Physical Sciences and Technology. Vol. 12 (No. 3-4), 2025: 60-70

IRSTI 53.43.35 https://doi.org/10.26577/phst20251226

Prediction of the pitting corrosion rates in AISI 304 steel heat exchangers in industrial circulating waters

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(Received October 7, 2025; received in revised form November 27, 2025; accepted December 11, 2025)

A predictive methodology was developed to estimate the pitting growth rate on AISI 304 steel surfaces operating in industrial circulating waters. The approach is based on the observation that most pits formed near oxide inclusions are metastable and repassivate within minutes; therefore, corrosion losses of Cr, Ni, and Fe are attributed primarily to stable pits. Stable pits were identified using selective dissolution coefficients for chromium (ZCr) and nickel (ZNi). Second-order regression models were established to correlate corrosion losses (ΔCr, ΔNi, ΔFe) with steel composition, structural features, and water parameters such as chloride concentration and pH. Results show that ΔCr is mainly influenced by chloride content and microstructural factors, including the number of oxides (1.98–3.95 μm), average austenite grain size, and δ-ferrite volume. Factors reducing ΔFe losses follow the order: chloride concentration < Ni content < acidity < grain size < number of 1.98–3.95 μm oxides; whereas smaller oxides, inter-oxide spacing, and δ-ferrite promote higher losses. ΔNi primarily depends on structural heterogeneity rather than chemical composition or water parameters. The resulting model enables prediction of average pitting growth rates on AISI 304 steel in circulating waters with an accuracy of ±19%, providing a practical tool for assessing corrosion resistance in heat exchanger applications.

Keywords: pittings growth rate, model circulating waters, corrosion losses of metals in pittings.
PACS number(s): 81.65.Kn; 82.45.Bb.

1. Introduction

Nowadays, heat exchange equipment is widely used in various technological processes for the production of industrial products [1]. Therefore, many designs of heat exchangers have been developed by engineers and researchers, with different materials selected depending on the corrosive aggressiveness of the media. In practice, circulating waters containing chlorides and other anions are often used for cooling technological products. The heat exchange components of such equipment are made from stainless austenitic steels of the 18-10 type [2–4] and the alloy 06KhN28MDT (analogue to AISI 904L steel) [5, 6]. These materials are resistant to general corrosion in many media but can be susceptible to pitting corrosion in chloride-containing media, particularly in industrial circulating waters [7, 8]. It is generally known [9, 11] that passivated steels and alloys often undergo pitting in the presence of chlorides in the media. Therefore, the assessment and prediction of their resistance to pitting corrosion during equipment operation is a relevant issue. In low-mineralized solutions, such as industrial circulating waters, the resistance of stainless steels and alloys to pitting corrosion is evaluated based on their critical pitting temperatures [2–4, 12, 13]. This parameter characterizes the resistance of passive films on the surface of steels and alloys to pitting corrosion at a given chloride concentration and medium acidity. It is very convenient for practical use during the operation of heat exchangers, since by adjusting the flow rate of circulating water, it is possible to influence this parameter, provided that the chloride concentration increases or the acidity of the circulating water changes. Such an approach makes it possible to counteract pitting corrosion of steels during the operation of heat exchangers in recirculating water systems in the event of parameter changes. However, this may contribute to a reduction in production efficiency, since their operating modes are altered. Therefore, during the operation of heat exchangers in industrial process lines at enterprises, even in the event of pitting corrosion caused by changes in recirculating water parameters, it is not possible to immediately change their operational regimes. Thus, a question arises in industrial practice: "How much time remains until perforation of heat exchange elements (plate, tube) of specified thickness (from 0.2 to 2.0 mm)?". To answer this question, a methodology has been developed for calculating the pitting growth rate on the surface of AISI 304 steel in model circulating waters. This methodology is based on previously established patterns and mechanisms of pitting corrosion of austenitic stainless steels and chromium-nickel-molybdenum alloys in chloride-containing media [2–8, 14], as well as on widely known information about the theory of localized corrosion [15–20].

2. Materials and methods

To develop a methodology for calculating the pitting corrosion growth rate on the surface of AISI 304 stainless steel exposed to model circulating waters with pH values ranging from 4 to 8 and chloride concentrations between 300 and 600 mg/L, mathematical models were constructed based on experimental data on corrosion losses of chromium (ΔCr), nickel (ΔNi), and iron (ΔFe) associated with pitting corrosion [21,22].

