

SECTION 2

THERMAL AND FAST REACTOR MATERIALS

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REVIEW: INVESTIGATION OF PROCESSES OF DAMAGE TO THE HEAT TRANSFER TUBE OF WWER-1000 STEAM GENERATORS

O.G. Rudenko, S.V. Gozhenko

National Science Center “Kharkov Institute of Physics and Technology”, Kharkiv, Ukraine

E-mail: rudenko@kipt.kharkov.ua

The analysis of operational damages of heat transfer tubes (HTT) in steam generators (SG) of WWER-1000 power units was carried out. The processes of contamination of the surfaces of HTT of SG PGV-1000M are considered. The processes of formation and growth of corrosion pits on the surface of heat transfer tubes are analyzed. The process for the initiation of corrosion pits and the development of cracks in the heat transfer tubes metal, which includes the combined action of a corrosive environment and non-metallic inclusions in heat transfer tubes, was proposed.

INTRODUCTION

Heat transfer tubes (HTT) are one of the most critical elements that determine the actual service life of steam generators (SG) at NPPs with WWER. HTTs are a barrier between the first radioactive circuit and the water of the secondary circuit, which has contact with the environment, and therefore this barrier must be reliable. The heat transfer tubes in the SG operate under very harsh conditions and damage to the HTT during operation leads to a long shutdown of the entire power unit of the nuclear power plant and to significant economic losses.

In the process of SG operation in water containing corrosive components, corrosion cracking (CC) of HTT occurs due to the occurrence of chemical and electrochemical processes at the interface between the metal surface and the aggressive environment. At present, a large complex of scientific works is being carried out in the world and many works have been published [1–5] on the study of the problems of damage to the CC of the HTT.

However, the problem has not been finally resolved. The conducted studies have shown that the HTT resource during SG operation is determined by the rate of several main damaging processes [6–8]:

- chloride-oxygen CC;
- the formation of corrosion pits due to the presence of copper in sediments;
- hydrogen embrittlement.

The joint effect of corrosion processes leads to damage to the HTT metal much faster than the action of each separately, which leads to complications in predicting the life of the HTT. Due to the need to consider several simultaneously occurring processes of HTT damage, there is no unified approach that allows us to evaluate the effect of the above processes on the corrosion resistance of austenitic stainless steel 08Kh18N10T.

The target of the work is to analyze damage to the metal of heat transfer tubes, at which it is possible to predict the resource of HTT. When analyzing the HTT damage mechanisms, materials were used summarizing

the operating experience of SG WWER-1000, the results of HTT monitoring during operation, the results of metallographic studies of cut samples of damaged heat exchange tubes, as well as literature data on the conditions for the initiation and growth of defects during SG operation.

1. INFORMATION ABOUT STEEL GRADE 08Kh18N10T AS HTT SG

Heat transfer tubes are made of steel grade Kh18N10T. The chemical composition of the alloy, in particular the concentrations of carbon, chromium, nickel, titanium, nitrogen, molybdenum, etc., has a significant effect on the tendency of steel to undergo corrosion. Based on this, it is necessary to know the effect of alloying and structural components of austenitic stainless steel on their corrosion resistance. Table 1 [9–11] summarizes the known literature data on the effect of the main alloying elements, carbon and second phases on the resistance of austenitic steels against local corrosion.

Table 1

Influence of alloying elements and second phases of austenitic steels on their resistance to local corrosion: positive (o), negative (x), neutral (xo)

Alloying elements/ phases	Corrosion resistance in chloride-containing environments		
	CC	slotted	ICC
Cr	o	o	o
Mo	o	o	o
Si	o	o	o
N	o	o	o
Ni	o	o	o
Mn	x	x	x
C	x	x	x
σ -phases	x	x	x
δ -ferrite	xo	xo	xo
α' -phases	x	x	x

It is well known that an increase in the corrosion resistance of austenitic steels is achieved by reducing the carbon content in them and additional alloying with elements that passivate the metal surface. However, the problem of preventing local corrosion of austenitic steels in a corrosive environment has not been solved now and, therefore, it is necessary to continue research.

