Nickel-based Superalloys Heat Resistance BRANN Simulation and Analytical Approximation

ANDREY TYAGUNOV, OLEG MILDER, DMITRY TARASOV
Department of IT and Automation
Ural Federal University
Mira 32 – R041, Ekaterinburg 620002
RUSSIA
datarasov@yandex.ru http://www.urfu.ru

Abstract: - Nickel-based superalloys containing many different chemical elements are systems with complex doping. These materials are widely used for the gas turbine engines parts and other heat-resistant devices manufacturing, which makes them extremely important for the industry. One of the main service property of the superalloys is the heat resistance that is expressed by the value of the ultimate tensile strength (UTS) or creep to rupture (σ). The level of damage required to cause failure is measured after the metal is heated and maintained to a certain temperature for a specific time interval. The heat resistance of alloys with different chemical compositions is often estimated using the complex Larson-Miller parameter (P_{LM}), which combines the temperature and the exposure time. The development of new alloys takes considerable time and is quite expensive. A model describing the dependence of UTS on the alloys composition would be an essential help for the developers. In our work, we have applied a statistical method for modelling the properties of alloys according to their composition. The approach is based on the use of artificial neural networks with preliminary processing of the input data. This allowed us to obtain a series of dependences $\sigma = f(P_{LM})$ for a large number of superalloys compositions. The simulation results are in good agreement with the experimental data. Plots of heat resistance vs P_{LM} have a characteristic exponential form for all alloys, however, each composition has its own characteristics reflected in the graph's slope coefficient, which indicates the thermal stability of an alloy.

Key-Words: - nickel-based superalloys, artificial neural network, ultimate tensile strength, heat resistance, Larson-Miller

1 Introduction

Nickel-based, but containing significant amounts of at least twelve other elements, including exotic rhenium, ruthenium, tantalum and hafnium, superalloys are high-temperature materials that demonstrate excellent resistance to mechanical and chemical degradation at temperatures close to their melting points. Since they first appeared in the mid-20th century, these alloys have had a unique impact on industry and technology. The remarkable characteristics of superalloys are the result of many years of developers' efforts. Superalloys products must retain the service complex of properties under conditions of high temperatures and mechanical loads throughout the design life cycle. Plenty of researchers and technologists have worked hard to develop a basic understanding of their physical behavior and the more practical aspects necessary for the best utilization of these alloys. In addition, it became obvious that the themes of alloy design, process development, component design, life expectancy, and material behavior are closely interrelated [1-7].

The main service properties of the alloys are heat resistance and structural phase stability. The heat resistance is the ability of material to resist the load at high temperatures, without undergoing permanent deformation or fracture. The heat resistance of nickel alloys is estimated by the long-term strength limit, i.e. the greatest mechanical stress that the material could resist without failure at a given temperature, test duration and working atmosphere (the ultimate tensile strength, UTS). The phase stability is the ability of a material to retain structural properties during a period of prolonged isothermal exposures.

TABLE 1. Chemical compositions of some common cast superalloys forming a sub-sample, wt% (Ni is balanced)

Alloy	Cr	Co	Mo	W	Al	Ti	Nb	Ta	Re	Ru	Hf	C	B	Zr
CMSX-4	6.5	9.0	0.6	6.0	5.6	1.0	-	6.5	3.0	-	0.1	-	-	-
PWA1480	10.0	5.0	-	4.0	5.0	1.5	-	12.0	-	-	-	-	-	-
RENE N5	7.0	8.0	2.0	5.0	6.2	-	-	7.0	3.0	-	0.2	-	-	-
IN100	10.0	15.0	3.0	-	5.5	4.7	-	-	-	-	-	0.18	0.004	0.06
MAR-M247	8.0	10.0	0.6	10.0	5.5	1.0	-	3.0	-	-	1.5	0.15	0.015	0.03
MC-NG	4.0	-	1.0	5.0	6.0	0.5	-	5.0	4.0	4.0	0.1	-	-	-

In order to increase their service properties, doping of the alloys with Cr, Co, Mo, W, Al, Ti, Nb, B, Fe, Y, Zr, Ta, Re, Ru, V, Ce, La, Si, Mn, Mg, Hf, Ir *etc.* is carried out. Some of the alloying elements are used to strengthen the nickel matrix by creating a solid solution. The other part forms excess phases: intermetallides, carbides and borides. Under operating conditions at high temperatures, the hardening phases dissolve and embrittlement is released. The later this occurs, the more heat-stable the alloy is. Frequently, new alloy compositions with sufficiently high initial heat resistance are not thermally stable and lose their properties pretty fast. In any case, the composition of the alloys is extremely diverse. As an example, we give the compositions of some common superalloys (see Table 1).