The developed mathematical models were based on second-order regression dependencies. In particular, the mathematical model (1) can be written in the form:

$$Y(\Delta Cr; \Delta Ni; \Delta Fe) = M_0 + \sum_{j=1}^{N} M_j \times x_j + \sum_{j=1}^{N} M_{(N+j)} \times x_j^2, \quad (1)$$

where: Y is the corrosion loss ΔCr, ΔNi or ΔFe of AISI 304 steel in pittings, mg for 240 hours of testing in model recycled water with pH 4-8 (x_1) and chloride concentration (x_2) of 300 and 600 mg/l [21, 22]. Here M is a mass coefficient of components x_j, and x_j is variables, in particular: x_3 – V_{ox}, vol. %; x_4 – the number of oxides up to 1.98 μm in size at 100 fields of view of an optical microscope (×320) in steel, pcs; x_5 – the number of oxides from 1.98 up to 3.95 μm in size, pcs; x_6 – L_{ox}, μm (mean distance between oxides in steel); x_7 – d_g, μm (mean grain diameter of austenite in steel); x_8 – volume of δ-ferrite vol. %; x_9 – C, wt. % (carbon content in steel); x_{10} – Mn; x_{11} – Si; x_{12} – Cr; x_{13} – Ni; x_{14} – N; x_{15} – Ti; x_{16} – S; x_{17} – P; x_{18} – χ_0, m^3/kg (specific magnetic susceptibility of steel). The chemical composition and structural constituents of the investigated AISI 304 steel heats were determined previously [23]. The specific paramagnetic susceptibility of austenite and the volume fraction of δ-ferrite were established in [24].

To build the second-order regressions (1), we used the analysis of the influence of individual factors X = {x_1, x_2, ..., x_{18}} on the resultant (output – Y = {y_1, y_2, ..., y_7} indicators [25, 26]. The dependent variables are: Y_1 = ΔCr, mg (10^{-5}) – chromium loss in pittings, Y_2 = ΔFe, mg (10^{-5}) – iron loss in pittings, Y_3 = ΔNi, mg (10^{-5}) – nickel loss in pittings,

$$Y_4 = \frac{Y_1}{Y_2} = Z_{Cr}, \quad (2)$$

where: Y_4 is the coefficient of selective dissolution of Cr in pittings,

$$Y_5 = (Y_4 \geq 1) = \left\{ \frac{1}{Y_4} \geq 1 \right\}, \quad (3)$$

- if $Z_{Cr} < 1$, then steel in a solution with such parameters (variables (x_1 (pH);) x_2 (C_{Cl-}, mg/l)) undergoes pitting corrosion with the formation of stable pittings, otherwise, steel in a solution with such parameters (variables x_1, x_2) pits with the formation of metastable pittings [22].

The coefficient of selective nickel dissolution from pittings, Z_{Ni}, is determined according to equation (4):

$$Y_6 = \frac{Y_3}{Y_2} = Z_{Ni}, \quad (4)$$

$$Y_7 = (Y_6 \geq 1) = \left\{ \frac{1}{Y_6} \geq 1 \right\}. \quad (5)$$

If $Z_{Ni} < 1$, then the pittings are growing intensively, otherwise, the pittings are not growing intensively [22].

The developed mathematical models based on the second-order regressions Y = {y_1, y_2,…, y_7} were used to determine the corrosion losses ΔCr, ΔN, and and ΔFe in pittings, identify metastable and stable pittings, and the intensity of their growth. At the same time, Y_1 – Y_5 were used to determine the rate of growth of stable pittings on the surface of AISI 304 steel in model recycled water with pH 4-8 and chloride concentration from 300 up to 600 mg/l, based on established approaches and analytical dependencies for determining the coefficients of participationof inclusions in pitting of steel [27].

The main approaches to calculating the average growth rate of pittings on the surface of AISI 304 steel are based on the fact that pittings nucleate and develop in the vicinity of inclusions [8, 28–30]. They are predominantly metastable and, therefore, repassivate within several minutes [31]. Consequently, it was assumed that all corrosion losses, ΔCr, ΔNi, and ΔFe, are concentrated only in stable pittings. These were identified using the coefficients of selective chromium dissolution from pittings (Z_{Cr}) [22]. It was also assumed that stable pittings form in the vicinity of inclusions located at the intersections with austenite grain boundaries in AISI 304 steel.

The quality of the developed methodology for calculating the growth rate of stable pittings on the surface of AISI 304 steel during pitting in model circulating waters with pH values from 4 up to 8 and chloride concentrations ranging from 300 up to 600 mg/l was evaluated using equation (6), based on the sum of squared instantaneous errors [32].

$$E = \sum_{S=1}^{S} \left(y^{S} - y^{S*} \right)^{2}, \tag{6}$$

where: y^S is the actual depth of stable pittings measured with an MMP-2R optical microscope, µm; y^{S*} is the calculated depth of stable pittings on the surface of AISI 304 steel, µm.

The quality of each developed mathematical model (1–5) was evaluated based on the sum of squared errors (SSE) and the mean squared error (MSE) [32].

3. Results and discussion

To calculate the average growth rate of pittings on the surface of AISI 304 steel, it is proposed to use the calculated values of ΔCr, ΔNi, and ΔFe from pittings (1–5), depending on the parameters of the model circulating waters within the pH range of 4 up to 8 and chloride concentrations from 300 up to 600 mg/l.

