically equilibrium path of structure formation in a multi-component Fe-W-Mo-Cr-Ti-Mn-Si alloy of tribotechnical purpose, containing 0.72 wt. % C and 2.75 wt. % B is analyzed. Computer simulation of the crystallization process was performed using the program "Thermo-Calc Software". It was found that the alloy belongs to the hyper-eutectic compositions, since its crystallization begins at 1472 ºC with the formation of primary boride WB and carbide TiC. After a series of subsequent eutectic reactions (in the range of 1126-923 ºC) and solid-phase transformations, a set of equilibrium phases is formed in the alloy, which at room temperature consists of borides WB, MoB, Fe_2B, Cr_4B, Mn_4B, carbides TiC, M_7C_3, and ferrite. The total volume fraction of borides and carbides is 45.05 vol. %. A comparison of the obtained data with the results of the study of the manufactured alloy showed that the simulation with "Thermo-Calc Software" provides satisfactory adequacy in predicting the structure in the alloys of the selected alloying system.

Keywords: multi-component cast iron, boride, carbide, microstructure, modelling.

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Introduction

Tribotechnical alloys are designed for operation under heavy friction and wear. Their chemical composition is constantly being upgraded to meet the growing demands of the intensification of equipment usage. There are different approaches to the selection of chemical composition and heat treatment of wear-resistant alloys, most of which are aimed at obtaining a heterophase structure with a high volume fraction of hard phase components which are compounds of transition metals with C, N, B [1, 2]. White alloy cast irons are widely used in various fields of related to the grinding, processing, and transportation of minerals (mining, metallurgy, heat energy production, etc.) [3, 4]. This group of alloys includes multi-component cast irons alloyed with a complex of carbide-forming elements (W, Mo, V, Cr), taken in equal proportions, forming a multiphase structure with a set of different carbides (MC, M_2C, M_6C, M_7C_3), distributed within austenitic-martensitic matrix [5, 6]. Due to this structure, multi-component cast irons performed an improved service life in terms of metallurgical and cement equipment [7]. An alternative approach is the replacement of carbon in cast iron with boron, allowing the formation of borides and carboborides, which have an advantage over carbides in hardness and wear resistance [8-10]. In this case, boron is introduced into unalloyed or low-chromium cast iron [11], as well as to high-speed steels [12, 13].

In [14], the authors proposed a new "hybrid" approach, which consists in combining the multi-component alloying principle with adding an increased amount of boron and such a strong carbide-forming element as titanium [15]. This approach aims at the formation of

complex carboboride phases with advanced tribological characteristics [16]. The implementation of new ideas should be based on computer modelling, which is now widely used at the preliminary stage of the alloy design [17, 18]. This work is devoted to forecasting the phase-structural state on the basis of thermo-dynamic modelling performed by the "Thermo-Calc Software".

I. Results and Discussion

The study material was a high-boron multi-component alloy of chemical composition (wt.%): 0.72 C; 2.75 B; 0.90 Mn; 1.10 Si; 10.35 Cr, 5.05 W, 5.57 Mo, 2.60 Ti, Fe - residue. The technology of manufacturing this alloy, the method of its study and phase-structural state are described in detail in [14]. According to Figure 1, the alloy is hyper-eutectic since its microstructure consists of (a) coarse primary precipitates of complex-alloyed carboboride $M_2(B,C)_5$ (where M=W,Mo,Ti,Fe), (b) dispersed titanium carboboride Ti(C,B), and (c) two types of eutectic which are "Chinese Script"-shaped (based on carboboride $M_2(B,C)_5$) and "Rosette"-shaped (based on carboboride $(Fe,Cr,Mo,W,Mn)_7(B,C)_3$) [14]. The metal matrix of the alloy is predominantly ferritic with a small amount of pearlite. The sequence of formation of phase-structural components during crystallization of this alloy under equilibrium conditions was studied by thermodynamic modelling using the "Thermo-Calc Software" [18].

Figure 2 shows the equilibrium diagram "M-Boron", where M includes C, Mn, Si, W, Mo, Ti, Fe in amounts corresponding to the chemical composition of the alloy. The location of the alloy on the diagram is indicated by a dotted line. The solidification path, presented in Figure 3, is determined by the change in the free Gibbs energy of the individual phases, shown in Figure 4.