The development of the superalloys was inextricably linked with the history of the jet engine for which they were originally developed. The superalloys work in the hottest sections of turbines under the heaviest loads, with primary importance being given to ensuring the integrity of the components made from them. Currently, an active search for further improvements in the temperature characteristics of the superalloys is underway.

Nowadays, the single-crystal superalloys are being used in increasing quantities in the gas turbine engines as they exhibit excellent properties at high loads rather than the polycrystalline superalloys. New developments in the superalloy metallurgy are required for the next generation of ultra-efficient power generation systems that also use turbines. However, the issue of existing alloys application in power turbines is practically not considered in the scientific literature, since all alloys are mainly investigated in a rather narrow range of thermal exposures.

One of the possible solutions to the problem of adapting existing alloys to new products being developed is a comprehensive "structure-properties" analysis of the known materials. Synthesis of new materials has always been based on knowledge of the behavior of alloying elements, their participation in the formation of structural components, and their contribution to the complex of service properties of products. Often, the role of alloying elements is considered individually, which generates a lot of conflicting information. The analysis of the effect of the chemical composition on the service properties of the final products is an entangled and multidimensional task, especially when it comes to create a fundamentally new family of materials. For the well-known systems, it is advisable to take into account the entire experimental experience using statistical analysis and modern numerical methods. As a result, the chemical composition of the material is optimized to solve specific design problems, the field of application of materials is expanded, and the selection of specific alloys is facilitated.

As we know, for many materials and under loading conditions, which are invariant in time, the creep strain rate is considered constant; i.e. it approaches a steady-state [8]. Design against creep usually necessitates a consideration of the time to rupture. Its multiplication with the creep strain rate gives a constant, which is numerically equal to the creep ductility, i.e. the creep strain to failure. All this made it possible for certain test conditions (applied stress σ (MPa), time τ (hours), and temperature T (K)) to derive the complex Larson-Miller parameter (P_{LM}) (1) where 20 is the Larson–Miller constant, which varies with the alloy type. For any given material, the dependence of the applied stress on the Larson–Miller parameter is approximately straight, although there is a tendency for some non-linearity, which results in the lines curving gently downwards [9].

$$P_{LM} = (t + 273) \times (20 + lg\tau) \ . \quad (1)$$

The chemistry of the single-crystal superalloys has been refined for last decades in order to improve their service properties. Often, instead of natural modelling of the alloys properties, various mathematical models are applied. However, up to date, there is no unified model that describes the behavior of the alloys with a specific composition under various test conditions. The most frequently used models are various statistical ones. Moreover, since the behavior of the alloys' properties are often non-linear, the statistical approach based on utilization of artificial neural networks is considered the most promising since the neural networks models have proven to be a great tool that are able to simulate significant non-linearity [10].

In this work, we describe an approach to predict UTS of superalloys based on the knowledge of their compositions (especially, the content of principal alloying elements). For modelling, we use the Bayesian regularized artificial neural network (BRANN). The test conditions are combined in the complex Larson-Miller parameter. The neural network, which has been trained on the alloys composition and the service properties of some known superalloys predicts the unknown values of the Larson-Miller parameter that corresponds to sets of isothermal exposures for whose this alloy has not been studied, yet. Also, we compare the obtained results with the known experimental data

2 Approach and Experimental

The main tasks of this work is to determine the dependence of the heat resistance and thermal stability variations against the Larson-Miller parameters of the tests using a database with information on the chemical compositions and properties of high-temperature nickel-based superalloys. Another one is to reveal the impact of major alloying elements on the heat resistance and thermal stability and to compare it with the known data.

