A Computational Approach to the Microstructural Design of High-Speed Steels

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Increasing requirements concerning the operational conditions and durability of tools create a demand for the optimization of tool steels. High-speed steels (HSS), for example, contain high amounts of carbides embedded in a secondary hardenable martensitic matrix. The wear behavior and the mechanical properties of HSS can be optimized for a certain application by adjusting the type and amount of carbides, as well as their compositions and the composition of the matrix. Computational thermodynamics based on the calculation of phase diagrams method allow the estimation of arising phases as well as phase compositions during the solidification or the heat treatment of a steel. However, in complex alloy systems, for example, HSS, the relationships between the content of alloying elements and the stability and the composition of phases can be complicated and nonlinear. Therefore, it can be difficult to find alloy compositions that are suitable to achieve a desired microstructure with iterative calculations. To handle this difficulty, a computational tool is developed, which determines compositions to obtain predefined HSS microstructures. The computational tool is based on a neural network that was previously trained with a thermodynamically calculated database. The efficiency of this approach is experimentally verified by producing and investigating laboratory melts of different HSS.

1. Introduction

The development of new high-speed steels (HSSs) requires a comprehensive knowledge about the alloying system and the interactions between the chemical composition and the microstructure. The amount and composition of stable phases in HSS does often have nonlinear dependencies on the overall chemical composition, which makes it difficult to determine the chemical composition to obtain a desired microstructure. The present work aims at designing a computational tool, which is capable of generating a chemical composition for a HSS with user-defined microstructural features.

In modern materials science, several computational methods have been established for the development, optimization and modeling of steels, which complement classical experiment-driven methods. Among them the use of neural networks as a form of artificial intelligence has gained increasing popularity. In the literature, numerous articles can be found in which neural networks are used in the context of steel research, for example, to optimize the heat treatment of HSS, for the prediction of mechanical properties, or delta ferrite prediction in stainless steel welds. Virtually, in all of the publications on this topic, experimental data is used to train the neural networks for the desired purposes. As the quantity of available training data limits the number of input and output parameters of a neural network, most of the works either require an enormous effort collecting experimental data or treat rather simple problems with a low number of input and output parameters. In the present work, an attempt is made to construct a neural network for the development of new HSS, but bypass the bottleneck of collecting experimental data. This is done by training a neural network with a set of thermodynamically calculated data instead of experimental data. This approach offers the use of an arbitrary quantity of training data, as thermodynamic calculations based on the calculation of phase diagrams (CALPHAD) method can be executed fast as well as completely automated. This allows to build the neural network with a high number of input and output parameters. Computational thermodynamics based on the CALPHAD method are widely used in materials science and are often applied for the development of new steels. It allows the calculation of phase equilibria, phase diagrams, and phase transformations as well as thermodynamic assessments in complex multi-component alloys. Coupling this well-established and powerful method with neural networks appears promising to provide new opportunities in the development and optimization of steels. The calculation of single-point equilibria provides information about the types, amounts, and compositions of stable phases for a given chemical composition and a given temperature, e.g., hardening temperature. This can be useful to estimate the resulting microstructure and additionally the resulting mechanical
properties. However, for the development of a new steel, it might be desirable to be able to define certain stable phases as well as their amounts and compositions at a temperature and to receive the corresponding chemical composition for the steel. This represents the reverse way of how classical CALPHAD calculations work. The target of the present study is therefore to build a software solution based on a neural network, which is capable of generating chemical compositions of HSS for defined parameters regarding stable phases and their compositions.

2. Neural Network Model

2.1. Network Architecture

Neural networks are mathematical models consisting of multiple layers of nodes or “neurons” that are interconnected with mathematical functions. This setup is inspired by the way in which the human brain processes data. Neural networks that rely on supervised learning methods are prepared for their tasks in a training process in which the neural network learns the relationships between existent pairs of input and output datasets, the so-called training data. This is done by continuously adjusting the weights of the connections between the neurons and thereby reducing the error between generated and the actual output data. After the training process, the neural network can be used to generate outputs for any new input data.[12,16,17]

The neural network used in this work was designed to be presented with a hardening temperature \( T_{\text{H}} \) and the microstructural features listed in Table 1 on the input side and to generate a corresponding chemical composition including the elements listed in Table 2 on the output side. The schematic structure of the neural network is shown in Figure 1. All of the microstructural features on the input side refer to the thermodynamic equilibrium at hardening temperature. The parameters were selected to represent the stable phases at hardening temperature on the one hand and the solution state of the matrix on the other hand. The portion of stable phases, i.e., the metal matrix as well as MC and M\(_6\)C carbides, can strongly influence the wear resistance of an HSS, which makes it important to set these values as manageable parameters.[18,19] The solution state of the metal matrix defines the secondary hardening potential of the steel during tempering and is therefore important to control the mechanical properties of the steel in the heat-treated state.[20] The C content of the metallic matrix is important to ensure hardenability, whereas the elements Cr, Mo, V, and W participate in carbide precipitation during tempering and cause secondary hardening.[21,22]

The neural network was implemented in the software MATLAB version 2016a using the Neural Network Toolbox and the Neural Net Fitting App. A multilayer feedforward network consisting of the input, output, and one hidden layer with 20 hidden neurons was used.