Other factors include microstructure and plastic deformation. The influence of the structural components of austenitic steel on corrosion resistance is mainly reduced to the theory of intergranular corrosion (ICC). The reason for the manifestation of the tendency of austenitic steels to intergranular corrosion is the precipitation of secondary high-chromium carbides of the Me_{23}C type at the boundaries of austenitic grains during heating in the temperature range of 450...850 °C, which leads to depletion of the grain body in chromium and an increase in the heterogeneity of the structure. It is noted in [12, 13] that if chromium carbides precipitate along grain boundaries, this causes not only transcrystalline cracking, but also mixed transcrystalline and intergranular cracking. Compared to iron, chromium has a greater affinity for oxygen, which increases the corrosion resistance of austenitic steels. It is noted in [14] that the presence of δ -ferrite in steel 08Kh18N10T increases its resistance to stress corrosion cracking (SCC) and reduces the tendency of steel to intergranular corrosion [15, 16]. Intergranular corrosion consists in the selective destruction of metal along grain boundaries, which usually leads to the formation of a network of cracks upon contact with an aggressive medium. In heat exchange tubes made of austenitic steel grade 08Kh18N10T, ICC occurs by the type of transgranular damage, but under appropriate conditions, intergranular corrosion can occur (Fig. 1).

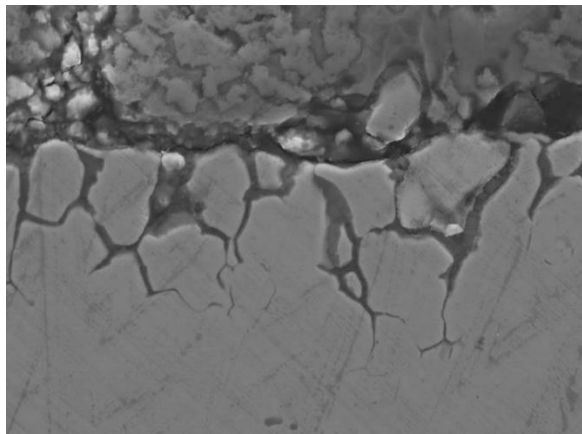


Fig. 1. ICC in heat transfer tubes made of austenitic steel 08X18H10T [17]

There are two different approaches to ensure the resistance of austenitic steel grade 08Kh18N10T against MCC:

1. Stabilization of steels with strong carbide-forming elements (Ti, Nb) while maintaining the carbon content within 0.06...0.12%. In stabilized steels titanium and niobium form very stable carbides with carbon, which do not dissolve in a solid solution even at 1323...1373 K. In this steel, the content of carbon not bound into titanium and niobium carbides should be

close to the solubility limit of carbon in austenite at room temperature. In this regard, the formation of secondary chromium carbides and the depletion of grain boundaries in chromium during heating in the temperature range of 450...850 °C are largely suppressed. For austenitic steels of the Kh18N10T type, a ratio was developed, where the contents of niobium and titanium in relation to carbon which should exceed at least 10- and 5-times the amount of carbon, respectively. If the steel is alloyed with nitrogen, then it is necessary to stabilize it with niobium, since it is more evenly distributed in the cast metal and is less prone to the formation of coarse carbonitride inclusions. The resistance of austenitic steels to ICC depends significantly on heat treatment and can be improved by dissolving chromium carbides. In any case, it is advisable to subject austenitic steel to stabilizing annealing at ≈ 1100 K. In this case, chromium carbides coagulate, and part of the carbon dissolved in the solid solution goes to form secondary TiC and NbC carbides.

2. The reduction in the carbon content in steel should not exceed 0.03%, without the introduction of stabilizing elements. However, it has also recently been noted in the literature that attention should be paid to the effect of austenite stability and second phase inclusions on corrosion pitting resistance for such steels. If stainless steel has an unstable austenite structure, then its long-term thermal aging can be accompanied by the formation of δ -ferrite and σ -phase, causing the metal to be prone to intergranular corrosion and to the formation of corrosion pits.

It should be noted that during deformation, bending, welding of austenitic steels (non-magnetic), the metal becomes magnetic. This is due to the formation of a ferrite phase. Part of the unstable austenite in stainless steels turns into martensite (α' -phase), which reduces the corrosion resistance of the metal. The metal of the spacer grating after punching and HTT during manufacture and assembly into a tube bundle (deformation, welding) becomes magnetic. Thus, the creation of local (permanent) magnetic fields, even with an insignificant magnetic intensity, can accelerate the process of accumulation of sediments of corrosive impurities from the secondary water on the surface of the HTT [18]. Therefore, after laying the HTT on the spacer grids in the SG, it is necessary to demagnetize the HTT along its entire length. In addition, during neutron irradiation of austenitic steel, inclusions of δ -ferrite can turn into a σ -phase (FeCr), which reduces the resistance of the metal against local corrosion in places depleted in chromium. Based on this, the main task is to reduce the tendency of the HTT metal to local types of corrosion in corrosive environments.