For modelling, an approach based on artificial neural networks is used. The input to the network is alloy composition and test conditions (t, τ). The task of the network is to predict the missing UTS values under the required conditions (t, τ). Validation of predicted data is carried out analytically. A trained neural network predicts the results for a selected sample of alloys (see Table 1) that did not participate in the training. The results of known tests and predictions are compared among themselves.

To solve the aforementioned problems, we have collected a database where the information on 308 nickel-based superalloys, containing their chemical compositions, heat resistances (UTS) under different test conditions has been stored. Data on alloys were collected from open sources (articles, catalogues, theses, etc.), as well as, in private conversations with representatives of manufacturers. About a half of the selected alloys are Soviet and Russian ones, the remaining half is made up by Western companies. The conditions and results of the tensile tests for these alloys formed 2,205 individual samples in the database, which later served as input data for neural networks training. Each known result of the strength tests under certain conditions of the isothermal exposure forms one sample.

In order to improve the information content of the database and to facilitate comparison of the test results for the heat resistance, the temperature and time of isothermal exposures were interpolated in accordance with the Larson-Miller parameter (P_{LM}) (1), however, we have modified it by dividing it with 10^5 (2) in order to obtain values of the same dimension as the alloying elements concentrations do (decimal format with values between 0 and 1). Physically, this only means the change of the input data scale and it does not affect the model. Moreover, the alloying elements concentrations have been normalized to the nickel concentration.

$$P_{LM} = T \times (20 + lg\tau) \times 10^{-5} \ . \quad (2)$$

Since the UTS range (σ, MPa) in experiments covers a band of several orders of magnitude (from one to thousands of MPa), it was decided using the logarithmic transformation (3). The use of the logarithm, also, makes prediction errors relative. This statement follows from the consideration that the expression (dy/y = const) is a differential equation of a logarithmic function. The network target values (y) for the inverse transformation (4) exclude the possibility of negative σ values, which are physically impossible. All this improves the predictive ability of the model.

$$y = \lg \sigma \ . \quad (3)$$

$$\sigma = 10^{y} \ . \quad (4)$$

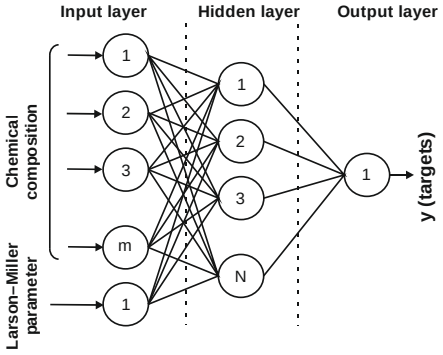
The network type selection has been made by analogy with the similar studies that present the results of superalloys compositions modelling [11–17] and with relation to our previous work [18]. The selected Bayesian regularized artificial neural network (BRANN) is more robust than standard back-propagation ones and is able to reduce or even eliminate the need of cross-validation. It has shown

satisfactory predictive ability along with resistance to overtraining.

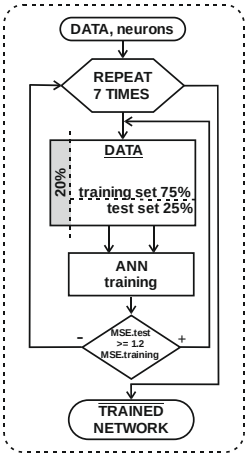
To create and train the BRANN, the MATLAB 2014a software package with the built-in MATLAB nntools was used. To improve the accuracy of simulations, a specially developed bootstrap algorithm and input data pre-processing were also applied. The configuration of the selected network and the training algorithm are shown in Fig.1.

We utilized a fixed configuration of the network that consisted of 13 neurons in the hidden layer (N=13). As the input data for the network, we have selected 23 detected alloying elements (m=23, with excluded balanced nickel) and P_{LM} for 308 alloys, which formed the array of the dimension (24×308). The output values are described by (3) and form the column vector (1×308).

