THERMODYNAMIC APPROACH TO THE DEVELOPMENT AND SELECTION OF HARDFACING MATERIALS IN ENERGY INDUSTRY

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Abstract:
The overall study objection is selection and optimization all available thermodynamic data required for using calculation of phase diagram (CALPHAD) technique within the Fe-C-Cr-Mn-Si-Ti system. Such data collected in the thermodynamic database can be used for predicting the phase constitution states of a given composition for Fe-based hardfacing materials, which often use in energy industry in order to increase the abrasion and impact wear resistance of equipment parts. In order to compare theroretical calculation results with experimental data, four different types of hardfacing were deposited using flux-cored arc welding. Microstructure and chemical composition of deposited layers was investigated using optical and scanning electron microscopy together with energy dispersive X-ray spectroscopy. Comparison of experimental and computed results shows that they are in good agreement in meaning of presence of all-important phase equilibrium regions. The developed database can be used for rational selection of hardfacing materials for energy industry equipment and reasonable choice of new alloying systems.

INTRODUCTION

Wear of energy equipment machine parts which caused by intensive abrasion wear together with high impact loads and aggressive environments is still an actually engineering and scientific problem. Most common equipment failures due to different types of wear take often place in oil and gas and coal industries during mining, transportation and processing of energy sources. Examples of such equipment are components of drilling rigs, coal crushers and conveyors, excavators, etc. In renewable energy industry intensive wear of working surfaces is the main reason of failure and decreasing productivity of equipment in the processing of wood biomass waste into fuel briquettes. One of the most rational and effective way to reduce wear in such cases is the creation of surface layers with high mechanical properties by forming microstructure, comprising hard wear-resistant phases in sufficient amounts. Technologically, this can be achieved by using hardfacing technique with alloys consisting of element combinations which allow to provide required phase composition and microstructure.

LITERATURE REVIEW

Traditionally the most commonly employed hardfacing materials for obtaining high abrasion resistance is the Fe-based alloys with large concentration of carbon and strong carbide forming elements such as Cr, Ti etc [1]. In conditions where high impact resistance needed, using Fe-based materials alloyed with austenite-forming elements such as Mn, Ni is much preferred [2]. In some cases, the mechanical properties of the coating are not so important as corrosion resistance and antifriction properties, which can be improved by alloying with Cr, Si, Cu, Nb [3, 4, 5]. However, in all cases the key factor which determines the complex of surface layer properties is reasonable choice of multicomponent alloying system based on analysis of all possible interactions between components. That type of analysis can be performed on the basis of equilibrium thermodynamics rules using modern technique of calculation of phase diagrams (CALPHAD). This technique allows to predict properties of multicomponent system (alloy) by the extrapolation data from lower-order binary and ternary systems.

Thus, the accuracy of predicting properties depends on the amount of thermodynamic data assessed from experimental investigation and (or) first principles calculations for binary and ternary interactions between the system components.

Taking into account importance of the most commonly used elements for alloying of hardfacing materials for energy industry equipment, thermodynamic dataset must cover at least binary and ternary iterations in Fe-C-Cr-Mn-Si-Ti[1] system. According to literature data the most complete and precision database, which partially covers chosen system except Cr created by B. Hallstedt [6]. On the other hand, some binary and ternary interactions Cr with Fe, Mn and C was investigated by B.-J. Lee et al. [7, 8]. K. Wang [9], L.Y. Chen [10] et al. conducted thermodynamic assessments for modelling phase equilibria in Ti-Cr-Fe and Ti-Cr-Mn metal ternary systems. Calculation from first principles together with experimental measurements in ternary systems containing Cr and Si were carried out by A. Berche [11, 12, 13].

The main objective of this paper to create thermodynamic database including accurate models for prediction phase composition of hardfacing materials of Fe-C-Cr-Mn-Si-Ti system with the aim of development new high-performance coatings for energy industry equipment.

