1. Introduction

In the water recycling systems of barometric condensers of enterprises, which produce chlorine and sodium hydroxide by the electrolysis method, the basic mineralization of water is determined by the Cl\(^{-}\) ions. The chloride content is up to 80% of the total salt content of water in the circulating water of chlorine enterprises that operate employing a drainless circuit.

The main structural materials of the circulating water supply systems are carbon steel of St.3 grade and gray cast iron of Cl 18-36 grade. This is due to their low cost, which is combined with good technological properties. The elements of pipelines, heat exchangers, grids, cooling towers are often made of carbon steel and cast iron.

In terms of relation with ferrous metals, chloride ions are among the most aggressive. Aggressiveness of chloride ions is explained by the ability to adsorption. In this case, oxygen is displaced from the metal surface, which leads to disruption of the continuity of the protective film and increases the rate of its corrosion.
The volume of circulating water in the chemical industry, in particular chlorine production, consists of hundreds of thousands of cubic meters. The main discharge of circulating water occurs due to its natural evaporation and the need for periodic water escape due to the accumulation of admixtures above permissible limits. Admixtures cause increased mineralization of water, which leads to the acceleration of corrosion destruction of metallic materials of the cooling systems. Creation of closed cycles of recycling water supply, which are able to work with high-mineralized water, will make it possible to solve the following tasks:

1) to reduce the costs related to the processes of discharge and compensation of a large volume of water, which will enable reducing the ecological load on the environment;
2) to reduce the cost of repair and replacement of equipment due to corrosion destruction.

2. Literature review and problem statement

Scientific data on the corrosive aggressiveness of chloride ions in water solutions with respect to ferrous metals are very numerous [1–10]. Most of the research in this direction has been carried out for alloy steels. Studies into corrosion of carbon steels have been performed in various media or using alloys containing different amounts of carbon or other admixtures. There are no data on the corrosion resistance of grey cast iron in water with an increased content of chlorides.

For example, in paper [1], an investigation was conducted to study the effect of NaCl on the corrosion of steel in solutions containing H₂S. The establishment of a partial contribution of chloride ions to steel corrosion in this case is impossible because of the low pH value of hydrogen sulfide-containing solutions.

Studies into corrosion of mild steel, carried out in paper [2], have been accompanied by a change in the pH of water due to the interaction of corrosion products with the components of the solution. Data from article [3] have been obtained as a result of a study of the corrosion of samples in the marine atmosphere. Although the corrosive process in this case occurs in the condensate film, it is impossible to use these data for the circulating cycles. Corrosion in the volume of the solution differs from corrosion in a thin layer of the solution under atmospheric corrosion.

Pitting corrosion of steel investigated in stratal waters [4], in addition to chloride ions, is determined by the presence of sulfate, hydro carbonate- and sulfide ions. Data of study in paper [5] refer to the corrosion of steel in neutral solutions of sodium chloride. Measurements of pH of the medium have not been carried out in this work, which does not make it possible to extend its results to circulating waters with a constant pH value.

The process of corrosion of stainless steel in salt water with an alkaline pH value has been investigated in article [6]. The authors have found out that increasing of the pH of water leads to a decrease in the rate of corrosion. The results relate to the investigation of local corrosion of welded joints and cannot be used to assess the corrosive behavior of the material in the product volume.

Paper [7] reported the investigation of intergranular corrosion of complex chromium-nickel-molybdenum steels. The paper presents data on the propensity of austenitic steels to intergranular, crevice and pitting corrosion, and also compares the steels of one class but with different compositions. Corrosive medium is sea water, the mineralization of which occurs due to the presence of not only chloride ions. Therefore, it is incorrect to use these results to estimate the corrosion of ferrous metals in circulating water.

Data of study [8] have shown that an increase in the concentration of chloride ions leads to a deterioration in the protective properties of a passive film of alloyed chromium-nickel steel. The authors think that it is caused by the conversion of iron hydroxide to a soluble iron oxychloride. Corrosion process has been studied in acidic solutions (pH=2), and circulating water have had a neutral or slightly alkaline pH value. The same applies to data from study [9], where a solution of FeCl₃, which is a strong oxidant and is absent in circulating water, has been used as a corrosive medium.

The admixture of sodium chloride causes the emergence of pitting corrosion of chromium-nickel steel in solutions of sulfuric acid and an increase in the admixture content accelerates the growth of active pitting and initiates the resumption of growth of passive pitting [10]. The studies have been carried out in solutions of sulfuric acid using samples of alloyed steel, which is not typical for circulating water supply systems.