In particular, for the mathematical model Y_1 (ΔCr) (1), based on data [21-24, 27, 33, 34], the regression coefficients were determined (Table 1).

Table 1 – Regression coefficients for Y_1.

M_2 (x_1)	$-499081 \cdot 10^{-6}$
M_9 (x_8)	$-1454{,}943182 \cdot 10^{-6}$
M_{20} (x^2)	$36285 \cdot 10^{-6}$
M_{21} (x_2^2)	$-1{,}52682 \cdot 10^{-6}$
M_{23} (x_4^2)	$-3{,}26616 \cdot 10^{-6}$
M_{24} (x_5^2)	$2771 \cdot 10^{-6}$
M_{25} (x_6^2)	$-1904 \cdot 10^{-6}$
M_{26} (x_7^2)	$12033 \cdot 10^{-6}$
M_{32} (x_{13}^2)	$329880 \cdot 10^{-6}$

According to the data (Table 1), it was established that chromium corrosion losses (ΔCr) from pitting of AISI 304 steel decrease with increasing alkalinity of model recirculating waters, δ-ferrite volume fraction, and average spacing between oxide inclusions. In particular, it was found that the ΔCr parameter of AISI 304 steel from pitting decreases by 20×10^{-6} mg with an increase in pH of model recirculating waters from 4 up to 8. It should be noted that the effect of δ-ferrite volume fraction in the investigated steel on its ΔCr from pitting was considerably less pronounced, as these values decreased by 0.00083×10^{-6} mg with an increase in its volume from 50×10^{-3} up to 168×10^{-3} vol. %. However, it can be stated that the size of oxide inclusions in AISI 304 steel has the greatest effect on its ΔCr from pitting. Indeed, these increase by 487.3×10^{-6} mg with an increase in average spacing between oxide inclusions from 150 up to 173 µm. From the analysis of regression coefficients M_{21} (x_2^2) and M_{23} (x_4^2), it follows that ΔCr of AISI 304 steel from pitting weakly depends on chloride concentration (x_2) in model recirculating waters and the quantity of fine oxides (x_4) therein. It was revealed that ΔCr of steel from pitting increases by 9797×10^{-5} mg with an increase in chloride concentration in model recirculating waters within the investigated range. In this case, the indicators of structural heterogeneity of AISI 304 steel have an effect that is more than an order of magnitude smaller on this parameter. Indeed, it was determined that ΔCr of AISI 304 steel from pitting increases by only 160×10^{-5} mg with increasing quantity of oxides sized 1.98 … 3.95 μm within the investigated range and by 89×10^{-5} mg with an increase in average austenite grain diameter from 49 up to 86 μm. At the same time, it has been proven that the chemical composition of the investigated steel practically does not affect chromium corrosion losses in pitting within the investigated model recirculating waters.

Summarizing the foregoing, it can be noted that ΔCr of AISI 304 steel from pitting depends mainly on the media parameter (x_2) and its structural heterogeneity. Moreover, parameter (x_2) has 32.9 times stronger effect on ΔCr from pitting than x_5, x_7, x_8 taken together.

For the mathematical model Y_2 (ΔFe), based on data [21-24, 27, 33, 34], its coefficients were established (Table 2). From the analysis of data (Table 2), it can be noted that Y_2 decreases with increasing pH (x_1) of model recirculating waters, chloride concentration therein (x_2), quantity of oxides in steel sized from 1.98 up to 3.95 μm (x_5), mean austenite grain diameter (x_7), and nickel content therein.

Table 2 – Coefficients of mathematical model Y_2.

M_2 (x_1)	$-4765640\cdot10^{-6}$
M_9 (x_8)	$1536023300\cdot10^{-6}$
M_{20} (x_1^2)	$38068\cdot10^{-6}$
M_{21} (x_2^2)	$-2,37036\cdot10^{-6}$
M_{23} (x_4^2)	$83,3358\cdot10^{-6}$
M_{24} (x_5^2)	$-2800\cdot10^{-6}$
M_{25} (x_6^2)	$2420\cdot10^{-6}$
M_{26} (x_7^2)	$-12540\cdot10^{-6}$
M_{32} (x_{13}^2)	$-443110\cdot10^{-6}$

