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Corrosion behavior of austenitic steels in chloride-containing media during the operation of plate-like heat exchangers

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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 on a feed-forward neural network for a 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 amount 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.

Key words: plate-like heat exchanges, circulating water, pitting corrosion, structure, neural networks.
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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 [1-5]. 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 circumstance 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 the chemical, oil and gas refining, energy, and other industries [6–10]. Today, particular successes have been made in the field of studying corrosion properties of structural steels: the effect of chemical composition and roughness on pitting corrosion [10–12], the destruction of AISI 304 steel in a marine environment underwear and corrosion conditions [13], as well as the effect of tribocorrosion conditions on pitting susceptibility and synergistic material loss for AISI 304 stainless steel [14]. In addition, it was found that parameters of recycled waters and structural heterogeneity of AISI 304, 12Kh18N10T,

8Kh18N10, AISI 321 sheets of steel significantly affect their pitting resistance in recycled waters, and the effect of their chemical composition is not so significant, and is determined only by the amount of Cr in their composition [15-17].

However, to date, there is no information in the literature on modeling and assessing the pitting resistance of structural materials from which smelting exchangers are made. Nowadays modern models describing the dependence of corrosion-resistant steels' and alloys' CPT on the parameters of circulating water (pH, chloride concentration) and steel parameters (chemical composition and structural heterogeneity) do not currently exist. Therefore, the prediction of corrosion behavior during operation and finding a correlation between model prediction and experimental data is an urgent problem.

The main goal of this work is to model the corrosion behavior of structural steels AISI 304, AISI 321, 12Kh18N10T, and 08Kh18N10, to determine the role of chromium and other components of these steels in their pitting resistance and to build mathematical models based on linear quadratic regressions and on a two-layer neural network of direct signal propagation. We have not find similar approaches because this is a pioneer work. The article summarizes experimental data of corrosion studies that we have conducted in the laboratory of the Azov Machine-Building Plant in Berdyansk city of Zaporozhye region for seven years.

2 Materials and research methods

Five industrial smeltings 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, 2), and structural heterogeneity was determined earlier in [15].

Table 1 – Chemical composition of steels AISI 304 and 08Kh18N10 [15]

№ of smelting	*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 [15]

№ of smelting	*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

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

Mathematical models of the dependence of the critical pitting temperature (CPT) of steels depending on their chemical composition (Tables 1, 2), structural heterogeneity and parameters of model circulating waters (pH 4…8, chloride concentration C_{Cl} = 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 [18]. Linear quadratic equations were constructed using these parameters (1). We have used standard feed-forward neural network, widely describeld in literature [19]:

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

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 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 chemical composition (x_3 – chromium), mass% [18].

A neural network model based on a two-layer feed-forward neural network for a reduced number of input features (x_1, x_2 and x_3) is described by formula (2) [20]. To simulate and train the neural network we have used online scripts of MATLAB [17]. The training have been provided using Levenberg-Marquardt algorithm.We have used 60% of the data as training and 40% as validation. To assess the suitability of the model, it is advisable to test it on data that was not used for training. That is why 60% (n=1200) of sample were used . This value is given as a guideline.

$$y = w_0^{(2,1)} + \sum_{i=1}^{1200} w_i^{(2,1)} \psi^{(1,i)} \times$$

$$\times \left(w_0^{(1,i)} + \sum_{j=1}^{800} w_j^{(1,i)} x_j^{(1,j)} \right), \qquad (2)$$

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:

$$E = \sum_{s=1}^{S} (y^s - y^{s*})^2 , \qquad (3)$$

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 [18].

The S is a number of instances (observations) in a sample. We use the all data for a model building. We have used online scripts of MATLAB for all model building. This software use 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 consistent with known literature data [21–26]. At the same time, it should be noted that for the square of the constituent x_2^2 (Table 3, item 4), taking into account its weight coefficient. w_k = -1,67·10^{-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. 3). 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 [27-30].

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_1^2	-0,9845
4	x_2^2	-1,67×10^{-5}
5		0,1735

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 content in them Cr within the standard. At the same time, the results of the analysis (Table 3, № 5) of the mathematical model (1) are harmonized with the data of works [16, 31-35] CPT 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 [36–40]. In addition, this element affects the solid-phase diffusion of Cr atoms to the surface of metastable pits and promotes their repassivation [33-35]. There is an evidence that Cr [41] increase the solubility of nitrogen in corrosion-resistant steels, and, consequently, their pitting resistance.

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)) and their Cr content. Other chemical elements in the studied steels (Tables 1, 2), the volume of titanium oxides and nitrides do not affect their y(CPT) and, accordingly, pitting resistance.

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 0,0382 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 smeltings 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, and x_3) (2) makes it possible to obtain much more accurate calculated values of y(CPT) for the steels under study, depending on the parameters of circulating water (x_1, x_2) and chemical elements (x_3) than the mathematical model (1). Since the total squares error for model (2) is 1.7994(3). 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).