Thus, corrosive aggressiveness of sodium chloride under specific conditions of operation of circulating water supply systems has been insufficiently explored. The influence of Cl ions on the corrosive behavior of carbon steel and gray cast iron in industrial water at elevated temperatures (40–70 °C) and alkaline pH has not been sufficiently illuminated. There is insufficient information on the corrosive behavior of ferrous metals in such water in the waterline zone. Such experimental data are necessary for the development of a new method for protection of structural materials of circulating water supply systems. This method is based on the formation of passive films on the surface of ferrous metals when sodium hydroxide is added to circulating water [11].

3. Study objective and tasks

The objective of present study was to investigate the effect of parameters of circulating water (the concentration of chloride ions and temperature) on the rate and nature of the corrosive destruction of carbon steel and gray cast iron.

To achieve this objective, the following tasks were set:

- to study the impact of chloride ions on the corrosion of ferrous metals in circulating water with pH=12 and to determine the concentration limit of NaCl, at which the metals under study have corrosion resistance acceptable for circulating water supply cycles;
- to study the effect of the circulating water temperature on the rate and nature of corrosion of steel and gray cast iron in circulating water with a fixed concentration of NaCl and pH value.

4. Methods of study of the corrosion of cast iron and carbon steel in circulating water

Preparation of samples has been carried out according to the generally accepted scheme [12]. Samples have been ground with a flap wheel and polished with a felt wheel using chrome paste. The samples have been degreased with an alkaline solution, washed with distilled water, and dried. Samples have been hung up on glass hooks and placed in 1-liter
flasks with a corrosive medium. The tests have been carried out on samples made of carbon steel St. 3 grade of a size of 40×25×3 mm and cast iron CI 18-36 grade of a diameter of 30 mm and a thickness of 2–3 mm.

The study has been carried out at a solution temperature of 22 °C. The pH value of the solution has been monitored every 24 hours and, if necessary, corrected. The change in the mass of the samples has been determined by weighing them on the ANG 200 scale. Investigations into the impact of water temperature on the corrosion of ferrous metals have been carried out using the UTU-4 thermostat. The nature of the corrosion damage of the metal has been assessed visually and with the help of a magnifier with a magnification of 8–12 times. The study duration was 550 hours. The corrosion rate of the samples has been evaluated by the value of the mass index $k_a$ (g·m$^{-2}$·h$^{-1}$).

Electrochemical study was carried out using the IPC-PrO potentiostat. The investigation cell has been a U-shaped glass vessel in which the anodic and cathodic spaces have been separated by a glass diaphragm. Platinum has been used as an auxiliary electrode. Electrode potentials have been measured relatively to the silver chloride reference electrode. Cylindrical samples of steel St. 3 grade and cast iron of CI 18-36 grade of a diameter and length of 10 mm have been used for the study. The working surface was the end of the sample. The samples were kept in the solution for 30 minutes before the experiment. The current values on the polarization dependences have been fixed after holding the electrode for a given potential for 5 minutes. As a result of the polarization measurements, the stationary potential $E_0$, the pitting potential $E_p$, the region of the passive state $ΔE$ of the samples studied have been determined.

The results have been processed using the Origin 7.5 software product. Approximation of experimental data in the software was carried out using a polynomial function of the third degree.

5. Results of study of corrosion of St. 3 steel and CI 18-36 cast iron in circulation water

5.1. The impact of the concentration of chloride ions on the corrosion of steel and cast iron

The results of weight tests of the influence of NaCl concentration on the corrosion rate of carbon steel and gray cast iron in circulating water with pH=12 are shown in Fig. 1. Based on the data obtained, it can be said that the reliable retarding action of NaOH at pH=12 becomes apparent in a solution with a NaCl concentration of up to 10 g·l$^{-1}$. The mass corrosion index of the metals investigated $k_a$ becomes larger than 0.1 g·(m$^2$·h)$^{-1}$ with an increase in the concentration of more than 10 g·l$^{-1}$. The corrosion resistance of the metals studied is 6, that is, they belong to the group of low-resistant metals. The nature of corrosion of the samples tested in the volume of the solution is general, of the samples tested in the zone of the waterline – it is pitting.

The influence of NaCl concentration on the electrochemical behavior of CI 18-36 cast iron (Fig. 2) in circulating water with pH=7, as well as CI 18-36 cast iron and St.3 steel in circulating water with pH=12 (Fig. 3), were investigated. Analysis of the course of the anode curves obtained in water with pH=7 shows that the cast iron is in the active state. The active state of cast iron does not depend on the concentration of NaCl. The incidence of the polarization curves increases with increase of NaCl concentration. This indicates the activation of the anode process by chlorine ions.

