Mathematical Modeling of the Corrosion Behavior of Austenitic Steels in Chloride-Containing Media During the Operation of Plate-Like Heat Exchangers

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Article info	Abstract
	Mathematical models that describe the dependences of the critical temperatures of pitting formation of AISI 304, 08Kh18N10, AISI 321, 12Kh18N10T steels in model circulating waters with pH 4…8 and chloride concentrations from 350 to 600 mg/l on their chemical composition and structure have been developed. They are based on linear squares regressions and a feed-forward neural network for reduced feature numbers. Using the developed mathematical models, it was found that the critical pitting temperatures of these steels increase with an increase in the pH of the circulating water, the number of oxides up to 3.95 μm in size, the average distance between titanium nitrides, the Cr content and a decrease in the concentration of chlorides in the circulating waters, the average distance between oxides and average austenite grain diameter. At the same time, it was found that the geometric dimensions of the steel structure most intensively affect their pitting resistance in circulating waters, and the effect of their chemical composition is minimal and is determined by the amount of Cr, which contributes to an increase in the pitting resistance of steels, probably increasing the solubility of nitrogen in the austenite solid solution. It is proposed to use the developed mathematical models to select the optimal heats of these steels for the production of heat exchangers and predict their pitting resistance during their operation in circulating waters.

1. Introduction

Corrosion-resistant steels of the austenitic class are widely used in the production of heat-exchange equipment, given their high corrosion resistance in many environments [1–4]. Currently, plate-like heat exchangers are widely used, because they are more compact than shell-and-tube heat exchangers, and also have less weight and more efficient thermal conductivity due to a significantly smaller thickness (0.3 ... 1.0 mm versus 1.0 ... 3.0 mm) of heat transfer elements. However, the latter cir-

cumstance increases the likelihood of perforation of plates of plate heat exchangers in the case of pitting corrosion in circulating waters, which are used to cool technological products in chemical, oil, and gas refining, energy, and other industries [5–8]. Therefore, the assessment of the pitting resistance of construction materials from which heat exchangers are made and the prediction of their corrosion behavior during operation is an actual problem. It was established in [9–11] that the parameters of circulating water and the structural heterogeneity of AISI 304, 12Kh18N10T, 08Kh18N10, AISI 321 steels significantly affect their pitting resistance in circulating water, while the effect of their chemical composition is not so

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significant and is determined only by the amount of Cr in their composition. To determine the role of chromium and the structural components of these steels in their pitting resistance and its prediction, mathematical models were built that are based on linear quadratic regressions and a two-layer neural network of direct signal propagation for a reduced number of input features.

2. Materials and research methods

Five industrial heats of steels of the austenitic class AISI 304, AISI 321, and one 12Kh18N10T and 08Kh18N10 were studied. Their chemical composition is presented in (Tables 1 and 2), and structural heterogeneity was determined earlier in [9–10].

The data-driven approach allows building a model on the experimental data only without any expert knowledge (physical, chemical, etc. theoretical models). Such a technique provides an opportunity for model building in insufficiently explored problems.

Mathematical models of the dependence of the critical pitting temperature (CPT) of steels depending on their chemical composition (Tables 1 and 2), structural heterogeneity, and parameters of

model circulating waters (pH 4…8, chloride concentration CCl = 350, 400, 500, 550, 600 mg/l). These parameters have the greatest effect on the pitting resistance of steels, since the ratio of the concentrations of chlorides in them to other anions (sulfates, nitrates, etc.) does not reach a critical value, and the rate of recycled water outflow is laminar [12]. Linear quadratic equations were constructed using these parameters (1). We have used a standard feed-forward neural network, widely described in the literature [13]:

$$y = \sum_k w_k c_k , \qquad (1)$$

where: y – is the critical pitting temperature (CPT) of steels, °C; w_k – the weight coefficient of the components (see Table 3); c_k – the feature component x_i (see Table 3).

In particular, the output feature of the model (1) is the CPT of steels AISI 304, 08Kh18N10, AISI 321, 12Kh18N10T in model circulating waters, and the variable features x_i are the indicators of model circulating waters (pH(x_1); chloride content (x_2), mg/l); components of the steel structure (x_3 – the volume of oxides, rot.%; x_4 – the amount of oxides up to 1.98 μm in size per 100 microscope