THERMODYNAMIC MODELS

The CALPHAD method was used for calculation of phase equilibria in the Fe-C-Cr-Mn-Si-Ti system. For the solid solution phases subregular solution model with two sublattices [14] was applied (one sublattice filled with Fe, Cr, Mn, Si, Ti atoms and the second one with C atoms and vacancies), so the molar Gibbs energy of each phase was represented as:

$$G_m^f = \sum_i y_i \left(y_c \, ^\circ G_{i:c}^\phi + y_{va} \, ^\circ G_{i:va}^\phi \right) + RT \sum_i y_i \ln(y_i) + \\ cRT(y_c \ln(y_c) + y_{va} \ln(y_{va})) + \, ^E G_m^\phi + \, ^{mg}G^\phi \tag{1}$$

where:

i = Fe, Cr, Mn, Si, Ti, y_i, y_c, y_{va} are the site fractions of metal, carbon and vacancies on the relevant sublattices. $^oG^\phi_{i:va}$ and $^oG^\phi_{i:c}$ are the Gibbs energy of the pure element at the stable state, according to [15] and Gibbs energy of the state where all the interstitial sites filled with carbon. The term $^{mg}G^\phi$ represent magnetic contribution into Gibbs energy according to [16]. Parameter L denote the interaction energy between elements. $^EG^\phi_m$ is the excess Gibbs energy and is expressed as follows:

$$^E G_m^\phi = \sum_i \sum_j y_i y_j \left(y_c L_{i,j:C}^\phi + y_{Va} L_{i,j:Va}^\phi \right) + \\ y_C y_{Va} \sum_i y_i L_{i:C,Va}^\phi + \sum_i \sum_j \sum_k y_i y_j y_k \left(y_C L_{i,j,k:C}^\phi + \\ y_{Va} L_{i,j,k:Va}^\phi \right) \\ i \neq j \neq k \tag{2}$$

The liquid phase was modeled as substitutional solution with Gibbs energy represented as follows:

$$G_m^{liq} = \sum_i x_i \, ^\circ G_{i:c}^\phi + RT \sum_i x_i \ln(x_i) + \, ^E G_m^{liq} \tag{3}$$

where:

x_i is the mole fraction of the component and

$$^E G_m^{liq} = \sum_i \sum_j x_i x_j L_{i,j}^{liq} + \sum_i \sum_j \sum_k x_i x_j x_k L_{i,j,k}^{liq} \\ i \neq j \neq k \tag{4}$$

All other solid phases such as carbides, silicides, intermetallic phases and ordered solutions was modeled using energy compound formalism described in [17]. Thermodynamic properties for pure elements and some compounds were taken from [15]. The thermodynamic parameters and models descriptions for binary and ternary systems, which are included into Fe-C-Cr-Mn-Si-Ti system were taken and optimized from the list in Table 1 and presented in Appendix section.

Table 1
The systems included into database and corresponding references

System	Reference	System	Reference
C-Cr	[7, 8, 18]	C-Cr-Fe	[7, 8, 40]
C-Fe	[7, 8,19, 20, 21, 22, 23, 24]	C-Cr-Mn	[8]
C-Mn	[21, 25, 26]	C-Cr-Si	[29]
C-Si	[6, 27, 28, 29]	C-Cr-Ti	[41]
C-Ti	[30]	C-Fe-Mn	[23]
Cr-Fe	[9]	C-Fe-Si	[28, 42]
Cr-Mn	[11]	C-Fe-Ti	[43, 44]
Cr-Si	[11, 31, 32]	C-Si-Ti	[45]
Cr-Ti	[9, 32]	Cr-Fe-Mn	[10]
Fe-Mn	[23, 33]	Cr-Fe-Si	[46,47]
Fe-Si	[28, 34, 35, 36]	Cr-Fe-Ti	[9]
Fe-Ti	[9, 37]	Cr-Mn-Si	[12]
Mn-Si	[13, 35]	Cr-Mn-Ti	[10]
Mn-Ti	[38]	Cr-Si-Ti	[32]
Si-Ti	[6, 32, 39]	Fe-Mn-Si	[6, 35]

MATERIALS AND METHODS

The hard facings for investigation were deposited using flux-cored arc welding (FCAW) technique on the mild carbon steel substrate using Lastek welding machine with a direct current of 150 A, at the reverse polarity.