According to the diagram, crystallization in the alloy begins at 1472 °C by precipitation from the liquid of primary inclusions of tetragonal tungsten boride WB by reaction (1):

$$Liquid(L) \rightarrow L + WB(1472 - 1462\,°C) \quad (1)$$

The equilibrium boron content in WB is 94.45 wt.% (Table 1)). Along with reaction (1), at 1460 °C, the cubic carbide TiC begins to crystallize (reaction (2)), and in a fairly wide range of temperatures (up to 1126 °C) these two phases are formed simultaneously with an increase in their total volume fraction to about 6 vol. %

$$L + WB \rightarrow L + WB + TiC\ (1460 - 1126\,°C) \quad (2)$$

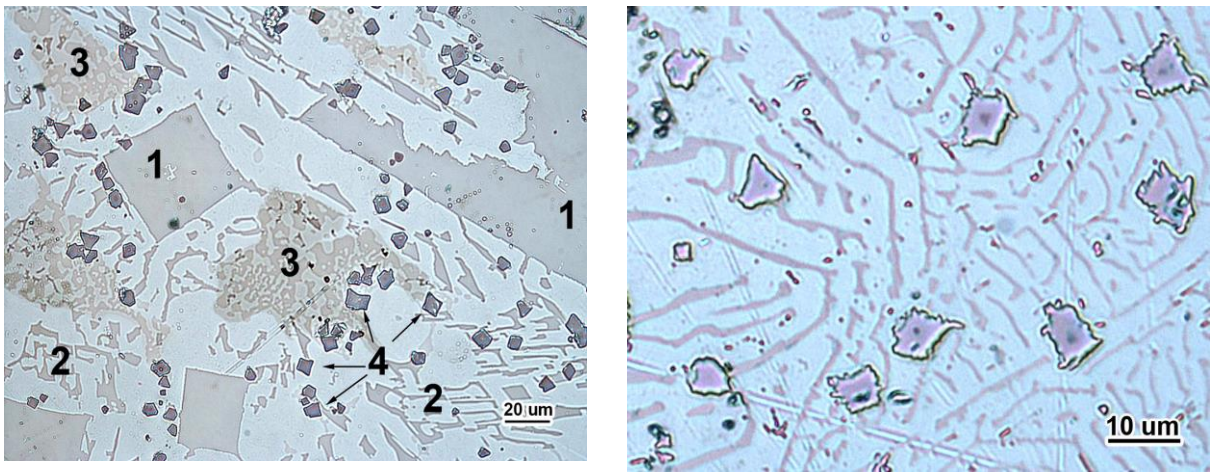

Fig. 1. Microstructure of the experimental alloy: (a) general view, (b) precipitates of titanium carboboride on the background of the "Chinese Script" eutectic (1 – primary precipitates $M_2(B,C)_5$, 2 – "Chinese Script" eutectic, 3 – $(Fe,Cr,Mo,W,Mn)_7(B,C)_3$-based eutectic, 4 – carboboride Ti(C,B)).

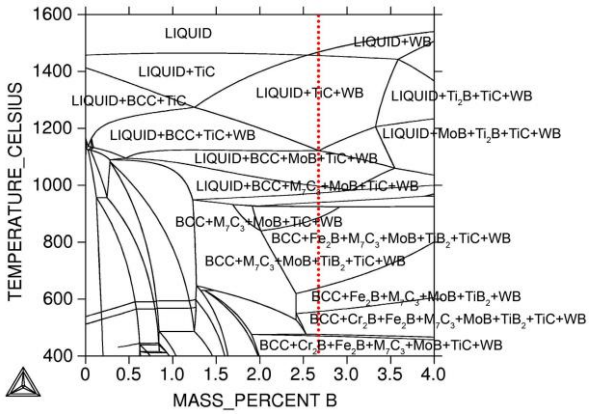

Fig. 2. The diagram "M-Boron" (where M includes C, Mn, Si, W, Mo, Ti, Fe in the above amounts).

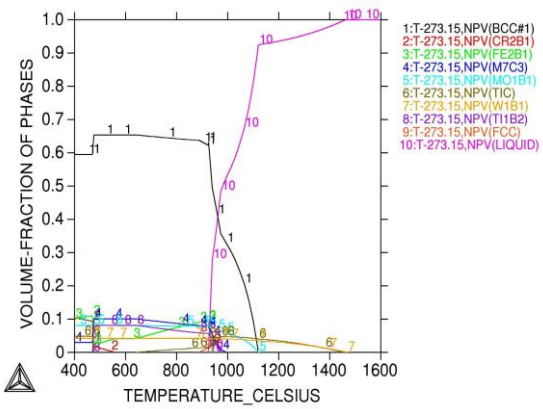

Fig. 3. Crystallization path in alloy.