According to the analysis of Eq. (1) and data (Table 2), it has been established that ΔFe losses from pitting in steel decrease by 19.063×10^{-5} mg with an increase in pH (x_1) of the media from 4 up to 8, by 0.6367×10^{-5} mg with an increase in chloride concentration (x_2) within the range of 300–600 mg/l, by 155.4×10^{-5} mg with an increase in the quantity of oxides of the aforementioned size (x_5) in the steel, by 62.7×10^{-5} mg with an increase in the mean austenite grain diameter (x_7) from 49 up to 86 μm, and by 9.7×10^{-5} mg with an increase in nickel content (x_{13}) in the steel from 8.1 up to 9.3 wt. %. It should be noted that ΔFe of steel from pitting increases with increasing pH (x_1), volume of δ-ferrite inclusions (x_8), quantity of oxides sized up to 1.98 μm (x_4), and mean distance between oxides (x_6) (Table 2). Calculation of the effect of $X = \{x_1, x_4, x_6, x_8\}$ on ΔFe showed that ΔFe of AISI 304 steel from pitting increases by 18.2726×10^{-5} mg with increasing pH (x_1) of the media from 4 up to 8, by 7.5523×10^{-5} mg with increasing quantity of fine oxides (x_4) from 300 up to 425, by 18×10^{-5} mg with increasing mean distance between oxides (x_6) from 150 up to 173 μm, and by 152.4×10^{-5} mg with increasing volume of δ-ferrite inclusions (x_8) from 0.014 up to 0.113 vol. %.

Summarizing the effect of parameters $X = \{x_1, x_2, x_4, x_5, x_6, x_7, x_8, x_{13}\}$ on ΔFe of AISI 304 steel from pitting, it should be noted that the significance of factors causing a decrease in Y_2 increases in the following sequence: x_2, x_{13}, x_1, x_7, x_5, while those causing an increase in Y_2 are: x_4, x_6, x_1, x_8. Thus, it can be noted that the parameters of model recirculating waters – pH (x_1) and chloride content therein (x_2) – have weak effect on ΔFe of AISI 304 steel from pitting. However, the structural constituents of AISI 304 steel primarily determine corrosion losses ΔFe from pitting. These decrease with increasing quantity of oxides sized 1.98 up to 3.95 μm, mean austenite grain diameter, and increase if the mean distance between oxide inclusions and volume of δ-ferrite inclusions in steel increase. A hypothesis has been proposed that stable pittings nucleate and grow in the vicinity of large oxides at their intersection with austenite grain boundaries. Indeed [22], it has been established that the greater the quantity of large-sized oxides in whose vicinity pittings nucleate and grow, the lower the current density of anodic currents therein due to their redistribution among pittings and the lower the intensity of ΔFe dissolution. Obviously, the greater the mean distance between oxides, the larger they are. According to the data presented in reference [22], in the vicinity of larger inclusions, there are more structural defects in the steel, which accelerates selective dissolution of ΔFe.

It should be noted that ΔCr from pitting of AISI 304 steel depends more on the parameters of model recirculating waters (x_1, x_2) than on the constituents of its structure $X = \{x_3 \ldots x_8\}$, whereas ΔFe correlates predominantly with the elements of its structure $X = \{x_5, x_6, x_7, x_8\}$. Most likely, this is related to the more negative value of the standard electrode potential of Cr compared to Fe [31].

For mathematical model Y_3 (1), using data $Y = Y_3$ [24] and $X = \{x_1 \ldots x_{18}\}$ [26], regression coefficients for Y_3 were established (Table 3).

Table 3 – Coefficients of mathematical model Y_3.

M_2 (x_1)	$-3977620 \cdot 10^{-6}$
M_9 (x_8)	$-2367054950 \cdot 10^{-6}$
M_{20} (x_{12}^2)	$278768 \cdot 10^{-6}$
M_{21} (x_2^2)	$1{,}94385 \cdot 10^{-6}$
M_{23} (x_4^2)	$3{,}74289 \cdot 10^{-6}$
M_{24} (x_5^2)	$-3397620 \cdot 10^{-6}$
M_{25} (x_6^2)	$2980 \cdot 10^{-6}$
M_{27} (x_7^2)	$278770 \cdot 10^{-6}$
M_{32} (x_{13}^2)	$1{,}94385 \cdot 10^{-6}$

Analysis of Table 3 data demonstrates that Y_3 (ΔNi) decreases with increasing pH (x_1) of model recirculating waters, volume of δ-ferrite inclusions in AISI 304 steel (x_8), quantity of oxide inclusions sized 1.98–3.95 µm (x_5), and with decreasing chloride content in model recirculating waters (x_2), number of oxides sized up to 1.98 µm (x_4), mean distance between oxides (x_6), mean diameter of austenite grains (x_7), and Ni concentration (x_{13}). Assessment of the significance of parameters $X = \{x_1, x_2, x_4, x_5, x_6, x_7, x_{13}\}$ on Y_3 was performed using mathematical model (1), with utilization of data from Table 3 and results from references [22, 24]. It was established that Y_3 decreases by 13.59×10^{-5} mg upon increasing pH (x_1) of recirculating waters from 4 up to 8, by 26.01×10^{-5} mg upon increasing volume of δ-ferrite inclusions in steel from 0.050 up to 0.168 vol. %, by 1.88513×10^{-3} mg upon increasing the number of inclusions sized 1.98–3.95 µm from 46 up to 240 per 100 microscope fields of view (×320). At the same time, it was calculated that Y_3 increases by 8.92×10^{-5} mg upon increasing pH (x_1) from 4 up to 8, by 0.35×10^{-5} mg upon increasing chloride concentration (x_2) from 300 up to 600 mg/l, by 0.2×10^{-5} mg upon increasing the number of fine oxides sized up to 1.98 µm (x_4) from 300 up to 425, by 22.14×10^{-5} mg upon increasing mean distance between oxides (x_6) from 150 up to 173 µm, by 1.3924×10^{-3} mg upon increasing mean austenite grain diameter (x_7) from 49 up to 86 µm, and by 47.43×10^{-11} mg upon increasing Ni content in steel (x_{13}) from 8.09 to 9.34 wt. %.