Flux-cored welding wires were prepared by drawing powder mixtures of initial components into low carbon steel sheath using drawing machine of Paton EWI NASU construction. Commercially available powders of titanium, chromium, ferrochromium, ferro silicomanganese with an average particle size less than 100 µm were used as a starting materials. Microstructures investigations were performed using optical and scanning electron microscopy (SEM) together with energy-dispersive X-ray spectroscopy (EDS). The geometric parameters of the microstructure were measured by random sections method using Image-ProPlus software.

Four types of the hard facing materials of different alloying systems were selected as samples for microstructure investigations and comparison experimental and calculated relationship between composition and phase constitution data (Table 2).

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[1] Thermodynamic database of Fe-C-Cr-Mn-Si-Ti system in tdb file form can be accessed at: https://www.researchgate.net/publication/339528105_CCR-FEMNSITI

Sample No	Element content (wt. %)					
	C	Cr	Mn	Si	Ti	Fe
S1	2.3	-	-	1.0	9.5	in balance
S2	0.25	10.2	14.3	3.0	-	in balance
S3	3.0	-	20.2	3.2	3.2	in balance
S4	3.8	14.3	12.9	6.0	3	in balance

RESULTS OF EXPERIMENTAL REVISION OF MODEL PARAMETERS

The microstructure of sample S1 mainly consists of TiC (MC) uniformly distributed grains with flower-like structure and ferrite (BCC)-TiC rod-like Fe-rich eutectic, as shown in Fig. 1.
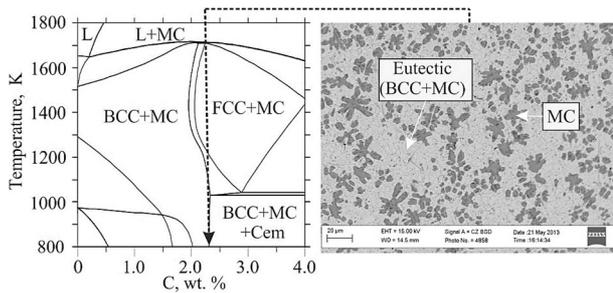
Fig. 1 Calculated fragment of the Fe-Ti9.5-Si1-C system polythermal section and corresponding microstructure of the sample S1

Corresponding phase region (BCC+MC) on the current polythermal section of Fe-Ti-Si-C system may exists below 1600 K in concentration range from 0 to 2.4 wt. % of carbon. This phase region has boundaries with liquid (L)+BCC+MC region, austenite (FCC)+BCC+MC and the region where ferrite and TiC coexists with metastable intermatalic phases. According to calculated results the structure formation of the typical hard facing at C < 2.0 % can be divided into three stages: primary crystallization of TiC in form of cuboid- shaped grains; TiC dendritic grain grown; eutectic (TiC+ferrite) crystallization.

In the hard facings of Fe-Cr-Mn-Si-C system which are similar to the S2 sample its major structure components are austenite (FCC) and ferrite (BCC) (Fig. 2).
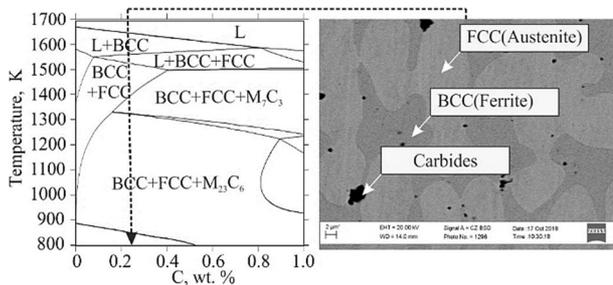