2.2. Network Training

For the training of a neural network with supervised learning techniques, sets of corresponding input and output data are required. In the context of the present work, this means that datasets of hardening temperatures with the defined microstructural features and corresponding chemical compositions are required. As the total number of parameters was chosen very high (10 input + 9 output), in this case, a quantity of 100 000 datasets was aimed. To build a database containing this amount of data, chemical compositions in a range that is reasonable for HSS were created randomly. This was done by generating 100 000 uniformly distributed random numbers for each alloying element in the ranges listed in Table 3 and combining them to random chemical compositions. To receive the corresponding

| Table 1. Input parameters for the training of the neural network. |
|---|---|---|---|---|---|---|---|---|
| \( T_{\text{H}} \) | Matrix | MC | Mo | V | W | Co | % |
| °C | [vol%] | [vol%] | [wt%] | [wt%] | [wt%] | [wt%] | |
| Input | 80.0 | 20.0 | 0 | 0.65 | 5.0 | 0.75 | 3.0 | 5.0 |
| Output_eq | 79.6 | 18.2 | 2.2 | 0.67 | 5.2 | 1.9 | 0.86 | 2.9 | 5.0 |
| HS\(_6\)Mo | 80.0 | 20.0 | 0 | 0.65 | 5.0 | 0.75 | 0 | 5.0 |
| Output_eq | 80.0 | 18.9 | 1.1 | 0.69 | 4.9 | 3.9 | 0.89 | 0 | 5.1 |
| HS\(_6\)S-2-5 | 90.4 | 13.8 | 8.3 | 0.61 | 4.2 | 2.6 | 1.11 | 2.5 | 4.8 |

| Table 2. Ranges for the generation of random numbers that were used as chemical compositions in the training data. Values in wt%. |
|---|---|---|---|---|---|---|
| C | Si | Mn | Cr | Ni | Mo | V | W | Co | Fe |
| Min. | 1 | 0.5 | 0.2 | 3 | 0.05 | 0 | 0 | 0 | 0 | bal. |
| Max. | 3 | 0.75 | 0.4 | 5 | 0.25 | 10 | 10 | 15 | 11 | bal. |

Figure 1. Schematic illustration of the structure of the used neural network.
microstructural features, thermodynamic equilibrium calculations were performed for each of the randomly generated chemical compositions and a randomly generated hardening temperature between 1100 and 1200°C. The thermodynamic equilibrium calculations were conducted using the ThermoCalc Toolbox for MATLAB together with the database TCFe8.

As an example of the data finally stored in the database, Figure 2 compares the distribution of the C content of the randomly generated chemical compositions and the C content of the metallic matrix in the thermodynamically calculated data. Figure 2a shows that the C content in the random chemical compositions is uniformly distributed, whereas the C content of the metallic matrix shown in Figure 2b is distributed very unevenly. The counts of database entries decrease with the C content of the metallic matrix, which reveals that only a small number of random chemical compositions enable high C contents in the metallic matrix.

For the training procedure, the data was divided into three subsets. The training subset, which contained 70% was used for the actual training process in which the network is presented with the data and is iteratively adjusted to it. 15% of the data were used as the test subset and the validation subset, respectively. The test subset is used to check the network response for new data during the training process, whereas the validation subset is used to evaluate the performance of the neural network after training. The training process itself can be carried out using different training algorithms with different characteristics and performance. In the present work, a scaled conjugate gradient algorithm was used, which enables a fast training process with large datasets.

3. Validation

The efficiency of the developed optimization tool was tested by applying it to an optimization of the HSS HS6-5-2-5. The optimization aimed on an improvement of the wear resistance by changing the carbide configuration and on an improvement of the secondary hardness by adjusting the matrix composition.