Fig. 1. Influence of NaCl concentration on corrosion rate of St. 3 steel and CI 18-36 cast iron in circulating water with pH=12 and temperature 22 °C: 1 – St. 3 in the volume of electrolyte; 2 – St. 3 in the waterline zone; 3 – CI 18-36 in the volume of the electrolyte; 4 – CI 18-36 in the waterline zone

Fig. 2. Anodic polarization dependencies of CI 18-36 cast iron in water with pH=7. $c_{NaCl}$ g·l$^{-1}$: 1 – 0; 2 – 5; 3 – 6.6; 4 – 10.0

Fig. 3. Influence of pH value on the anodic polarization dependences: a – polarization dependences of steel St. 3 in water at pH=12. $c_{NaCl}$ g·l$^{-1}$: 1 – 0; 2 – 0.165; 3 – 0.195; 4 – 1.0; 5 – 1.65; 6 – 5.0; 7 – 6.6; 8 – 9.9; 9 – 16.5; 10 – 20.0

With the increase in NaCl concentration, $E_{corr}$ of cast iron is shifted to the negative side much more than $E_{corr}$ of carbon steel. The shift of $E_{corr}$ to the negative direction reduces the region of the passive state of cast iron from 0.58 to 0.12–0.25 V, and of carbon steel from 0.7 to 0.2 V. These data show that more reliable protective films are formed on the steel. The region of the passive state on the anode curves
taken in media with pH=12 exists for solutions with NaCl concentration up to 10 g/l. At $c_{NaCl}=16.5$ g/l, the passive state is minimal, and at 20 g/l is absent. The presence of a passive state at $c_{NaCl}<10$ g/l explains the decrease in the corrosion rate of carbon steel and cast iron during anodic control of the process. Thus, the results of electrochemical investigations confirmed and explained the results of weight tests: an effective inhibiting effect of NaOH at pH=12 is observed only in a solution with a NaCl concentration of up to 10 g/l.

There is a region of the passive state on all anodic polarization curves at pH=12. The values of the stationary potential $E_s$, the potential of pitting, $E_{pi}$, and the extent of the region of the passive state $\Delta E$ depend on the concentration of sodium chloride in the circulating water (Table 1).

**Table 1**

| Metal          | $c_{NaCl}$ g·l$^{-1}$ | $E_s$ V | $E_{pi}$ V | $E_{st}$ V | $\Delta E$ V |
|----------------|------------------------|---------|------------|------------|-------------|
| CI 18-36       | 0.000                  | -0.25   | 0.70       | 1.00       |             |
| CI 18-36       | 0.165                  | -0.25   | 0.24       | 0.49       |             |
| CI 18-36       | 0.495                  | -0.30   | 0.28       | 0.58       |             |
| CI 18-36       | 1.000                  | -0.30   | 0.08       | 0.38       |             |
| CI 18-36       | 4.950                  | -0.25   | -0.10      | 0.25       |             |
| CI 18-36       | 6.600                  | -0.40   | -0.28      | 0.12       |             |
| St. 3          | 0.000                  | -0.11   | 0.60       | 0.70       |             |
| St. 3          | 0.165                  | -0.12   | 0.60       | 0.70       |             |
| St. 3          | 0.495                  | -0.15   | 0.27       | 0.42       |             |
| St. 3          | 1.000                  | -0.12   | 0.15       | 0.30       |             |
| St. 3          | 1.650                  | -0.12   | 0.24       | 0.36       |             |
| St. 3          | 4.950                  | -0.13   | 0.14       | 0.34       |             |
| St. 3          | 9.900                  | -0.15   | 0.10       | 0.20       |             |
| St. 3          | 16.500                 | -0.15   | -0.10      | absent     |             |

The study conducted allow us to conclude that the corrosion rate of steel ($k_c=0.08$ g·m$^{-2}$·h$^{-1}$) and gray cast iron ($k_c=0.10$ g·m$^{-2}$·h$^{-1}$) in circulating water with $c_{NaCl}=10$ g/l and pH=12 does not exceed the rate of corrosion of these metals in circulating water with $c_{NaCl}=1$ g/l and pH=7.

### 5.2. Impact of water temperature on corrosion of steel and cast iron

The temperature of the hot water entering the cooling tower of the recycling cycle should be no higher than 35 °C, and the cooled one - no higher than 20 °C [12]. During the summer period, due to various reasons, these values may exceed the regulations by 10–20 °C. Therefore, the corrosion of steel and cast iron in open circulating systems at temperatures of 20–60 °C has been investigated. The investigation was carried out in recycling water with $c_{NaCl}=1.17$ g/l with pH=7 and in water with $c_{NaCl}=5.85$ g/l with pH=12. The results of the tests are given in Table 2.