Fig. 2 Calculated fragment of the Fe-Cr10.2-Mn14.3-Si3-C system polythermal section and corresponding microstructure of the sample S2

Such microstructure is typical for well known duplex steels. However, in presented system there are some features which are caused by two competing processes: stabilizing of ferrite by presence of silicon and chromium and stabilization of austenite through manganese partitioning between ferrite and austenite. As a result, both phases

become stable and coexist in wide temperature range from ~ 1500 K to the line determining beginning of (Fe, Cr, Mn)$_x$(C)$_y$ compounds precipitation. So, the structure formation process of hard facing includes crystallization of ferrite from liquid, crystallization of austenite in three-phase region (L+BCC+FCC) and finally precipitation of dispersed carbides and intermetallic phases. The microstructure of the hard facing of Fe-Mn-Ti-Si-C alloying system (sample S3) consists of manganese austenite grains, elongated in the same direction (Fig. 3). Most austenite grains contain fine, uniformly distributed facetted inclusions of TiC, which allocated mainly in the central areas of austenite grains. Intergranular space between austenite grains is filled with austenite (FCC) + carbides of cementite type (Cem) eutectic. According to calculated phase equilibrium plot structure formation process of such hard facings can be divided into three main stages: crystallization of TiC (MC) from liquid at high temperatures; austenitic nucleation and growth around TiC particles; eutectic ledeburite type reaction according to scheme: L→FCC+Cem.
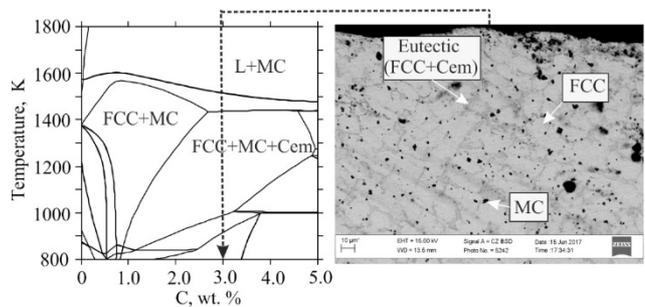

Fig. 3 Calculated fragment of the Fe-Mn20.2-Ti3.2-Si4.1-C system polythermal section and corresponding microstructure of the sample S3

The hardfacing alloys with chemical composition close to sample S4 crystallize according to a complex scheme, containing some features of structure formation of S1, S2 and S3 hard facing alloys. Solidification of that type of alloys begins at high temperatures from nucleation and grown of TiC (MC) fine particles which play important role as a modifiers for further structure formation.

At the next stage crystallization of the alloyed (Cr, Mn) austenite occurs by heterogeneous nucleation on TiC particles and further dendritic grain grown in the opposite direction to the heat sink. The last important stage of the alloy solidification is the eutectic reaction of cooperative FCC and M$_7$C$_3$ (Fig. 4) grain grown though the interdendritic space.
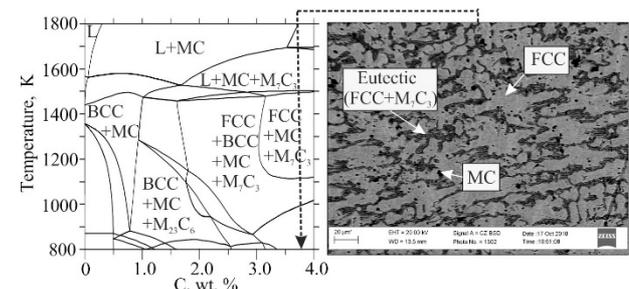

Fig. 4 Calculated fragment of the Fe-Cr14.3-Mn12.9-Ti3-Si6-C system polythermal section and corresponding microstructure of the sample S3

A comparative analysis of the structures and phase compositions of four experimental had facing alloys (S1-S4), which belong to different alloying systems with corresponding polythermal sections of the Fe-Cr-Mn-Ti-Si-C system, calculated using dataset developed in this study shows good agreement between experimental and thermodynamic modelling data. So, the thermodynamic calculation in current system can be significantly extended to a wide range of materials used in energy industry equipment.