The precipitation of WB and TiC leads to the depletion of the liquid by the corresponding chemical elements (C, B, W, Ti), which causes a sequence of eutectic reactions with the crystallization of αFe in the form of ferrite (BCC - Body Centred Cubic). Under crystallization, silicon and manganese are rejected by the growing carbides and borides into the residual liquid which further transforms into ferrite. As a result, the concentration of Si and Mn in ferrite increases up 1.59 wt.% and 1.70 wt. %, accordingly, which is more than 1.5 times the content of these elements in the alloy. The equilibrium concentration of other elements in ferrite is very low, especially for carbon and titanium.

In the temperature range 1126-1000 ºC, the eutectic (BCC + MoB (89.90 wt.% Mo)) is formed by reaction (3) (in parallel, the slow release of WB boride and TiC carbide continues):

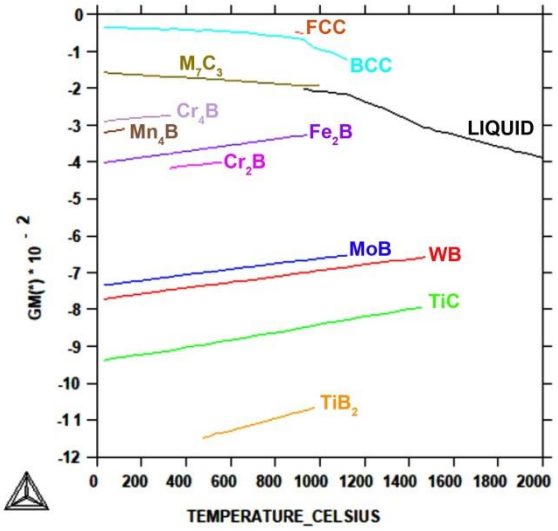

Fig. 4. Gibbs energy temperature dependences for different phases.

$$L + WB + TiC \rightarrow L + BCC + MoB + TiC + WB\ (1126 - 1000\ ^{\circ}C) \quad (3)$$

Precipitation of carbide-forming elements (Ti, W, Mo)-based carbides and borides with a total volume of about 17 vol. % and crystallization of 35 vol.% ferrite sharply increases the chromium content in the residual liquid at the end of the reaction (3). This encourages the formation of Cr-rich hexagonal carbide M_7C_3 (or rather $(Cr, Mn)_7C_3$, which contains 88.2 wt.% Cr and 2,81 wt.% Mn (Table 1)), which crystallizes in the temperature range of 1000-978 ºC by reaction (4), being involved in the eutectic "BCC + M_7C_3".

$$L + \mathbf{BCC} + MoB + TiC + WB \rightarrow L + BCC + M_7C_3 + MoB + TiC + WB\ (\mathbf{1000 - 978}\ ^{\circ}C) \quad (4)$$

As follows from Fig. 4, in the temperature range 978-475ºC it is titanium diboride TiB_2 that has the lowest Gibbs energy, so at 978ºC its crystallization by eutectic reaction (5) begins:

$$L + BCC + M_7C_3 + MoB + TiC + WB \rightarrow L + BCC + M_7C_3 + MoB + TiB_2 + TiC + WB(978 - 945\ ^{\circ}C) \quad (5)$$

Upon its completion, the liquid is much depleted in alloying elements; therefore the boron residue binds to iron to form boride Fe_2B during the eutectic reaction (6), which occurs in the range of 984-923ºC:

$$L + BCC + M_7C_3 + MoB + TiB_2 + TiC + WB \rightarrow L + BCC + Fe_2B + M_7C_3 + MoB + TiB_2 + TiC + WB(945 - 923\ ^{\circ}C) \quad (6)$$

Table 1

Equilibrium phase chemical composition of the experimental alloy at 500 K and 10^5 Pa.