The latter was pursued with two different concepts. In the first concept named HS_W+Mo, a high solution state of Cr, Mo, W, and C in the matrix was desired. In the concept named HS_Mo, a high solution state of Cr, Mo, and C was desired, eliminating the element W from the alloy. A high solution state of these alloying elements was targeted as Cr, Mo, and W form small carbides during tempering and thereby contribute to an increase in the secondary hardness maximum. Regarding the carbide configuration, an amount of exclusively 20 vol% MC carbides was desired to be stable at a hardening temperature of 1200°C in both variants to achieve a high resistance against abrasive wear. The positive effect of increasing MC carbide contents on the abrasive wear resistance of HSS can be related to their morphology as well as their high hardness which lies above the hardness of many common abrasives, e.g., Al₂O₃.[19,24,25]

The precise input parameters that were defined for each of the alloy concepts are given in Table 3. For these input parameters, the chemical compositions listed in Table 4 were proposed by the neural network. With the proposed chemical compositions and the hardening temperature of 1200°C, thermodynamic equilibrium calculations were then performed. The results named Output_eq are listed in Table 3 and can be compared to the input parameters to assess the efficiency of the neural network.

Subsequent to the computational alloy development, an experimental investigation of the proposed alloys was conducted. Therefore, HSS with the chemical compositions as proposed by the neural network were produced as 200 g laboratory melts without any further optimizations or adjustments.

| Table 4. Chemical compositions of the HSS HS_W+Mo and HS_Mo as proposed by the neural network and chemical composition of the reference steel HS6-5-2-5. Values in wt%. |
|---------------------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| C    | Si   | Mn   | Cr   | Ni  | Mo  | V   | W   | Co  | Fe  |
|------|------|------|------|-----|-----|-----|-----|-----|-----|
| HS_W+Mo | 2.76 | 0.61 | 0.32 | 5.18 | 0.19 | 3.86 | 8.41 | 7.56 | 4.97 | bal. |
| HS_Mo   | 2.85 | 0.63 | 0.33 | 5.22 | 0.24 | 7.83 | 9.00 | 0.13 | 5.05 | bal. |
| HS6-5-2-5 | 0.92 | 0.40 | 0.30 | 4.10 | 0.06 | 5.00 | 6.40 | 6.40 | 4.80 | bal. |
In addition, the steel HS6-5-2-5 was also produced in the laboratory scale to serve as a reference material. The laboratory melts were produced in a vacuum-induction furnace (Leybold–Heraeus GmbH, Germany) using ARMCO iron and different prealloys (FeV, FeMo, FeCr, etc.). The cast material was heat-treated by austenitizing at 1200 °C for 30 min in a protective gas furnace, followed by oil quenching. Subsequently, tempering experiments were conducted at temperatures between 480 and 580 °C. At each temperature, three tempering steps of 2 h each were performed. The hardness of the HSS in the as-quenched state as well as in different tempering states was tested using a Vickers hardness tester with a load of 30 kg.

For the investigation of the steels microstructures, samples were taken from the material in the as-quenched state and were metallographically prepared by grinding and subsequent polishing with 3 and 1 µm diamond suspension. The steels microstructures were observed by means of scanning electron microscopy (SEM) using a FEG-SEM MIRA 3 (Tescan, Czech Republic) operated at a working distance of 15 mm and an acceleration voltage of 20 kV. The carbide volume contents of the HSS were determined by means of digital image analysis using the methodology described by Benito et al.\[^{26}\] At least five backscattered electron (BSE) SEM images of each steel were evaluated. To identify the carbide types in the steel microstructures, electron backscatter diffraction (EBSD) measurements were conducted in the SEM using a NordlysNano detector (Oxford Instruments, UK). The specimens were polished with colloidal silica suspension with a particle size of 0.02 µm for the investigations.

### 4. Results and Discussion

A database consisting of random chemical compositions and corresponding thermodynamic equilibrium values was created and proved to be suitable for training a neural network. Thus, the trained neural network stores generalized relationships between the chemical compositions and the thermodynamic equilibrium values available in the training data.

To test the performance of the neural network, two HSS variants HS_W + Mo and HS_Mo were developed using the neural network. The chemical compositions of the two HSS listed in Table 4 were generated by the neural network for the input parameters given in Table 3. Both the proposed compositions appear reasonable for HSS with high carbide contents, which indicates the principal functionality of the approach. The main difference between the two proposed compositions can be found in their Mo and W contents, which corresponds to the different matrix contents of Mo and W that were defined in the input parameters. Interestingly, the Mo content of HS_Mo is virtually equal to the Mo content of HS_W + Mo plus half of the W content of HS_W + Mo. This follows the well-known mass ratio for the substitution of W by Mo, which is 2:1.\[^{26}\] A detailed judgment about the performance of the neural network can be made by comparing the input parameters of the HSS variants HS_W + Mo and HS_Mo to the thermodynamic equilibrium values that were calculated with the proposed compositions and the hardening temperature of 1200 °C (Table 3). The comparison shows that the carbide configurations in both the developed HSS are close to the targeted values of 20 vol% MC and no M_6C carbides. In both HSS, the attained MC carbide contents show relative deviations of less than 10% from the targeted values. However, small amounts of undesirable M_6C carbides are also present.