1.1. DAMAGE OF HEAT TRANSFER TUBES SG WWER-1000

The operating experience of the HTT SG has shown that Kh18N10T steel is not without drawbacks. During SG operation, the initiation and subsequent development of corrosion defects in the heat transfer tubes was recorded, which can lead to depressurization of the primary circuit. HTT defects were found both in free zones and in slit gaps between spacer grids of HTT

[19, 20]. The process of damage to the SG heat transfer tubes is due to the arrival of metal corrosion products of the equipment of the second circuit of the NPP, which are made of carbon steel and copper-containing alloys.

The following main types of operational corrosion damages were recorded on the heat transfer tubes SG surface in accordance with GOST 9.908-85 [21]:

- corrosion pits (damage, the depth of which is approximately equal to the width);
- single corrosion cracks (damage characterized by the formation of a deep, branched crack);
- pitting (damage, the depth of which is much greater than the width);
- corrosion spots (damage having a shallow depth and a large area).

The metallographic analysis of the cutout defective HTTs showed that, in general, the defects have the character of corrosion pits on the outer surface [22].

From the analysis of HTT damage, the process of CC of austenitic steel can be represented by the most common theory [22–24]. During operation, corrosion products are sediments on the surface of the HTT. In places where corrosion products are sediments, an aggressive environment is formed with an increased concentration of chlorides and a low pH value, which leads to irreversible destruction of the protective layer of austenitic steel by replacing oxygen in the oxide layer with chloride ions. In this electrochemical process, the unprotected metal surface acts as the anode, the oxide layer is the cathode, and the anode dissolves in such an aggressive environment with the formation of corrosion pits. In the presence of significant residual stresses and under the influence of working loads, CC occurs in the HTT metal. Thus, one of the main conditions for reducing HTT damage is to prevent the accumulation of sediments on the HTT, inside which an aggressive environment is formed containing chloride ions, oxygen, hydrogen and causing the destruction of the protective film. Therefore, it is necessary to strictly comply with the requirements of the water-chemistry regime (WCR) at Ph 9.2, according to the regulatory guide [28, 29].

2. THE MAIN FACTORS INFLUENCING THE PROCESS OF HTT DAMAGE

The main factors influencing the development of HTT injuries [25–28] are:

- Increase in the concentration of corrosive impurities in the SG water. Violation of the WCR.
- Excessive sediments on heat exchange tubes (higher than standard).
- Presence of internal residual stresses and external tensile stresses in HTT during SG operation.
- Influence of temperature on the corrosion damage of HTT.

At present, there is no unified approach to the process of corrosion damage in HTT, considering the joint influence of the above factors. Based on this, there is no single model that describes the process of corrosion failure of austenitic steel.

Corrosion damage to HTT is most often associated with the destruction of the protective oxide layer and with the anodic dissolution of the metal. The main

corrosive components in the SG water are chlorine ions and oxygen, at a certain concentration of which corrosion pits are formed in sediments on the HTT surface [29, 30]. It should be noted that the pH near cracks is significantly lower than the pH of the environment, differs by more than 3 values [31, 32], which also significantly accelerates corrosion.

In addition to the corrosive environment, the damage to the HTT is affected by the presence of internal residual and external tensile stresses. The presence of internal and external stresses contributes to an easier flow of SCC of HTT, especially at the operating temperature of the SG. SCC manifests itself as a brittle propagation of cracks in the metal. The types of cracks are of intergranular, transcrystalline, and mixed types and largely depend on the alloy composition, microstructure, emerging stresses, and corrosive environment. Mostly carbon steel, nickel-based alloys tend to intergranular cracking, and in austenitic stainless steels, as a rule, cracks are initiated at the bottom of corrosion pits and transgranular or mixed cracking is usually observed, while the cracks have a branching appearance.