Phase	Element content, wt. %:								
	C	B	W	Mo	Ti	Cr	Mn	Si	Fe
WB	-	5.55	94.45	-	-	-	-	-	-
TiC	16.46	-	-	-	83.54	-	-	-	-
MoB	-	10.10	-	89.90	-	-	-	-	-
BCC	$8.5\cdot10^{-13}$	$3.13\cdot10^{-6}$	$2.92\cdot10^{-6}$	-	$6.7\cdot10^{-8}$	0.29	1.59	1.70	96.42
M_7C_3 $(Cr,Mn)_7C_3$	8.99	-	-	-	-	88.20	2.81	-	-
Cr_4B	-	4.94	-	-	-	95.06	-	-	-
Fe_2B	-	8.81	-	-	-	-	-	-	91.19

Below 923ºC the alloy is in the solid state. In the temperature range 923-891ºC, the structures of the metal matrix are two-phase, consisting of ferrite (BCC) and austenite (FCC - Face Centred Cubic), and below 891ºC it is only ferritic. In the solid state in the range of 563-333ºC the formation of chromium boride Cr_2B occurs due to the dissociation of titanium diboride, and at temperatures below 333ºC Cr_2B transforms into Cr-rich Cr_4B boride (95.05 wt.% Cr). Below 120 ºC a manganese boride Mn_4B appears.

The temperature interval of equilibrium phases in the experimental alloy and their volume fraction (at room temperature) are shown in Table 2. The simulation showed that at room temperature the alloy should contain two carbide phases (TiC, M_7C_3) and five boride phases (WB, MoB, Fe_2B, Cr_4B, Mn_4B), and the metal matrix must be completely ferritic. The total volume fraction of carbides and borides should be 45.05 vol. %. By the structural status, the alloy is hyper-eutectic, because the crystallization of primary WB occurs before any eutectic transformation.

The thermodynamic characteristics of the chemical components and the phase chemical composition, calculated for 500 K (227 ºC) and a pressure of 10^5 Pa, are shown in Table 3. Under these conditions, the following data were obtained for the experimental alloy: molar mass is 51.90 g/mol; enthalpy (ΔH_{500}^o) is $-2.53 \cdot 10^4$ J/mol; Gibbs energy (ΔG_{500}^o) is –2.45·J/mol. According to the data in Table 1, the alloy is characterized by a significant inter-phase elemental redistribution. Entire ferrite-forming silicon and a significant part of austenite-forming manganese are concentrated in ferrite. Manganese is also present in chromium carbide. Strong carbide-forming elements are concentrated almost entirely in the corresponding borides and carbides (except for chromium, which is partially dissolved in ferrite).

Thus, the main sequence of formation of strengthening phases in the alloy during its crystallization is determined as WB → TiC → MoB → M_7C_3 → TiB_2 → Fe_2B → Cr_2B (Cr_4B) → Mn_4B. The crystallization of the alloy begins with the precipitation of primary borides and titanium carbides, after which it continued with the formation of eutectic which based on W(Mo)-rich borides. The formation of $(Cr,Mn)_7C_3$-based eutectic and TiB_2-based eutectic takes place at a much lower temperature. In the temperature range 923-891 ºC, the alloy had austenite in its structure, which then completely transforms into

Table 2

Temperature interval of phase precipitation and its equilibrium volume fractions at room temperature (RT).

Phase constituents	Temperature, ºC		
	Start	Finish	Volume fraction at RT, vol.%
Liquid	>1472	923	–
WB	1472	RT	4.30
TiC	1460	RT	4.90
MoB	1126	RT	8.09
BCC	1126	RT	54.95
M_7C_3	1000	RT	3.00
FCC	923	891	–
Fe_2B	945	RT	13.94
TiB_2	978	475	–
Cr_2B	563	333	–
Cr_4B	333	RT	9.96
Mn_4B	120	RT	0.86

Table 3

Thermodynamic characteristics of the components in the alloy (at 500 K and 10^5 Pa).

Component	Moles mass-fraction	Activity	Potential, J/mol
B	0.130	$8.51 \cdot 10^{-13}$	$-1.16 \cdot 10^5$
C	0.031	$5.63 \cdot 10^{-5}$	$-4.07 \cdot 10^4$
Cr	0,099	$2.36 \cdot 10^{-1}$	$-6.01 \cdot 10^3$
Fe	0.624	$9.43 \cdot 10^{-1}$	$-2.45 \cdot 10^2$
Mn	0.009	$5.19 \cdot 10^{-2}$	$-1.23 \cdot 10^4$
Mo	0.041	$1.11 \cdot 10^{-3}$	$-2.82 \cdot 10^4$
Si	0.018	$1.47 \cdot 10^{-11}$	$-1.04 \cdot 10^5$
Ti	0.027	$1.61 \cdot 10^{-14}$	$-1.32 \cdot 10^5$
W	0.021	$1.91 \cdot 10^{-4}$	$-3.56 \cdot 10^4$

eutectoid.