The increase of the temperature to 40 °C leads to an increase in the corrosion rate by a factor of 2–4. At temperatures of 50–60 °C, the rate of corrosion, especially in the waterline zone, becomes significant. Therefore, the water temperature should not be raised above 40 °C while operating the circulating system.

It should be noted that the rate of corrosion of carbon steel and cast iron at pH=12, even in mineralized water with a concentration of 5.85 g/l NaCl at all temperatures, as a rule, does not exceed the corrosion rate of these metals in circulating water with $c_{NaCl}=1$ g/l and pH=7. To explain these regularities, anode curves of carbon steel and cast iron have been obtained at different temperatures in the solutions under study (Figs. 4, 5).

**Table 2**

| Metal          | Test area     | $t$, °C | $k_c$ g·m$^{-2}$·h$^{-1}$ | Corrosion nature |
|----------------|---------------|---------|--------------------------|------------------|
| St. 3          | in the solution volume | 20     | 0.0080                  | spots            |
| St. 3          | in the solution volume | 60     | 0.1330                  | spots            |
| St. 3          | in the waterline zone  | 20     | 0.1860                  | pitting          |
| St. 3          | in the waterline zone  | 60     | 0.3500                  | pitting          |
| CI 18-36       | in the solution volume | 20     | 0.0130                  | uniform          |
| CI 18-36       | in the solution volume | 60     | 0.1290                  | spots            |
| CI 18-36       | in the waterline zone  | 20     | 0.1780                  | spots            |
| CI 18-36       | in the waterline zone  | 60     | 0.4446                  | spots            |

**Fig. 4.** Impact of water temperature on anodic polarization dependencies: $\alpha$ – polarization dependencies of steel St.3; $t$, °C: 20(1); 40(2); 50(3); 60(4). $c_{NaCl}=5.85$ g·l$^{-1}$; pH=12; $b$ – polarization dependencies of cast iron CI 18-36; $t$, °C: 1 – 20; 2 – 40; 3 – 50; 4 – 60. $c_{NaCl}=5.85$ g·l$^{-1}$; pH=12

**Table 3**

| Metal          | Test area     | $t$, °C | $k_c$ g·m$^{-2}$·h$^{-1}$ | Corrosion nature |
|----------------|---------------|---------|--------------------------|------------------|
| St. 3          | in the electrolyte volume | 20     | 0.0120                  | spots            |
| St. 3          | in the electrolyte volume | 40     | 0.0483                  | spots            |
| St. 3          | in the electrolyte volume | 50     | 0.0850                  | spots            |
| St. 3          | in the electrolyte volume | 60     | 0.0905                  | spots            |
| St. 3          | in the waterline zone  | 20     | 0.0636                  | spots            |
| St. 3          | in the waterline zone  | 40     | 0.1413                  | spots            |
| St. 3          | in the waterline zone  | 50     | 0.1706                  | spots            |
| St. 3          | in the waterline zone  | 60     | 0.3225                  | spots            |
| CI 18-36       | in the electrolyte volume | 20     | 0.0106                  | uniform          |
| CI 18-36       | in the electrolyte volume | 40     | 0.0453                  | uniform          |
| CI 18-36       | in the electrolyte volume | 50     | 0.1241                  | uniform          |
| CI 18-36       | in the electrolyte volume | 60     | 0.0807                  | uniform          |
| CI 18-36       | in the waterline zone  | 20     | 0.1067                  | spots            |
| CI 18-36       | in the waterline zone  | 40     | 0.1321                  | spots            |
| CI 18-36       | in the waterline zone  | 50     | 0.1813                  | spots            |
| CI 18-36       | in the waterline zone  | 60     | 0.3073                  | spots            |
The passive state decreases on carbon steel from 0.3 to 0.1 V, on cast iron from 0.25 to 0.05 V with an increase in temperature in mineralized water with pH=12. Dependencies obtained at higher temperatures shift to the direction of more negative potentials. This indicates a lesser polarization of the anode process and explains the increase in the corrosion rate of steel and cast iron with an increase in temperature.

The passive state is absent on the anode curves at all temperatures in the mineralized water with pH=7 and NaCl concentration of 1.17 g l⁻¹ (Fig. 5).

![Graph showing impact of water temperature on anodic polarization dependencies of steel.](image)

With an increase in temperature, the anode curves of the steel are shifted toward negative potentials, which also indicates a decrease in the polarization of the anode process. Probably, in this case the corrosion rate of the metal will be determined by the flow of oxygen to the cathode sections. The limiting stage of the corrosion process is the diffusion of the depolarizer to the cathode regions.