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*
j	**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
	12 (x_3)	-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

It should be noted that in the mathematical model (1) based on linear squares regressions, the following xi variables are significant: (x1, x2 are the pH of the model circulating waters and the concentration of chlorides in them) and x3 is the chromium content in steels. And in a neural network model based on a neural network of direct signal propagation for a reduced numbers of features (2): (x1, x2, and x3). 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.

In order to study the influence of chromium on the critical pitting temperature (CPT) of steels, five industrial melts of AISI 304 and AISI 321 steel alloys and one melt each of 08X18N10 and 12X18N10T steels, which are analogues of the above-mentioned steels, were studied. Samples with a diameter of 42 mm and a thickness of 1 mm were polished, and their opposite surface and edge were shielded with fluoroplastic. Taking into account the possibility of contamination of the surface of the heat transfer plates of heat exchangers with sediment from water, solutions that are most often encountered during the operation of heat exchangers were chosen (according to the statistics of OJSC "Pavlogradhimmash" plant). The samples were subjected to pitting corrosion in aqueous solutions of magnesium chloride $MgCl_2$ with a chloride concentration of 350; 400; 500; 550 and 600 mg/l for 5 hours at a given temperature (± 0.5)0C and pH 4...8. The temperature of the solutions was maintained in a TS-80M-2 thermostat. For each study, 800 samples of each smelting were taken. Arithmetic mean values of the obtained parameters were used in the calculations. Pittings were recorded visually in 100 fields of view of the MMP-2P microscope. The samples were thermally tested in chloride-containing solutions to detect "active" pitting, i.e. with a diameter of 7 µm or more. In their absence, the temperature of each subsequent series of tests was increased by 2 oC until their appearance [13]. Table 5 reflects an experimental dependence of critical pitting temperature (CPT) of AISI 304 and 08Kh18N10 steels on chlorine concentration in model circulating waters (x2) pH (x1) and the content of Cr in steel (x3).

Table 5 – Critical pitting temperature (CPT) of AISI 304 and 08Kh18N10 steels in chloride-containing solutions

pH	C_{Cl^-}, mg/l	Steel AISI 304, smelting №					Steel 08X18H10
		1	2	3	4	5	CPT, ^{0} C
		The mean content of Cr, mass%					
		16,9	17,1	17,4	17,5	17,7	
		CPT, ^{0} C	CPT^{0}C	CPT, ^{0} C	CPT, ^{0} C	CPT, ^{0} C	
1	2	3	4	5	6	7	8
8	600	45	47	46	43	44	45
4		46	47	48	43	46	46
5		50	52	52	45	47	46
6		54	53	56	46	51	48
7		55	58	57	52	53	49
8	550	47	51	49	45	46	47
4		47	51	53	49	48	48
5		51	53	55	49	50	49
6		53	57	58	54	54	50
7		58	59	60	56	57	52
8	500	51	52	53	47	46	50
4		53	56	54	50	42	52
5		53	56	57	54	55	55
6		58	60	62	56	54	57
7		60	62	61	57	59	58
8	400	58	58	59	49	55	52
4		59	61	60	52	56	55
5		60	62	63	55	59	58
6		62	65	65	56	59	60
7		68	68	69	65	63	60
8	350	61	60	61	54	55	57
4		61	62	62	56	56	60
5		66	67	67	58	63	62
6		67	69	69	60	65	63
7		69	70	70	62	67	63

The mean content of Cr:

$$\overline{x} = \frac{1}{n}\sum_{i=1}^{n} x_i, \text{ n=800}, \quad (4)$$

Experimental critical pitting temperature $\overline{CPT}$ was obtained by formula:

$$\overline{CPT} = \frac{1}{n}\sum_{i=1}^{n} y_i, \text{ n=800}, \quad (5)$$

The line graph (Fig. 1) illustrates the dependence of calculated steel's CPT in model chlorine-containing recycled waters from the chromium concentration in steel. In numerical experiments on the pitting processes in model chloride-containing media, the vast possibilities of [17] "MATLAB" methodology for mathematical modeling of chromium-containing steel corrosion were used. The predicted dependence is shown in Figure 1. An analysis of numerical experimental results showed that the CPT of steel AISI 304, 08X18H10, calculated by the mathematical model (1), taking into account the weighting factor of 5.46 for the components x3 (Cr), grows in a straight line with an increase in the chromium content from 17.44 steel 08X18H10 to 18 .3 mass%, smelting 3, AISI 304 steel. The slope of the line indicates that a change in chromium content in such an interval considerably affects the CPT of these steels in the studied circulating waters. At the same time, the analysis showed that the CPT of AISI 321 and 12Kh18N10T steel also significantly climbs by 8.3 °C with an increase in the chromium content in them from 16.46, melt 2 of AISI321 steel to 17.97 mass% of 12Kh18N10T steel the straight-line dependence indicates that the CPT of the studied AISI304, 08Kh18N10, AISI321, 12Kh18N10T steels and their pitting resistance in model circulating waters strongly depends on the chromium content in them.