Table 1
Chemical composition of steels AISI 304 and 08Kh18N10

# of heat	Content of chemical elements, mass%								
	C	Mn	Si	Cr	Ni	N	Ti	S	P
1	0.071	1.23	0.22	17.96	9.34	0.048	-	0.001	0.027
2	0.067	1.74	0.50	18.22	8.09	0.046	-	0.001	0.028
3	0.075	1.65	0.43	18.25	8.09	0.055	-	0.004	0.024
4	0.050	1.70	0.41	18.30	8.10	0.044	-	0.002	0.028
5	0.030	1.81	0.39	18.10	8.20	0.039	-	0.001	0.034
08Kh18N10	0.060	1.34	0.32	17.44	9.77	–	–	0.006	0.035

Table 2
Chemical composition of steels AISI 321 and 12Kh18N10T

# of heat	Content of chemical elements, mass%								
	C	Mn	Si	Cr	Ni	N	Ti	S	P
1	0.035	1.66	0.54	17.10	9.10	0.012	0.32	0.001	0.026
2	0.060	1.59	0.66	16.43	9.14	0.011	0.34	0.002	0.027
3	0.064	1.22	0.52	17.43	9.70	0.012	0.41	0.001	0.026
4	0.030	1.62	0.41	17.41	9.24	0.013	0.31	0.002	0.028
5	0.040	1.70	0.49	17.70	9.10	0.013	0.35	0.001	0.026
12Kh18N10T	0.070	1.70	0.49	17.97	10.46	–	0.46	0.007	0.027

fields (×320), units; x_5 – the amount of oxides with sizes from 1.98 to 3.95 µm per 100 fields of view of a microscope, units; x_6 is the average distance between oxides, µm; x_7 is the average austenite grain diameter, µm; x_8 is the volume of δ-ferrite, rot.%; x_{19} is the volume of titanium nitrides, rot.%; x_{20} is the average distance between them, µm) and their chemical composition (x_9 – carbon content, wt.%; x_{10} – manganese; x_{11} – silicon; x_{12} – chromium; x_{13} – nickel; x_{14} – nitrogen; x_{15} – titanium; x_{16} – sulfur; x_{17} – phosphorus) and x_{18} – specific magnetic susceptibility, m³/kg [12].

A neural network model based on a two-layer feed-forward neural network for a reduced number of input features (x_1, x_2, x_6, x_{10} and x_{12}) is described by formula (2) [14]. To simulate and train the neural network we have used online scripts of MATLAB. The training has been provided using Levenberg-Marquardt algorithm. We have used all the data as training and validation, because of the small data set size. We understand that to assess the suitability of the model, it is advisable to test it on data that was not used for training. However, due to the limited sample size, the observations were forced to use the entire sample. The resulting error estimate may be underestimated to the data that were not used in the construction of the model. This value is given as a guideline.

$$ y = w_0^{(2,1)} + \sum_{i=1}^{15} w_i^{(2,1)} \psi^{(1,i)} \left(w_0^{(1,i)} + \sum_{j=1}^{7} w_j^{(1,i)} x_j^{(1,i)} \right), $$

(2)

where $\psi^{(1,i)}(a) = \dfrac{2}{1+e^{-2a}} - 1$ – activation function of the i-th neuron of the first layer of the network,

$w_j^{(1,i)}$ – weight coefficient of the j-th input of the i-th neuron of the network's first layer,

$w_i^{(2,1)}$ – weight coefficient of the i-th input of the single neuron of the network's second layer.

The values of the weight coefficients $w_j^{(1,i)}$ and $w_i^{(2,1)}$ are presented in (Table 4).

The weight coefficients of the regression model (1) were determined by the least squares method, and the quality of mathematical models was evaluated by the sum of squares of instantaneous errors:

$$ E = \sum_{s=1}^{s} (y^s - y^{s*})^2 , $$

(3)

where: y^{s*} – calculated value of the output feature for the s-th instance of observations (CPT); – y^s the value of the output feature for the S-th instance of observations (CPT) determined experimentally [12].

The S is the number of instances (observations) in a sample. We use the all data for model building. We have used online scripts of MATLAB for all model building. This software uses a least-squares method for regression models.

3. Research results and discussion

Analysis of the C_k constituent of the developed linear squares regression model (1), taking into account the established weight coefficients W_k (Table 3), showed that the CPT of the studied AISI 304, 08Kh18N10, AISI 321 and 12Kh18N10T steels increases by 54.2 °C with increasing pH(x_1) of model circulating water from 4 to 8 (see item 1 of Table 3) and decreases by 12.0 °C with an increase in the concentration of chlorides in it from 350 to 600 mg/l. This trend is harmonized with well-known literature data [15–18]. At the same time, it should be noted that the square of the constituent x_2^2 (Table 3, item 6), taking into account its weight coefficient. $w_k = -1.67 \cdot 10^{-5}$ the increase in the concentration of chlorides $x_2(C_{Cl})$ within the above-mentioned limits has practically no effect on the value of y(CPT) of the steels under study. At the same time, for the constituent x_1^2 (pH), an increase in pH(x_1) of model circulating waters from 4 to 8 contributes to a decrease in y(CPT) of steels by 47.3 °C (Table 3, p. 5). At the same time, taking into account that for the constituent x_1 (Table 3, item 1) an increase in its value within the above limits contributes to an increase in y(CPT) of steels by 54.2 °C, and for a decrease by 47.3 °C, the total the well-known tendency to increase the CPT of steels in chloride-containing media is not violated. Therefore, we can state the fact that y(CPT) of the studied steels increases on average by 6.9 °C with an increase in pH(x_1) of model circulating waters from 4 to 8. This value is harmonized with experimental data [19, 20].