Based on the analysis performed, the significance of parameters $X = \{x_1, x_5, x_8\}$ that contribute to the decrease in Y_3 upon their increase is arranged in the following order: x_1, x_8, x_5, whereas parameters that contribute to the increase in Y_3, $X = \{x_1, x_2, x_4, x_6, x_7, x_{13}\}$, are: x_{13}, x_4, x_7. This demonstrates that Y_3 in AISI 304 steel decreases upon increasing the number of large oxide inclusions sized 1.98–3.95 µm, in the vicinity of which stable pittings nucleate at their intersection with austenite grain boundaries, upon increasing volume of δ-ferrite inclusions, and upon decreasing the mean austenite grain diameter.

Summarizing the foregoing, it can be stated that corrosion losses ΔNi from pitting are determined predominantly by the structural heterogeneity of the steel and are practically independent of changes in its chemical composition within standard ranges and parameters of model recirculating waters that promote pittings formation.

To identify metastable and stable pittings on the surface of the investigated AISI 304 steel in model recirculating waters, mathematical model (2) was developed, whose regression coefficients are presented in Table 4.

Analysis of Table 4 data demonstrates that the selective dissolution coefficient of Cr from pitting decreases with increasing chloride concentration (x_2), quantity of oxide inclusions sized up to 1.98 µm (x_4), mean distance between them (x_6), volume of δ-ferrite inclusions in AISI 304 steel (x_7), and with decreasing quantity of oxides sized 1.98–3.95 µm (x_5) and Ni concentration therein. At the same time, it should be emphasized that the regression coefficients M_2 (x_1) and M_{20} of model (2) are 7.107 and –11.8736, respectively. Thus, the coefficient Z_{Cr} decreases with increasing pH (x_1) of model recirculating waters from 4 up to 8 by only 4.77.

Table 4 – Coefficients of mathematical model $Y_4 = Z_{Cr}$.

M_2 (x_1)	$7276907 \cdot 10^{-6}$
M_9 (x_8)	$-1020086466 \cdot 10^{-6}$
M_{20} (x_1^2)	$-593681 \cdot 10^{-6}$
M_{21} (x_2^2)	$-4{,}02701 \cdot 10^{-6}$
M_{23} (x_4^2)	$-6{,}66467 \cdot 10^{-6}$
M_{24} (x_5^2)	$1834 \cdot 10^{-6}$
M_{25} (x_6^2)	$-1790 \cdot 10^{-6}$
M_{26} (x_7^2)	$-8099 \cdot 10^{-6}$
M_{32} (x_{13}^2)	$280000 \cdot 10^{-6}$

Thus, pH (x_1) of the media has practically no effect on the coefficient Z_{Cr}. At the same time, based on the results of analysis of the effects of parameters $X = \{x_1, x_2, x_4, x_5, x_6, x_7, x_8, x_{13}\}$ on Y_4, it was established that these decrease by 101; 19; 1.09; 6.04; 13.3; 40.45 upon increasing volume of δ-ferrite inclusions in steel (x_8), chloride concentration in model recirculating waters (x_2), quantity of oxide inclusions sized up to 1.98 μm (x_4), mean distance between oxides (x_6), and mean diameter of austenite grains (x_7) within the specified ranges indicated above. At the same time, Y_4 increases by 101.76 and 6.08 upon increasing in the steel the quantity of oxides sized 1.98–3.95 μm (x_5), and by 6.08 upon increasing the Ni content therein within the aforementioned ranges.

Based on the analysis performed, it can be stated that the coefficient Z_{Cr} decreases with increasing volume of δ-ferrite inclusions (x_8) in steel, mean diameter of austenite grains (x_7), and with decreasing quantity of oxide inclusions sized 1.98–3.95 μm, in the vicinity of which pittings nucleate at their intersection with austenite grain boundaries. Thus, the results of previous investigations and the hypothesis regarding the effect of volume of δ-ferrite inclusions in steel on selective dissolution of metals from stable pittings are confirmed. In this connection, it is important to note that the specific magnetic susceptibility of austenite (χ_{18}) has no effect on the process of selective metal dissolution in pittings.