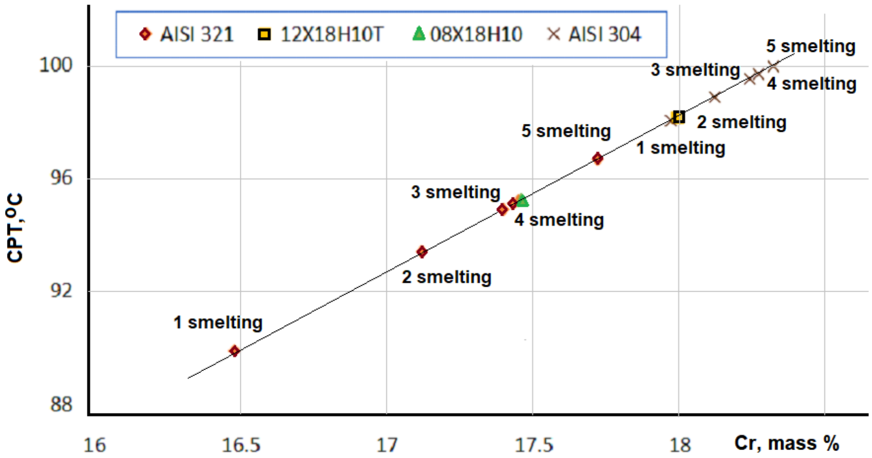

Figure 1 – Dependence of calculated steel's CPT in model chlorine-containing recycled waters on chromium l (x_3) concentration (pH=6, C_{Cl}=500 mg/l)

An analysis of the experimental results (Fig. 2) showed that the CPT of AISI304, 08Kh18N10, AISI321, and 12Kh18N10T sheets of steel increases linearly with increasing chromium concentration. In general, the pitting temperature is directly proportional to the chromium content. The deviation from the theoretical value is due to the attendance of other elements in the steels, which also affects the pitting process. As can be seen from the comparison of theoretical and experimental graphs, the proposed model is in good agreement.

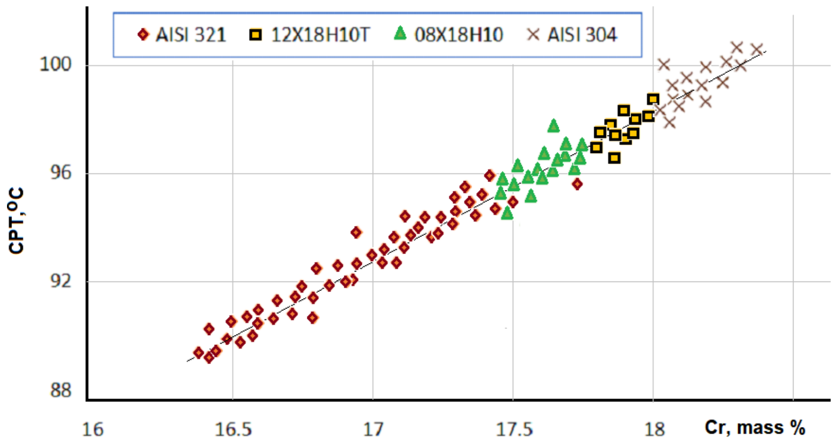

Figure 2 – Dependence of experimentally determined steel's CPT in model chlorine-containing recycled waters on chromium concentration (pH=6, CCl=500 mg/l)

In order to estimate the extent of data distribution (or spread) around calculated CPT the Dispersion (R^2) was determined as:

$$R^2 = \frac{1}{n}\sum_{i=1}^{n}(y_i - y_{i,calc})^2, \text{ n} = 800, \quad (6)$$

where yi is an experimental and yi,calc is a calculated data. The Root Mean Square Error (RMSE):

$$\text{RNSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(y_i - y_{i,calc})^2}. \quad (7)$$

The Mean Percentage Error (MPE) was calculated as:

$$\text{MPE} = \frac{1}{n}\sum_{i=1}^{n}\left(\frac{y_i - y_{i,calc}}{y_{i,calc}}\right)100\%, \quad (8)$$

Mean percentage error is 4,5-5,2%.

Thus, it is shown that using the methodology at the stage of the technical design of steel composition makes it possible to simulate the pitting formation dynamics and, due to this, reduces the time for choosing a steel grade, decline the number of bench tests required, and improve the quality and efficiency of structures in chlorine-containing recycled waters.

4 Conclusions

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

ii. It has been established that their pitting resistance increases with an increase in the pH of the circulating water, the Cr content and a decrease in the concentration of chlorides in the circulating water. 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.

iii. The 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 smeltings 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.

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