6. Discussion of the study results of corrosion of St. 3 steel and CI 18-36 cast iron

The protective action of sodium hydroxide is associated with the formation of oxide films on the surface of steel and cast iron, which transfer the metal into a passive state. The formation of mixed oxide-hydroxide films is more likely to occur on the surface of iron alloys on alkaline media. The formation of pure hydroxide films is unlikely, since the friable and porous sediment of iron hydroxides formed in alkali, which is weakly bound to the metal, cannot cause the passivation. Formation of the protective layer occurs according to the following scheme: ions Fe²⁺, which react with OH⁻ ions and form Fe(OH)₃ sediment, enter the solution. The sediment undergoes further anodic oxidation, and a dense oxide layer is formed. When anodic polarization occurs in the region of the pitting potential, Fe(OH)₃ is formed on the metal, the amount of which depends on the number of pits formed under the action of Cl⁻ ions. When the polarization is reversed to the potential (−0.6) V, Fe(OH)₃ is transformed to Fe(OH)₄. The amount of Fe(OH)₄ depends on the concentration of NaCl. Peaks are observed on the cathode and anode curves at low NaCl concentrations in the potential region (−0.3) to (−0.5) V. According to Pourbaix diagram, these peaks correspond to the following reactions:

$$3\text{FeO}_3 + 2\text{H}^+ + 2\text{e}^- \rightarrow 3\text{Fe(OH)}_3 + \text{H}_2\text{O};$$

$$E^\circ = 0.271 – 0.059\text{pH};$$

(1)

The pitting potential of the steel is shifted toward negative direction at NaCl concentration of more than 9.9 g l⁻¹ in a solution with pH=12. This reduces the area of the passive state. At a NaCl concentration of 16.5 g l⁻¹, this region is absent, i.e., the surface is in the active state.

The reason for the activation of iron in the presence of chloride ions is their introduction into the surface oxide. Activation of iron by chlorine ions is associated with their introduction into the γ-Fe₂O₃ lattice with the formation of a substitution structure [13]. Chlorine, when replacing oxygen, acts as a donor forming a center with a positive effective charge. This leads to the appearance of an equivalent number of free electrons in the oxide and an increase in the dissolution rate of iron due to the facilitated transfer of charge and material through the surface layer.

The results of adsorption investigations show that a phase oxide film, which is capable of adsorbing chloride ions, is formed on the metal surface [14]. An increase in the adsorption of Cl⁻ ions is observed at those pH values where the protective action of OH⁻ ions, which is determined by the formation of Fe(OH)₂, with a developed surface, is realized. As the pH increases, the amount of adsorbed chlorine ions increases, which is associated with an increase in the thickness of the hydroxide films due to a decrease in their solubility. The presence of a threshold concentration of NaCl, at which the anode process is sharply activated, is due to the achievement of a concentration gradient, at which Cl⁻ ions can diffuse to the metal surface through the protective layers.

7. Conclusions

1. The impact of Cl⁻ ion concentration on the rate and nature of the corrosive destruction of carbon steel of St. 3 grade and gray cast iron of CI 18-36 grade in circulating water has been investigated. It has been shown that the increase in NaCl concentration causes a shift in the stationary potentials of iron and steel to the negative direction, and this effect is more pronounced for the first alloy. The shift of the value of $E_{\text{p}}$ to the negative direction reduces the region of the passive state of cast iron from 0.58 to 0.12–0.25 V, and of carbon steel from 0.7 to 0.2 V. These data have confirmed that steel forms more stable passive films. The passive state of ferrous metals at pH=12 exists for solutions with NaCl concentration of up to 10 g l⁻¹. With $c_{\text{NaCl}}=16.5$ g l⁻¹, the region of the passive state is practically absent. The effective inhibitory effect of NaOH at pH=12 is observed in solutions with a concentration of $c_{\text{NaCl}}<10$ g l⁻¹.

2. An increase in the temperature of the circulating water leads to an increase in the rate of corrosion by a factor of 2–4. At temperatures of 50-60 °C the overall corrosion rate of steel and cast iron becomes significant. Particularly increases the rate of corrosion in the waterline zone. Therefore, when using equipment made of St.3 steel and CI 18-36 cast iron, the temperature of the circulating water should not exceed 40 °C. An increase in the pH of the circulating water causes a decrease in the rate of corrosion of these alloys. In the circulating water with $c_{\text{NaCl}}=5.85$ g l⁻¹ and pH=12, the corrosion rate of these metals does not exceed the corrosion rate in circulating water with $c_{\text{NaCl}}=1.2$ g l⁻¹ and pH=7.
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