The training procedure consists of seven cycles (corresponding to seven independent networks created) with a certain number of iterations (up to 200). Each iteration consists of wee (up to 30) training epochs. In each iteration, the input data vary based on a quintile decomposition of the input database. For the training cycle, only randomly selected 80% of initial data are applied. At each iteration, the formed dataset is randomly divided into the training (75%) and the test (25%) sub-samples that are involved in training of this particular network.



a)



b)

Fig. 1. ANN Architecture: (a) ANN configuration; (b) ANN training algorithm

The stop criterion for the iteration is the desired level of mean squared error (MSE) (5) achieved during the training. The target MSE value is chosen empirically and is set slightly below the level at which the MSE of training and test samples obtained after the next training epoch begin to diverge. In our case, based on preliminary study, we selected the following criterion: MSE.test ≥ 1.2×MSE.training.

$$MSE = \frac{\sum_{i=1}^{n}(y_{predicted\_i} - y_{fact\_i})^2}{n} \quad . \tag{5}$$

Thus, after numerous experiments, an artificial neural networks set with the highest accuracy of prediction ($MSE = 1.5 \times 10^{-3}$) has been selected. The set consists of seven networks that work in parallel. The further predictions made are calculated as average values of all seven predictions accounts for the back transformation (4). The final network performance is evaluated by the related root mean squared error (RRMSE) (6) of the known σ values predictions.

$$RRMSE = \sqrt{\frac{\sum_{j=1}^{k}\left(\frac{\sigma_{predicted\_j} - \sigma_{real\_j}}{\sigma_{real\_j}}\right)^2}{k}} \quad . \tag{6}$$

The performed ANN computations made it possible to supplement the missing parameters in the database on the service properties of high-temperature nickel-based superalloys. After that, the dependences of the long-term strength on the Larson-Miller parameter for all compositions of alloys contained in the database were reconstructed.

For most alloys, the dependence $\sigma(P_{LM})$ demonstrates a noticeable nonlinearity and the asymptotic tendency of σ to zero with P_{LM} greater than 30. Therefore, we have decided to approximate only the part of the area of the most intense structural changes (P_{LM} greater than 22) by the exponent using equation (7) under the assumption that $x \equiv P_{LM}$.

$$\sigma(x) = \sigma_0 \times \exp(-\frac{x - x_0}{p}) \quad . \tag{7}$$

were σ_0, x_0, p are the parameters that are established during the approximation. The area below P_{LM}=22 is out of practical interest and is not considered. The most common test conditions for alloys is an exposure of 1000 hours (τ=1000) at a temperature of

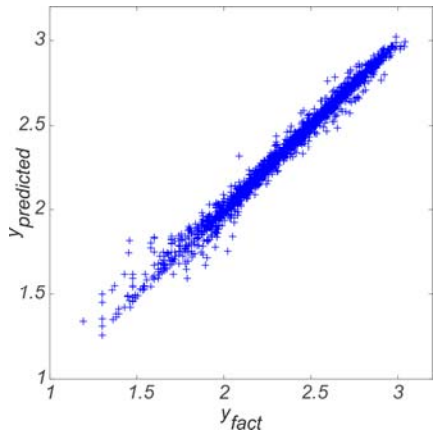
1000°C (t=1000). This condition (σ_{1000}^{1000}) is equal to x=29.3.

Therefore, for the further visualization of the results, we utilized UTS with the Larson-Miller parameter equal to 29 (σ(29)). The σ_0 parameter has no physical sense and was introduced just as the exponent (7) parameter; x_0 is a shift. The most physically inspired parameter in the model (7) is the slope factor p. It is p what responsible for the description of the thermo-temporal dynamics of the alloys phase stability. The higher the p, the slower the process of thermo-temporary degradation of the alloy structure goes.

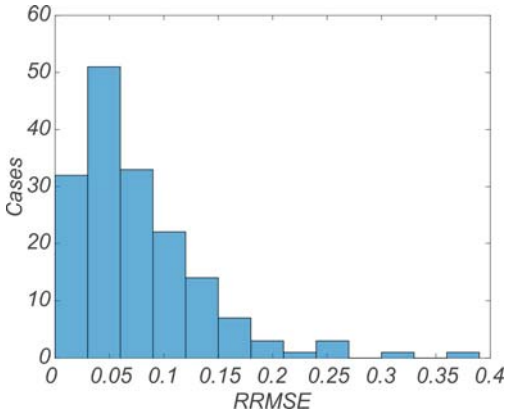
3 Results and Discussion

On the basis of artificial neural networks, a model for simulation UTS of nickel-based superalloys has been created. The network performance is reflected in Fig.2(a) as the dependence of predicted values vs experimental ones (R^2=0.986). In accordance with (3), the y parameter is an order of UTS values. As it can be seen (Fig.2(a)), the most residues are observed in the area of up to 100 MPa (y≤2).