In the AISI 304 and 08Kh18N10 steels under study, titanium nitrides are absent, because, unlike AISI 321 and 12Kh18N10T steels, they are not stabilized by titanium (Tables 1 and 2). At the same time, in AISI 321 and 12Kh18N10T steels, the titanium content varied from 0.32 mass% (heat 1) to 0.46 mass% (steel 12Kh18N10T), and the volume of titanium nitrides – from 0.2336 rot.% (heat 4)

up to 0.4745 rot.% (heat 1). Thus, taking into account the data (Table 2) on the content of nitrogen and titanium in AISI 321 and 12Kh18N10T steels, it can be noted that the volume of titanium nitrides (x_{19}) in these steels does not depend on the content of these chemical elements in them. In addition, according to (Table 3; p. 4, 15), a change in the volume of titanium nitrides in these steels from 0 rot.% in steel AISI 304 and 08Kh18N10 to 0.4745 rot.% heat 1 of AISI 321 steel does not affect their y(CPT) in model circulating waters. In this case, according to (Table 3, p. 16) and the data of [21], an increase in the average distance between titanium nitrides from 66 μm in heat 5 of AISI 321 steel to 91 μm in heat 2 promotes an increase in their y(CPT) by 99 °C. Thus, it turns out that the greater the average distance between titanium nitrides in these steels, the smaller their size and the higher y(CPT), which characterizes pitting resistance. These results are harmonized with the data of [22] in terms of the critical size of inclusions at which stable pits can form and develop near them.

The volume of oxides (x_3) in the studied steels varies in the range from 0.0027 rot.% for steel 12Kh18N10T to 0.0324 rot.% for heat 4 of AISI 304 steel, therefore, according to (Table 3; #7), the influence of this parameter on y(CPT) is practically absent. Similarly to titanium nitrides and oxides, a change in the volume of δ-ferrite in these steels (x_8) in the range from 0.0138 rot.% heat 3 steel AISI 304 to 0.372 rot.% steel 12Kh18N10T practically does not affect their y(CPT) in model circulating waters (Table 3, # 3, 12). Such results are consistent with the data of works [9] for steel AISI 321 and [10, 23] for AISI 304. At the same time, according to the data (Table 3, items 8, 9), 1 and 54 °C with an increase in the amount of oxides in them up to 1.98 microns in size (x_4) from 12 units steel 12Kh18N10T to 425 heat 1 steel AISI 304 and from 1.98 to 3.95 microns (x_5) from 2 units steel 12X18H10T up to 280 units steel 08Kh18N10, respectively. Such dependencies are due to the fact that stable pittings are formed near inclusions that are larger than the critical size [23, 24], for austenitic steels, about 5 μm [25]. Accordingly, in oxides up to 3.95 μm in size, metastable pits were formed, which were repassivated with time. At the same time, it is known [26] that the more metastable pits on the steel surface, the lower the probability of their transition to a stable state. At the same time, it should be noted that stable pittings can form near inclusions much smaller than 5 μm if they are located at the boundaries of

Table 3
Feature constituents x_i and their weighting coefficients

#	Additive component c_k	Weight coefficient w_k
1	x_1	13.54
2	x_2	-0.0481
3	x_8	0.0153
4	x_{19}	-0.0493
5	x_1^2	-0.9845
6	x_2^2	-1.67×10^{-5}
7	x_3^2	-8.34×10^{-4}
8	x_4^2	1.22×10^{-4}
9	x_5^2	6.9×10^{-4}
10	x_6^2	-2.76×10^{-4}
11	x_7^2	-0.0014
12	x_8^2	5.46×10^{-6}
13	x_{10}^2	-0.0076
14	x_{12}^2	0.1735
15	x_{19}^2	1.02×10^{-5}
16	x_{20}^2	0.0254

austenite grains. It is obvious that the larger the average austenite grain diameter, the lower the probability of inclusions intersecting with grain boundaries and, accordingly, the lower the probability of pitting corrosion and the higher the CPT of the steels at which it develops.