Regarding the matrix compositions, only marginal deviations between the desired and the achieved values can be found. The highest deviations can be observed in the matrix contents of C and V, which are slightly higher than desired. It can be concluded that the overall performance of the neural network in generating chemical compositions to obtain an HSS with the specified features is very good. This means that the challenge of developing an HSS with arbitrary microstructural features can be bypassed using a sufficiently trained neural network. In consideration of the deviations between the targets and the results, it can be stated that the chemical composition generated this way can at least be used as an initial value for a manual optimization. Naturally, this only holds true under the premise that the desired microstructure is thermodynamically stable at any reasonable heat-treatment temperature.

However, the neural network tool that was built and applied in the present work undoubtedly leaves room for further improvement, as some crucial boundary conditions for a practical use are not yet taken into account. For example, the system does consider what phase field, e.g., fcc + MC, has to be reached, but it does not consider the exact location inside the phase field. As a consequence, a proposed chemical composition and heat-treatment temperature could be located in the direct proximity of an undesired phase field which would be unfavorable for a practical use.

Despite the weaknesses the presented approach still has, the developed HSS variants HS_W + Mo and HS_Mo were produced in a laboratory scale and characterized as described in Section 3. Figure 3a shows an SEM image of the microstructure of the HSS HS6-5-2-5, which serves as a reference steel in the experimental part, in the hardened state. The microstructure features a carbide network that consists of bright skeleton-like carbides as well as small and blocky carbides appearing dark. The microstructures of the two developed HSS, as shown in Figure 4a and 5a, exhibit a much coarser carbide structure with bright skeleton-like carbides as well as dark and blocky carbides of different sizes. By means of EBSD measurements, the dark carbides could be identified as vanadium carbides of the type VC in all three HSS. The bright carbides could be identified as M_6C carbides of the type Fe_3W_2C in the HSS HS6-5-2-5 and HS_W + Mo, whereas M_2C carbides were detected in the HSS HS_Mo. All three carbide types are well known to form in HSS.\[^{27}\] The carbide volume fractions determined via quantitative image analysis are given in Table 5. The reference material HS6-5-2-5 mainly features M_2C carbides, whereas mainly primary MC carbides can be found in the microstructures of the two newly developed HSS.

The measured volume contents of MC carbides approximately match the thermodynamically calculated values listed in Table 3. The measured M_2C/M_6C volume contents in the HSS HS_W + Mo and HS_Mo are distinctly higher than the thermodynamically calculated values. A reason for this deviation is that the as-cast state can be referred to as a nonequilibrium state and the heat treatment with a duration of 30 min is not sufficient to transfer the microstructure into a state close to the
Figure 3. a) SEM-BSE image of the microstructure of the HSS HS6-5-2-5. In the image the spots of the EBSD phase analysis are marked. b) Solved Kikuchi pattern obtained at spot 1. c) Solved Kikuchi pattern obtained at spot 2. d) Solved Kikuchi pattern obtained at spot 3.

Figure 4. a) SEM–BSE image of the microstructure of the HSS HS_W + Mo. In the image, the spots of the EBSD phase analysis are marked. b) Solved Kikuchi pattern obtained at spot 1. c) Solved Kikuchi pattern obtained at spot 2. d) Solved Kikuchi pattern obtained at spot 3.

Figure 5. a) SEM–BSE image of the microstructure of the HSS HS_Mo. In the image, the spots of the EBSD phase analysis are marked. b) Solved Kikuchi pattern obtained at spot 1. c) Solved Kikuchi pattern obtained at spot 2. d) Solved Kikuchi pattern obtained at spot 3.
The use of training data consisting of a combination of randomly generated chemical compositions and corresponding thermodynamic equilibrium values instead of experimental data, proved to be adequate for the implementation.

Testing the neural network by using it for the optimization of an HS6-5-2-5 showed that the chemical compositions generated by the neural network are appropriate to fulfill the desired characteristics with only small deviations.

The experimental validation revealed that some larger deviations between the desired microstructure and the microstructure of the actually produced material may be expected. This originates from the fact, that the neural network relies on thermodynamic equilibrium data, but actual steel microstructures represent nonequilibrium states.

Conflict of Interest

The authors declare no conflict of interest.

Keywords
calculation of phase diagrams (CALPHAD), high-speed steels, neural networks, simulation, steel design

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