A comparison of the obtained results and the data of the article [14] shows the both coincidences and discrepancies between thermodynamic calculations and experimental data for the studied alloy. The simulation results are confirmed in terms of the structural status of the alloy, which is indeed hyper-eutectic. The structure of the real alloy revealed the presence of the predicted phase/structural constituents such as primary inclusions, and the eutectics of different types (one of which is based on W(Mo)-rich borides, another one is based on Cr-rich carboboride $M_7(B,C)_3$). In addition, the presence of pearlite in the structure of the real alloy confirms the adequacy of modelling, according to which austenite was part of the structure in the temperature range of 923-891ºC (as known, pearlite is formed only from austenite during eutectoid transformation).

The differences include the fact that the modelling did not predict the formation of carboborides. Also, instead of a number of forecasted eutectic reactions with the formation of different borides, there was actually only one reaction with the formation of "Chinese Script" eutectic which was based on carboboride $M_2(B,C)_5$. According to [14] $M_2(B,C)_5$ contains 23 wt.% Fe, 11-18 wt.% W, Mo, Cr (each), as well as 1.5 wt.% Ti. These data show that under crystallization in the sand form, instead of the formation of different boride phases (which presumes the high-content fluctuation of the element in the liquid), only one compound was formed, the nucleus of which grew by attaching nearby dissimilar atoms with high affinity to carbon (boron). Obviously, this process was the most kinetically advantageous in a view of the limited diffusion mobility of the alloying elements atoms. It was unlikely that the local concentration of a particular element may reach the equilibrium level for a particular boride (Table 1). For a similar reason, a significant part of chromium was replaced by iron (35 wt.%) and other elements (Mo, W, Mn) in carboboride $M_7(B,C)_3$ and a significant proportion of titanium was substituted by tungsten (10 wt.%) and molybdenum (3.5 wt.%) in carboboride Ti(C,B).

In general, satisfactory adequacy of the results of modelling the phase-structural state of the studied high-boron multi-component alloy is confirmed. This indicates the prospects for more extensive use of "Thermo-Calc Software" in the design of alloys of similar complex alloying systems.

Conclusion

The peculiarities of thermodynamically equilibrium phase-structural state formation in a multi-component 0.72 %C-Fe-W-Mo-Cr-Ti-Mn-Si alloy with high boron content (2.75 wt.%) was studied by thermodynamic modelling using the computer program "Thermo-Calc Software". It was found that crystallization in the alloy begins at 1472ºC with the formation of primary boride WB and carbide TiC and continues with a series of successive reactions with the formation of double eutectic "Ferrite + B" (where B is one of MoB, TiB_2, Fe_2B) and eutectic "Ferrite + Carbide M_7C_3». Borides of chromium (Cr_2B, Cr_4B) and manganese (Mn_4B) are formed in the solid state. The total volume fraction of borides and carbides is 45.05 vol. %. The results of the simulation mostly coincide with the real structural state of the alloy. The exception is that under nonequilibrium crystallization a complex carboboride $(W,Mo,Ti,Fe)_2(B,C)_5$ is formed instead of individual borides, and a carborides Ti(C,B) and $(Fe,Cr,Mo,W,Mn)_7(B,C)_3$ appears instead of carbides TiC and $(Cr, Mn)_7C_3$, accordingly.

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Моделювання фазово-структурного стану в гібридному мультикомпонентному сплаві з високим вмістом бору

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В роботі проаналізовано структуроутворення у мультикомпонентному Fe-W-Mo-Cr-Ti-Mn-Si сплаві триботехнічного призначення, що вміщує 0,72 мас. % С та 2,75 мас. % В. Комп'ютерне моделювання рівноважного процесу кристалізації виконували із застосуванням програми «Thermo-CalcSoftware». Сплав відноситься до заевтектичних композицій; його кристалізація починається при 1472 °С з формування первинного бориду WB і карбіду TiC. Після низки наступних евтектичних реакцій (в інтервалі 1126-923 °С) та твердофазних перетворень в сплаві формується набір рівноважних фаз, що на момент досягнення кімнатної температури складається із боридів WB, MoB, Fe_2B, Cr_4B, Mn_4B, карбідів TiC, M_7C_3 та фериту. Загальна об'ємна частка боридів та карбідів становить 45,05 об. %. Співставлення отриманих даних з результатами дослідження реального сплаву показало, що моделювання із «Thermo-CalcSoftware» забезпечує задовільну адекватність при прогнозуванні структури в сплавах обраної системи легування.

Ключові слова: мультикомпонентний сплав, борид, карбіди, мікроструктура, моделювання.