Analysis of the data (Table 3, # 11) of the model (1) is harmonized with this statement because it was found that y(CPT) of the steels under study decreases by conditional 135.5 °C with an increase in the average austenite grain diameter from 23 to 312 μm because the larger the average grain diameter, the higher the degree of the incoherence of adjacent grains and the higher the defectiveness of their boundaries. In addition, it should be noted that the theory of the formation of stable pits at the intersection of austenite grain boundaries in steels with small inclusions is harmonized with the results of analysis (Table 3, item 10) of the model (1), because it was found that y(CPT) of the steels under study decreases by the conditional 167.8 °C with an increase in the average distance between oxides from 150 to 794 μm [27]. Since it is obvious that the greater the average distance between inclusions in steels, the smaller their size and the greater the number. It turns out that the

greater the amount of oxides in the studied steels, the higher the probability of their intersection with the boundaries of austenite grains and the higher the probability of pitting corrosion. Summarizing the above, it can be noted that the risk of pitting corrosion of the steels under study decreases with an increase in the amount of oxides up to 3.95 μm in size, located in the austenite solid solution, due to the redistribution of the anode current density between a large number of metastable pittings that located near these oxides, at a decrease in the degree of incoherence between adjacent austenite grains due to a decrease in their average diameter, as well as a change in the amount of small oxides due to a decrease in the average distance between them.

Studies of the pitting resistance of AISI 304 and AISI 321 steels showed that it mainly depends on the parameters of model circulating waters (x_i, x_2), the structural heterogeneity of these steels (x_6, x_7), and to a lesser extent, on the content in them Cr within the standard. At the same time, the results of the analysis (Table 3, # 13, 14) of the mathematical model (1) are harmonized with the data of works [9, 10, 26], because it was found that y(CPT) of the steels under study practically does not change with increasing Mn content from 1.22 to 1.81 mass% (Tables 1 and 2). At the same time, it increases by 7.4 °C with an increase in the Cr content in steels from 17.1 to 18.3 mass% (Table 1). The pitting resistance of steels and alloys alloyed with Cr is associated with oxide films formed by Cr with O [28–30]. In addition, this element affects the solid-phase diffusion of Cr, Ni, and Fe atoms to the surface of metastable pits and promotes their repassivation [25, 26]. At the same time, there is evidence that Cr [31] and, especially, manganese increase the solubility of nitrogen in corrosion-resistant steels, and, consequently, their pitting resistance. The resistance of austenitic steels alloyed with nitrogen to pitting corrosion is associated with an increase in the stability of the oxide film. Mechanisms for improving the passive stability of nitrogen alloyed steel films include:

– formation of ammonium ions or nitrate (nitrite) ions ;

– segregation of nitrogen on the surface during anodic dissolution ;

– formation on the surface bonds of Cr-Ni ;

– complex formation of ammonia or NO and ammonium salts.

In addition, it is considered that the positive effect of nitrogen is due to the inhibitory effect on the anodic dissolution of steel. In particular, there is evidence that nitrate ions stabilize the passive film, preventing the adsorption of chlorides and increasing the resistance of austenitic steels to pitting corrosion in chloride-containing media. It should be noted that the proposed mechanisms for increasing the pitting resistance of steels when alloyed with nitrogen involve the action of nitrogen dissolved in the solid solution of steel austenite. At the same time, the results of the analysis of the mathematical model (1) (Table 3) showed that N in the amount of 0.011 ... 0.013 mass% in heat of AISI 321 steel and 0.032...0.055 mass%. affects their y(CPT), i.e. pitting resistance in model circulating waters. This is due to its low solubility in the solid solution of steel austenite. However, as mentioned above, Cr increases its solubility in solid solution. Thus, nitrogen dissolved in the solid solution of austenite, by one or more of the above mechanisms, protects austenitic steels from pitting corrosion. Thus, summarizing the above data, it can be noted that the pitting resistance of austenitic steels AISI 304, 08Kh18N10, AISI 321, 12Kh18N10T, is determined by the parameters of recycled waters (pH (x_1), C$_{Cl^-}$(x_2)), which make up the structure (x_4, x_5 – the number of oxides sizes up to 1.98 and 1.98…3.95 μm, respectively; x_7 is the austenite grain diameter; x_6, x_{20} is the average distance between oxides and nitrides of titanium, respectively) of steels and their Cr content. For other chemical elements in the studied steels (Tables 1 and 2), the volume of titanium oxides and nitrides do not affect their y(CPT) and, accordingly, pitting resistance.