The UTS vs P_{LM} dependencies has been built for the alloys from the validation sub-sample (Table 1) in order to verify the model performance. The errors (RRMSE) of the UTS neural network predictions distribution are shown in Fig.2(b).



a)



b)

Fig. 2. (a) Predicted y (3, absolute values) vs experimental ones total for training and test data sets; (b) the UTS ANN predictions errors (RRMSE) distribution

As it can be seen, the error is smaller than 0.20 for most observations. Moreover, the median value is 0.06 and 75% percentile is just 0.10. The RRMSE values more than 0.2 are corresponded to the y values (see Fig.2(a)) less than 2. Thus, we might define the model as successful and the results as sufficiently precise. The greatest prediction errors of the model are localized in the area of stresses that does not represent much practical interest (100 MPa).

The examples of the model predictions of UTS together with the real experimental data and with approximations by (7) for six common superalloys (see Table 1) are shown in Fig.3. The figure shows the satisfactory accuracy for both, the ANN predictions and the exponential approximation. The slope factor p characterizes how close the exponential curve is to the abscissa axis, thus, we may deduce that p is an indirect indicator of the alloy thermal stability. The larger the p, the longer the structure resists destruction. It is also necessary to note the following important point resulting from image analysis. It is obvious that the approximation of the dependence of the heat resistance of the alloys on the Larson-Miller parameter by the exponent is valid only in the range of intense structural changes. Approximation of the entire range requires a more complex model that has yet to be developed. We might suggest the dependence would be sigmoidal.

For the further validation of the model, we have applied the calculated values of the heat resistance (σ) and the thermal stability (p) to visually assess the effect of particular doping. We took new data on molybdenum, chromium, rhenium, tantalum, cobalt, and tungsten (see Fig. 5) in comparison with known

information [19] (see Fig. 4) as these elements make the main contribution to the hardening of alloys.

The results of modelling show that Ta, Re (and Ru, as the literature confirms) increase the high-temperature strength of alloys; W has no significant influence; Cr, Co and Mo reduce it. These results are in good agreement with the experimental data given in [20], which confirms their adequacy. Two elements (Co and Mo) demonstrate inconsistent behavior when compared with [19], however, our data belong to a substantially wider range of chemical compositions, and consequently more trustworthy. Furthermore, we provide an initial background for the linear trends. Frankly speaking, it is obvious that linear regressions do not reflect the nature of the alloys properties behavior; however, we still have no reason to introduce another approximation model.

The exponential slope factor p demonstrates a weak dependence on the content of a particular element. The exceptions are possibly tungsten (there is a slight decrease in the thermal stability of the alloy with an increase in the concentration of the element) and molybdenum (the opposite is observed).

down the degradation. We could also add tantalum to this list. The issue of the mutual influence of alloying elements still raises a plenty of disputes. Partly, the answers are given in the framework of quantum chemistry, though such an analysis is beyond the scope of our work.

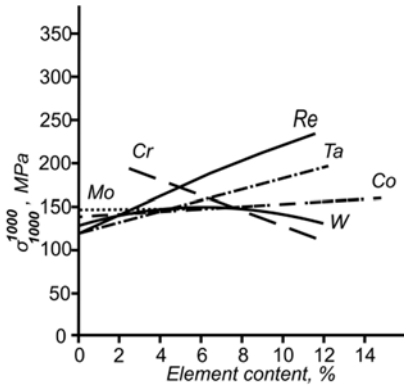


Fig. 4. The experimental dependences of the heat resistance against the content of certain alloying elements [19]

We should also add some more explanations from the metallurgical point of view. The basis of alloys' heat resistance is created by a complex structure: γ-nickel solid solution, the main