To identify metastable and stable pittings on the surface of AISI 304 steel, using computer processing $Y_5 = (Y_4 \geq 1) = \{1/Y_4 \geq 1\}$ as a function of variables $X = \{x_1, x_2, ..., x_{18}\}$, a mathematical model was constructed that is based on formula (3) and data from references [21, 27, 35]. The calculated coefficients of this mathematical model are presented in Table 5.

The proposed mathematical model (3) (Table 5) provides the ability to very rapidly calculate Z_{Cr} and assess the nature of pittings on the surface of AISI 304 steel as a function of recirculating water parameters and steel properties.

Table 5 – The coefficients of mathematical model $Y_5 = (Y_4 \geq 1) = \{1/Z_{Cr} \geq 1\}$.

M_2 (x_1)	$1966477 \cdot 10^{-6}$
M_9 (x_8)	$293560606 \cdot 10^{-6}$
M_{20} (x_2^1)	$-159659 \cdot 10^{-6}$
M_{21} (x_2^2)	$-1{,}18519 \cdot 10^{-6}$
M_{23} (x_4^2)	$-93{,}15195 \cdot 10^{-6}$
M_{24} (x_5^2)	$-581 \cdot 10^{-6}$
M_{25} (x_6^2)	$269 \cdot 10^{-6}$
M_{26} (x_7^2)	$-2465 \cdot 10^{-6}$
M_{32} (x_{13}^2)	$-63424 \cdot 10^{-6}$

To determine the coefficient Z_{Ni} for selective dissolution of nickel from pitting, a mathematical model was constructed using formula (4), and the established coefficients of this model are presented in Table 6.

Table 6 – The coefficients of mathematical model $Y_6 = \frac{Y_3}{Y_2} = Z_{Ni}$.

M_2 (x_1)	$16792025 \cdot 10^{-6}$
M_9 (x_8)	$-14718238697 \cdot 10^{-6}$
M_{20} (x_2^1)	$-1374383 \cdot 10^{-6}$
M_{21} (x_2^2)	$-16{,}2513 \cdot 10^{-6}$
M_{23} (x_4^2)	$-246 \cdot 10^{-6}$
M_{24} (x_5^2)	$27576 \cdot 10^{-6}$
M_{25} (x_6^2)	$-20603 \cdot 10^{-6}$
M_{26} (x_7^2)	$120672 \cdot 10^{-6}$
M_{32} (x_{13}^2)	$3481913 \cdot 10^{-6}$

According to the data in Table 6, Y_6 (Z_{Ni}) decreases by 1471.82; 4.39; 22.29; 156.06 with increasing parameters $X = \{x_3, x_2, x_4, x_6\}$ within the ranges specified above. At the same time, Y_6 (Z_{Ni}) increases by 1530.02; 602.76; 8983 with increasing parameters $X = \{x_5, x_7, x_{13}\}$ within the ranges specified above.

Summarizing the foregoing, it can be noted that the coefficients Z_{Ni} depend predominantly on variables $x_8, x_5, x_6, x_7, x_{13}$. Moreover, their influence increases in the following sequence: x_{13}, x_6, x_7, x_8, x_5. At the same time, the effect of x_7, x_8, x_5 is most substantial; therefore, volume of δ-ferrite inclusions, mean austenite grain diameter, and quantity of large oxides have the greatest effect on the process of selective nickel dissolution from pitting.

A mathematical model (5) was constructed that enables assessment of the intensity of pittings propagation on the surface of AISI 304 steel in model recirculating waters. The coefficients of mathematical model (5) are presented in Table 7.

Summarizing the foregoing, it can be noted that by applying information technologies and mathematical model (5), it is possible to rapidly assess the intensity of pitting propagation on the surface of AISI 304 steel. At the same time, analysis of model (5) showed that the Z_{Ni} coefficients of AISI 304 steel decrease with increasing volume of δ-ferrite inclusions (x_8) and with decreasing mean austenite grain diameter (x_7) and quantity of oxides (1.98–3.95 μm) (x_5). The influence of parameters

$X = \{x_5, x_7, x_8\}$ on Z_{Ni} was found to be most significant, while the influence of parameters $X = \{x_1, x_2, x_4, x_6, x_{13}\}$ was found to be insignificant.

Table 7 – Coefficients of the mathematical model (5).