At the same time, it should be noted that the root-mean-square error of determining y(CPT) of the studied steels using the mathematical model (1) (Table 3) is 3820.4 and the mean error is 0.0028. Thus, this mathematical model can be recommended to the industry for predicting the pitting resistance of heat exchange equipment using water circulation systems, as well as for selecting heats of these steels with optimal pitting resistance, depending on the operating conditions of this system. In addition, the developed mathematical model can be useful in the development of new steel grades proof to pitting corrosion.

The developed neural network model based on a two-layer neural network of forward propagation for a reduced number of input features (x_1, x_2, x_6, x_{10} and x_{12}) (2) makes it possible to obtain much more accurate calculated values of y(CPT) for the steels under study, depending on the pa-

rameters of circulating water (x_1, x_2), their structural element (x_6) and chemical elements (x_{10}, x_{12}) than the mathematical model (1). Since the total squares error for model (2) is 1.7994 (3), and the average $|y_{ex}-y_{calc}| - 0.0026$. In this case, the error in determining the CPT of the studied steels during the experiment is ±0.5 °C. The disadvantage of the mathematical model (2) is the inability to estimate the quantitative effect of the parameters of the model circulating water, structural heterogeneity and chemical composition of the steels under study on their y(CPT). The values of the weight coefficient ($w_j^{(1, i)}$) of the j-th input of the i-th neuron of the network's first layer and the weight coefficient of the i-th input of the single neuron of the network's second layer ($w_i^{(2, 1)}$) are presented in (Table 4).

It should be noted that in the mathematical model (1) based on linear squares regressions, the following x_i variables are significant: (x_1, x_2 are the pH of the model circulating waters and the concentration of chlorides in them); x_4, x_5 – the amount of oxides in the studied steels up to 1.98 μm in size and from 1.98 to 3.95 μm; x_6, x_{20} – the average distance between oxides and nitrides; x_7 is the average austenite grain diameter and x_{12} is the chromium content in steels. And in a neural network model based on a neural network of direct signal propagation for reduced numbers of features (2): (x_1, x_2, x_6, x_{10} and x_{12}). Moreover, these features are common to both mathematical models. Thus, it turns out that these features are the most important in terms of their influence on the pitting resistance of the steels under study. In this case, the proposed mechanisms of the effect of these features on y(CPT) of steels AISI 304, 08Kh18N10, AISI 321, 12Kh18N10T are described above.

Table 4

Values of the weight coefficients ($w_j^{(1, i)}$) of the j-th input of the i-th neuron of the network's first layer and the i-th input of the single neuron of the network's second layer ($w_i^{(2, 1)}$)

$w_j^{(1, i)}$	i	1	2	3	4
	0	-2.5702	-0.0005	0.0019	4.9133
	1 (x_1)	-1.8387	0.1347	0.2015	-1.9649
	2 (x_2)	0.7325	0.2347	-0.2193	0.065
j	**6 (x_6)**	-3.5582	11.3589	0.2667	1.5316
	10 (x_{10})	-20.461	12.6135	0.1583	0.0165
	12 (x_{12})	-20.6655	5.4093	0.1532	-0.2049
i	0	1	2	3	4
$w_i^{(2, 1)}$	-2.3433	17.5420	-1.0258	0.0047	8.3758

4. Conclusions

Two mathematical models have been developed, which are based on linear squares regressions and a feed-forward neural network for reduced numbers of features. They are proposed to be used to select the optimal heats of AISI 304, 08Kh18N10, AISI 321, 12Kh18N10T steels and predict the pitting resistance of plate heat exchangers from them in circulating water.

It has been established that their pitting resistance increases with an increase in the pH of the circulating water, the amount of oxides up to 3.98 μm in size, the average distance between titanium nitrides, the Cr content and a decrease in the concentration of chlorides in the circulating water, the average distance between the oxides and the average diameter of the austenite grain.

Based on the dependencies obtained and based on well-known literature data, the mechanisms of the influence of the parameters of circulating water, structural elements and the chemical composition of the studied steels on their pitting resistance in circulating water are proposed. In particular, metastable pittings are formed in a solid solution of steel austenite near oxides (1.98–3.95 μm) and repassivate without reaching a critical size of about 5 μm, which contributes to an increase in their pitting resistance.

The smallest oxides (up to ~ 1 μm) at the intersection with the boundaries of austenite grains contribute to the formation of stable pits, due to the highest degree of the incoherence of adjacent grains, which grows with an increase in the average diameter of austenite grains. Chromium promotes an increase in the solubility of nitrogen in the austenite solid solution and repassivation of pits under the action of anions of nitrogen compounds.

Acknowledgments

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