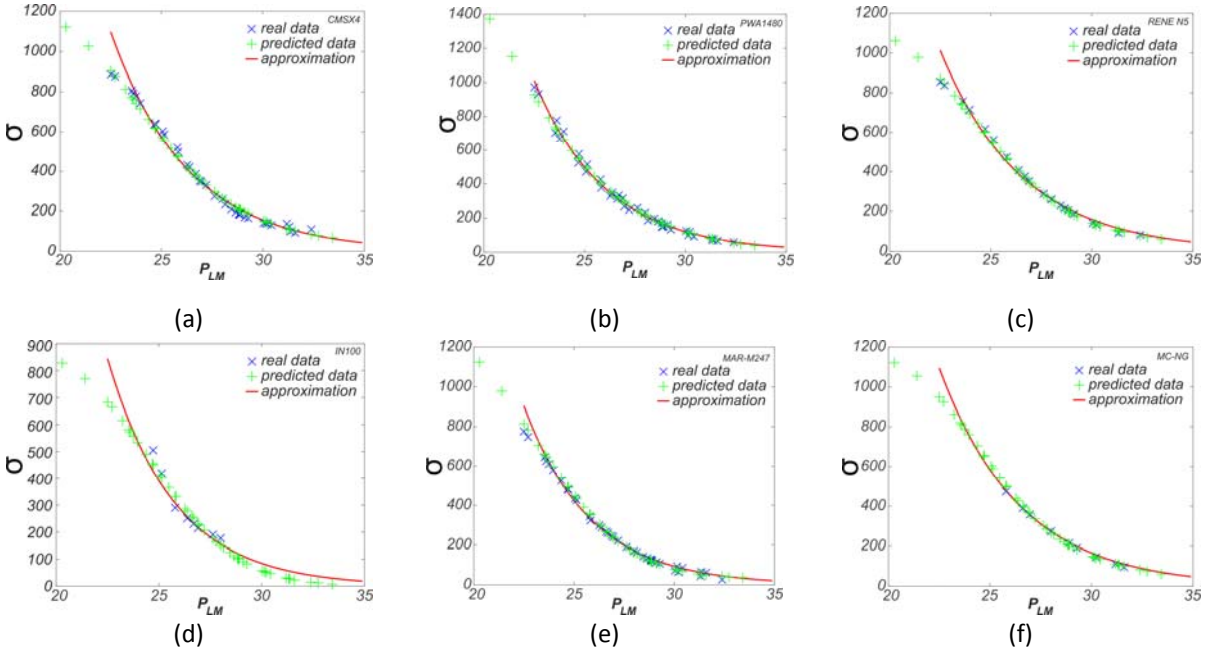


Fig.3. Calculated data on the effect of certain alloying elements on the ultimate tensile strength of nickel alloys

As follows from numerous studies, the overdeposition of high-temperature nickel-based alloys leads to an unbalanced phase composition and, consequently, to a decrease in thermal stability. Only rhenium and ruthenium block the development of embrittlement phases and substantially slow

reinforcing intermetallic γ'-phase with stoichiometry $(Ni, Co)_3(Al, Ti...)$, MC carbides, γ + γ' and MC+MB eutectics. There are two main mechanisms of hardening: solid solution and dispersion. They provide enhanced strength properties of alloys. Since the products that are

made of the alloys operate at high temperatures, certain changes in their structure are resulted by the influence of diffusion processes. Accordingly, the structure is degraded. The rate or degree of the structure degradation is described by another substantial property of thermal stability or phase stability. The higher the thermal stability, the longer the operation time of the product. Previously, thermal stability was described only empirically, according to morphological and quantitative changes in excess phases evaluation. The proposed analytical quantitative assessment of thermal stability as a coefficient p in the approximation formula (7) opens up new perspectives in modelling the properties of the alloys.

The heat resistance of the alloys, as a rule, is compared directly by the heat resistance change depend on the Larson–Miller parameter. The experimentally observed points commonly form these dependencies. However, this is not correct, since UTS characterizing the heat resistance cannot be less or equal to zero, just as the Larson-Miller parameter cannot take negative values. Therefore, this relation can only be described by an asymptotic (in particular, exponential) dependence.

For a computational experiment using ANNs, a database containing information on 308 cast chemical compositions of high-temperature nickel-based superalloys and on the results of measurements of UTS of single-crystal samples prepared from these alloys was created. Firstly, the ANN simulated the missing results for each alloy composition. Then, graphs of dependences of UTS on the Larson-Miller parameter for some well-known alloys were plotted (see Fig. 3). From the above results, it follows that all the graphs have the same exponential appearance of change in heat resistance with an increase in the Larson-Miller parameter, however, on each graph one can distinguish a point that characterizes a particular alloy. This is the locality of transition of the intense heat resistance decrease to a flatter area. It is obvious that the decrease in heat resistance is associated with the processes of the original structure degradation.