M_2 (x_1)	$1525000 \cdot 10^{-6}$
M_9 (x_8)	$-416666667 \cdot 10^{-6}$
M_{20} (x_1^2)	$-125000 \cdot 10^{-6}$
M_{21} (x^2_2)	$-5{,}92596 \cdot 10^{-7}$
M_{23} (x^2_4)	$-1{,}85183 \cdot 10^{-5}$
M_{24} (x^2_5)	$765 \cdot 10^{-6}$
M_{25} (x^2_6)	$-659 \cdot 10^{-6}$
M_{26} (x^2_7)	$3418 \cdot 10^{-6}$
M_{32} (x^2_{13})	$116313 \cdot 10^{-6}$

Analysis of the indicated models demonstrates that ΔCr from pitting depends primarily on chloride concentration in model recirculating waters and such steel parameters as oxide quantity, mean austenite grain diameter, and δ-ferrite content. A hypothesis has been proposed that pitting nucleation and propagation are caused by the intensity of chloride ion adsorption on structural imperfections of AISI 304 steel in the vicinity of these oxides at their intersection with austenite grain boundaries. It was established that ΔFe from pitting decreases with increasing oxide quantity in the steel (sized 1.98…3.95 μm), mean austenite grain diameter, and increases when δ-ferrite quantity and mean distance between steel oxides increase. It has been proven that ΔNi from pitting decreases with increasing oxide quantity in the steel (1.98…3.95 μm) and δ-ferrite content, and with decreasing mean austenite grain diameter. It was established that coefficients Z_{Cr} decrease with increasing δ-ferrite quantity in the steel, mean austenite grain diameter, and with decreasing oxide quantity (sized 1.98…3.95 μm). This may also contribute to the transition of metastable pittings to stable pittings. It was found that coefficients ΔNi decrease with increasing oxide and δ-ferrite quantity, and mean austenite grain diameter of the steel. At the same time, it was established that media pH, steel chemical composition within standard ranges, fine oxides, their volume, and specific paramagnetic susceptibility do not affect selective dissolution of ΔCr, ΔFe, and ΔNi from pitting on AISI 304 steel. The proposed models provide the ability to assess the pitting character of AISI 304 steel, calculate ΔCr, ΔFe, ΔNi from pitting and their propagation rates, which is important in heat exchanger operation.

Taking into account the approaches mentioned above and the constructed mathematical models (1–5), the determination of the average pittings growth rate on the surface of AISI 304 steel has been justified using formula (7):

$$v_p = \frac{h}{\tau}, \tag{7}$$

where: v_p is mean pittings growth rate, μm/h; h is mean pitting depth, μm; τ is a test time, hours (is a constant value equal to 240 hours).

The average pitting depth h is calculated using the next equation:

$$\bar{h} = R \big/ N_{s.p.} \,. \tag{8}$$

where: R is the radius of the conditional layer of metal dissolved from the steel during the test, μm; $N_{s.p.}$ is the number of stable pittings on the steel surface.

The parameter R is proposed to be determined by relation:

$$R = \sqrt[3]{\frac{3V}{4\pi}}, \tag{9}$$

where V is a volume of metal dissolved in pittings, μm^3.

The volume of metal dissolved in pittings is determined by:

$$V = V_{Fe} + V_{Cr} + V_{Ni}, \tag{10}$$

where $V_{(Fe,\,Cr,\,Ni)}$ is a volume of Fe, Cr, Ni dissolved on the pittings surface of, μm^3.

The volume of metals dissolved from the pitting surface is determined by equation (11):

$$V_{(Cr,Ni,Fe)} = m_{(Fe,Ni,Fe)} \big/ \rho_{(Cr,Ni,Fe)}, \tag{11}$$

where $m_{(Cr,\,Ni,\,Fe)}$ is the mass of Cr, Ni, Fe dissolved on the surface of the pittings, g; $\rho_{(Fe,\,Cr,\,Ni)}$ is the density of Fe, Cr, Ni, $g/\mu m^3$.

According to [36], with densities $\rho_{Fe} = 7.874 \cdot 10^{-12}$ $g/\mu m^3$, $\rho_{Cr} = 7.19 \cdot 10^{-12}$ $g/\mu m^3$, $\rho_{Ni} = 8.9 \cdot 10^{-12}$ $g/\mu m^3$, the values $m_{(Cr,\,Fe,\,Ni)}$ can be identified with ΔCr, ΔFe, and ΔNi of steel from stable pittings, the calculation of which is performed using mathematical model (1).

As shown above, formation of stable pittings on the surface of the investigated AISI 304 steel occurs predominantly in the vicinity of the largest oxide inclusions sized up to 4 μm at their intersection points with austenite grain boundaries. Around smaller oxides (up to 1.98 μm), metastable pittings predominantly nucleate, characterized by rapid repassivation. In connection with this, the coefficients of oxide participation in the pitting-forming process of AISI 304 steel were determined exclusively for stable pittings with size exceeding 5 μm [37]. It was previously established [22] that the coefficients of oxide inclusion participation in pitting of AISI 304 steel do not depend on variations in its chemical composition within standard ranges, but are determined by pH and chloride concentration in chloride-containing recirculating waters. Thus, the role of oxides in pitting formation of AISI 304 steel correlates with the pitting potential. Based on this, to calculate the coefficients of oxide participation in pitting of AISI 304 steel in model recirculating waters with pH 4–8 and chloride concentration 300–600 mg/l, the following relationships (12–16) are proposed:

$$K = 0{,}341 - 3{,}7\cdot10^{-4}\, C_{Cl^-}, \text{ at pH } 8; \qquad (12)$$

$$K = 0{,}081 + 1{,}20\cdot10^{-4}\, C_{Cl^-}, \text{ at pH } 7; \qquad (13)$$

$$K = 0{,}082 + 1{,}42\cdot10^{-4}\, C_{Cl^-}, \text{ at pH } 6; \qquad (14)$$

$$K = 0{,}085 + 1{,}40\cdot10^{-4}\, C_{Cl^-}, \text{ at pH } 5; \qquad (15)$$

$$K = 0{,}048 + 2{,}10\cdot10^{-4}\, C_{Cl^-}, \text{ at pH } 4. \qquad (16)$$

Where C_{Cl^-} is a concentration of chloride ions in the model recirculating water.

Relationships (12–16) enable determination of the fraction of oxide inclusions in AISI 304 steel that participate in the pitting-forming process. Calculations are conducted on a defined metal area, namely on 100 fields of view of an optical microscope (×320). In this case, the total number of oxides on the surface of AISI 304 steel is determined by metallographic analysis method [38]. It should be noted that modern optical microscopes are equipped with software that allows more rapid and more accurate determination of the quantity and size of inclusions in steels and alloys.

In model recirculating waters with pH 4–8 and chloride concentration 300–600 mg/l, fine oxide inclusions (up to 1.98 μm) in AISI 304 steel participated in the pittings-forming process in the range of 8.53–11.68 % of their total number on the sample surfaces, whereas oxides sized 1.98–3.95 μm were involved in pitting in the amount of 3.1–5.76 % of their total quantity [39]. Simultaneously, pittings that form in the vicinity of fine oxides rapidly repassivate because their sizes do not exceed 2 μm [40-45]. As a result, the parameters ΔFe, ΔCr, ΔNi obtained from such pittings are not taken into account in the calculation of the volume (V) of metal dissolved from stable pittings (10).

It is important to note that on mean, 4.43 % of oxide inclusions sized 1.98–3.95 μm were involved in nucleation of stable pittings. Taking this into account, the number of stable pittings per area of 100 microscope fields of view (×320) is recommended to be calculated using formula (17):

$$N_{s,p} = 0{,}0443\times K\times N_{ox}, \qquad (17)$$

where K is the participation of oxides involved in pitting, determined by formulas (12-16); N_{ox} is the total amount of oxides on the surface of AISI 304 steel per 100 microscope fields of view (×320).

Based on the analysis performed, it can be stated that the application of formulas (1–17) enables calculation of the mean growth rate of stable pittings on the surface of AISI 304 steel in model recirculating waters with pH 4–8 and chloride content 300–600 mg/l. At the same time, it should be emphasized that the pitting propagation rates of stable pittings on the surface of AISI 304 steel in recirculating waters calculated in this manner are characterized by a mean error of up to ±19 %. This finds confirmation in the results of laboratory studies and practical experience in heat exchanger equipment operation. This is due to the stochastic nature of the processes of pitting nucleation, development, and repassivation. Despite this, the proposed methodology for calculating the pitting propagation rate on the surface of AISI 304 steel in model recirculating waters enables prediction of the time to achieve perforation of heat transfer elements of heat exchangers in the event of their pitting.

4. Conclusion

The main approaches for calculating the average growth rate of pittings on the surface of AISI 304 steel in model circulating waters with pH values ranging from 4 up to 8 and chloride concentrations between 300 and 600 mg/l have been developed.

These approaches are based on the assumption that pittings nucleating and growing in the vicinity of oxide inclusions are predominantly metastable and, therefore, repassivate within several minutes. Consequently, it was assumed that all corrosion losses, ΔCr, ΔNi, and ΔFe, are concentrated only in stable pittings. It was proposed to identify them using the coefficient of selective chromium dissolution (Z_{Cr}). Second-order regression-based mathematical models were developed to establish the relationships between the corrosion losses of Cr, Ni, and Fe from pittings, the coefficients Z_{Cr} and Z_{Ni}, and the steel's chemical composition, structural constituents, and circulating water parameters. A method for calculating the growth rate of pittings on the surface of AISI 304 steel during the operation of heat exchangers in circulating water systems was developed and implemented in production. This method is based on the calculated values of corrosion losses ΔCr, ΔNi, and ΔFe from pittings and on the participation coefficients of oxide inclusions in pitting corrosion, determined using empirical formulas. It was shown that the sum of squared instantaneous errors for the average pitting growth rate determined by calculation and measurement is E = 0.250718, and the mean error is ± 19 %.

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