DISCUSSION

According to the analysis of presented thermodynamic description of Fe-Cr-C-Mn-Si-C system the Fe-based hypoeutectic (2-4 wt. % C) alloys, simultaneously high alloyed with Mn and Cr (10-15 wt. %) and low alloyed with Si and Ti (1-4 wt. %) are the most versatile hard facings capable of resisting abrasion, corrosion and impact. According to experimental and calculated data the generalized solidification scheme of such alloys can be divided into three main stages (Fig. 5): I – crystallization of TiC; II – crystallization of primary austenite on the TiC grains; III – eutectic reaction of liquid phase decomposition into austenite + M_7C_3 mixture. The main role of each alloying element in current system is as follows: Fe is the base of the hard facing alloy; Cr provides corrosion resistance, and formation of M_7C_3 abrasion resistant carbide; Mn provides impact resistance due to stabilization of high-alloyed austenite; presence of Ti allows to obtain inclusions of refractory phase (TiC) which provides more intensive nucleation of austenite grains, as well as promotes increased abrasive wear resistance.
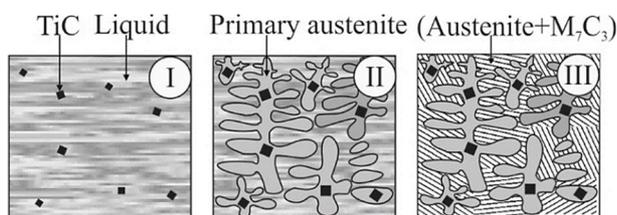

Fig. 5 Main stages in the solidification of the typical Fe-based hard facing alloy of Fe-Cr-C-Mn-Si-C system

The peculiarity of using the proposed database is that it is mainly focused on alloys, whose phase composition remains unchanged after the solidification, so no quenching or tempering is required for these hard facings. In most cases traditional heat treatment can be completely replaced by the welding thermal cycle during hard facing deposition by arc welding. In such conditions, the optimal temperature range for phase data analysis is 800-1200 K. In some cases, it is necessary to include into calculations phase transformations in non-equilibrium state, such as martensitic transformations, austenite decomposition on the ferrite-cementite mixtures, precipitation during age hardening etc. For that purpose, proposed database can be integrated into diffusion-controlled reactions software (DICTRA) with relevant mobility databases. Database structure makes it possible to use it with commercial Thermo-calc software as well as with freely available Open Calphad software.

In further investigations its important to extend current database by adding tungsten for covering tungsten carbide-based hard facing materials, which are widely used in oil and gas industry.

CONCLUSIONS

A thermodynamic database for Fe-based hard facing alloys of Fe-C-Cr-Mn-Si-Ti system has been developed to predict phase composition of surface layers deposited by flux-cored arc welding technique on the working surfaces of energy industry equipment. Database contains thermodynamic data and models for all binary interactions and fifteen out of twenty required ternary interactions between system components. Good agreement has been obtained between calculated phase composition and experimental data, observed by microstructural investigation of hard facing weld deposits of Fe-Ti-Si-C, Fe-Cr-Mn-Si-C, Fe-Mn-Ti-Si-C and Fe-Cr-Mn-Ti-Si-C alloying systems. It was found, that in investigated alloys phase composition calculated at temperature range of 800-1200 K remains stable during cooling to a room temperature. Such approach to the prediction of hard facing alloys phase composition substantially reduces number of trial experiments during selection and development materials for improvement durability of energy industry equipment. Taking into account that the software implementation of the proposed approach is widely represented by commercial and non-commercial software packages such as Thermo-Calc, OpenCalphad, MatCalc and many others, it can be easily integrated into automated materials quality management systems.

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