4 Conclusion

The high-temperature nickel-based superalloys are unique materials with complex doping and a heterophasic phase composition. This paper has addressed the following issues concerning the superalloys: modelling the variation in heat resistance under different values of the Larson-Miller parameter; definition of the thermal stability as a numerical parameter; modelling the influence of the alloying elements on the heat resistance and the thermal stability.

To solve the problems, we have engaged the following toolkit: the common Bayesian artificial neural network with a specially developed bootstrap training procedure; the database of 308 known superalloys contents and service properties that formed 2,205 individual samples for ANN training. We trained the network, then we predicted the unknown values of the alloys UTS (heat resistance), and thus filled the database. We approximated the dependence $\sigma(P_{LM})$ by the exponential equation (7). Based on the complete database, we verified the model performance by assessing the RRMSE values (the median RRMSE is 0.06) and by plotting the $\sigma(P_{LM})$ dependence together with the approximations for six common superalloys from the sub-sample, which did not take part in the training.

The verification has shown good predictive accuracy of the ANN model, as well as, the satisfactory description of the $\sigma(P_{LM}>22)$ dependence by an exponent. Nevertheless, we suggest that for the whole $\sigma(P_{LM})$ range, the better approximation equation is sigmoidal. This issue must be studied further. Thus, ANN calculations have helped to obtain the dependences of the heat resistance and the thermal stability on the Larson-Miller parameter. The results of the calculations coincide with the experimentally obtained data, which confirms the model adequacy.

From the metallurgical point of view, the structure of the cast metal is heterophasic: γ-solid solution, γ’ secondary phase, MC carbides, γ + γ’ eutectics, MC + MB (carbo-boride) eutectics and so on. The decrease in heat resistance with increasing Larson-Miller parameter is explained by the degradation of the structure with the release of new embrittling phases: TCP, M_6C, $M_{23}C_6$. The cuboid particles of the γ’ secondary phase are first coagulated, breaking down the partially coherent bond with the γ- matrix, then coalescing into blocks, after which they break up into a few rounded small particles and dissolve in the γ- matrix. Carbide eutectics of font morphology also break up into colonies of small round particles along the boundaries of polyhedral M-C carbides. Reactions also occur with the release of two new M_6C and $M_{23}C_6$, related to TCP phases. Eutectic γ, γ’ and

MC+MB also change their morphology and the number of secretions. The instability of the structural components leads to the nucleation and development of other TCP phases: sigma, Laves, etc. The meta-stability of the structure leads to the weakening of alloys, consisting in a two-stage decrease in heat resistance.

Since the basis of the heat resistance is the dispersion hardening by the intermetallic γ' phase, the solid solution and carbide hardening mechanisms still work at the end of the processes of the alloy structure degradation after completion of dissolution of the main hardening phase, in particular, if carbon is introduced into the alloy. The spread of predictions on the sloping area of the dependence of UTS by the Larson-Miller parameter is associated with these phenomena.

During the exponential approximation of the dependence, $\sigma(P_{LM})$ we found a characteristic feature for each composition of the alloy. It is the exponential parameters p, characterizing the thermal stability of the alloys. Dependencies of stress and slope on the composition of elements, judging by the plots (Fig.5), have opposite trends. All these suggest that the simultaneous achievement of heat resistance and thermal stability is a sophisticated issue, the solution of which cannot be based on the use of linear models. Moreover, taking into account the mutual influence of two or more alloying elements on the properties of the alloy, even using non-linear regression models, is an extremely resource-intensive task. Thus, approaches based on the application of artificial neural networks that do not require significant computational power, and at

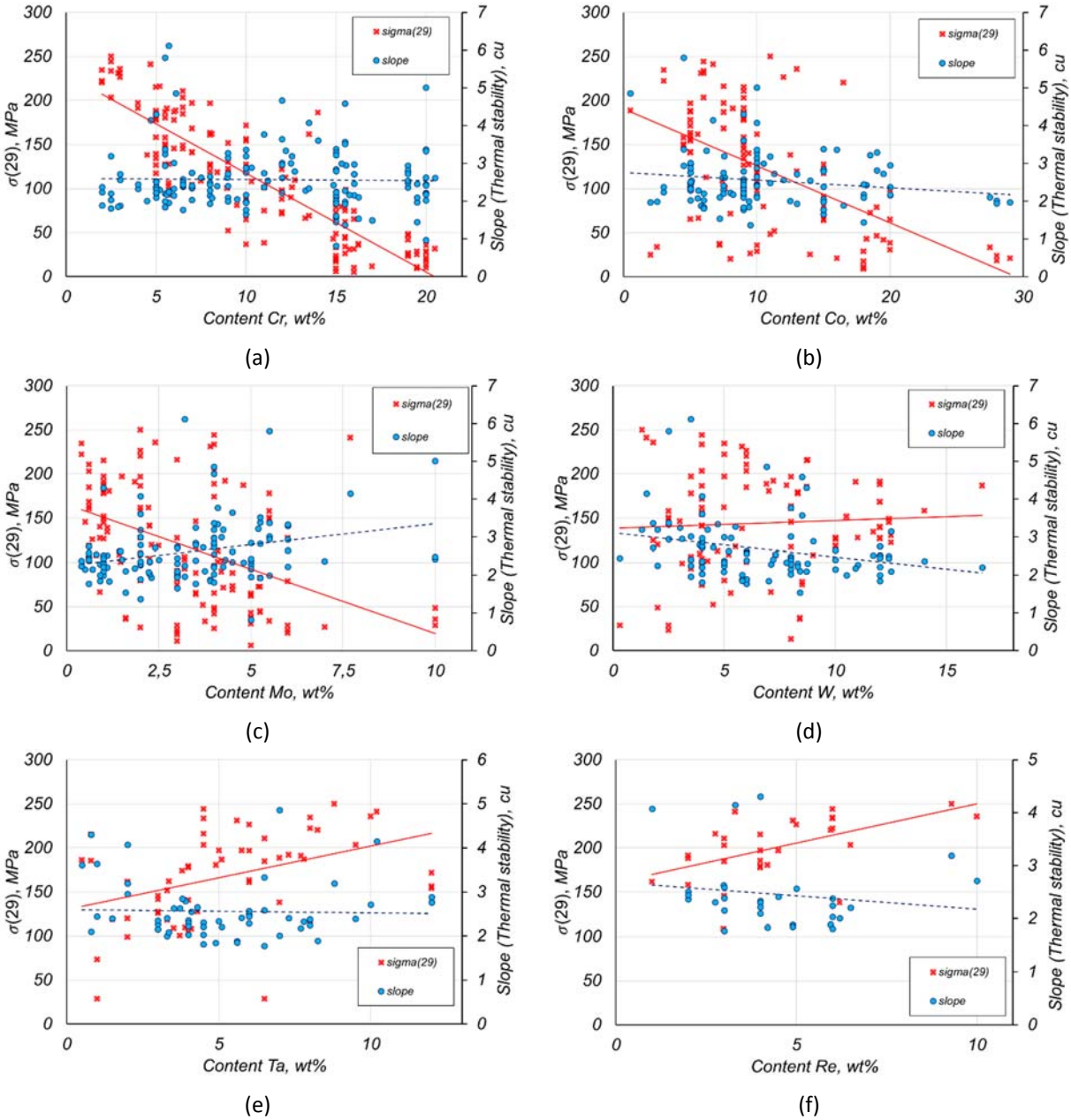


Fig.5. The effect of a particular element content (wt%) on the UTS σ(29) and thermal stability (p or slope): (a) Cr, (b) Co, (c) Mo, (d) W, (e) Ta, (f) Re; the solid lines indicate the linear regressions of UTS difference, the dotted lines indicate the linear regressions of the thermal stability difference

the same time have high modelling accuracy (with the correct training algorithms), seem very promising in solving such problems.

We tried to find similar works in which some mathematical method of predicting UTS would be applied. However, we were not able to detect them. Therefore, a comparison with any other model is currently impossible. The success of the neural model may inspire researchers to use regression models to solve similar problems. We hope this issue will find its development in the near future.

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