2.1. WATER-CHEMISTRY MODE OF THE STEAM GENERATOR

Salts, acids, chloride ions, sulfates, sodium, equipment metal corrosion products, and oxygen are the main water contaminants. During long-term operation of the SG, most of the corrosion products remain inside the vessel in the form of sediments, including on the HTT surface. The sludge can only be removed by periodic flushing during scheduled maintenance work.

The organization of the WCR has the goal of maintaining the concentration and quantitative composition of impurities within the limits that ensure reliable and safe operation of equipment for nuclear power plants with WWER [31]. The Table 2 shows the water quality standards in SG for WWER-1000 according to regulatory guide [28, 29].

Thus, to prevent damage to the HTT SG, it is necessary:

- reduction of the total salt content in the SG water (chlorides and sulfates);
- stable maintenance of the pH value of water in the weakly alkaline region in all modes of SG operation. If the pH drops below the neutral value, additional water treatment is necessary;
- low oxygen content in SG water.

To maintain pH within the established limits of 9.0 ± 0.2 , corrective water treatment with ammonia is carried out at all WWER-1000 power units [33, 34]. The volatile part (NH_3) does not accumulate in water, but is removed with steam, therefore, alkalization of the medium does not occur. In some works [28, 31, 32], it is noted that while maintaining the pH values of the medium at a level of up to 10, an increase in the corrosion resistance of austenitic steel is observed. It is noted in [31] that at low pH values < 3 , the general corrosion rate increases significantly and, according to GOST 13819-69 [36], becomes dangerous. A shift in pH to a higher region ($\text{pH} > 11$) increases the supply of

corrosive impurities and can provoke accelerated intergranular corrosion or alkaline CC [56].

Table 2
Requirements for the quality of water entering the SG WWER-1000 [35]

Control indicators	Maximum concentration of impurities in water of SG
Concentration of chloride ions, $\mu\text{g/kg}$	< 100
Sulfate ion concentration, $\mu\text{g/kg}$	< 200
Oxygen concentration, $\mu\text{g/kg}$	< 10
Iron concentration, $\mu\text{g/kg}$	< 15
Copper concentration, $\mu\text{g/kg}$	< 3
pH value at 25 °C	9.0±0.2
Sodium concentration, $\mu\text{g/kg}$	< 300
Hydrazine concentration, $\mu\text{g/kg}$	20

It should be noted that the observance of water chemistry in the secondary circuit is practically the only way to increase the duration of SG operation at moderate capital costs. However, it should be noted that, in principle, it is impossible to radically remove the corrosion of secondary circuit equipment elements by changing the water-chemistry.

2.2. THE FORMATION AND GROWTH OF SEDIMENTS ON THE HTT

The main part of the resulting corrosion products accumulates in the SG. Over time, they accumulate in the form of sediments on the HTT and the surface of the SG body. When studying the structure and properties of sediments on the heating surfaces of austenitic steels, some authors [35] concluded that the intensity of sediments in austenitic steel depends mainly on surface irregularities and roughness. On the surface of pipes there are always defects caused by the method of production, and which are acceptable in accordance with the regulatory documentation.

The formation of sediments is a complex physicochemical process associated with the formation of the smallest particles due to an increase in the concentration of salts during repeated evaporation of water in SG [36, 37]. There are centers of formation of sediments from particles, the chemical composition of which is closest to the saturation state. It should be noted that the rate of sediments accumulation at SG operating temperatures on Kh18N10T steel specimens is 3–4 times lower compared to tests at room temperature over the same period [38]. Based on the analysis of the sludge removed from the SG during chemical washings, it can be concluded that, as a rule, it is difficult for large particles to attach to the HTT surface, due to the water flow rate in the SG. They continue to grow in the liquid volume and settle on the inner surface of the SG. Blowing can remove only a part of the corrosion products (sludge) that have not yet formed in the form of insoluble sediments within 35,000 h.

The results of metallographic studies of the surface of the sediments showed the formation of channels with a diameter of ~10 μm [39]; in this case, settled spherical particles can be observed on the surface of the

sediments. In places where sediments are formed, overheating of the HTT metal occurs. Soluble salts, chlorides, oxides accumulate under the sediments, which, when a certain concentration is reached, affect the formation of corrosion pits.

Analysis of the data showed that the rate of accumulation of sediments of corrosion products on the surface SG for a yearly campaign with hydrazine-ammonia water chemistry is 11.8 g/m^2 , and with ethanolamine water chemistry it is 4.8 g/m^2 [40, 41]. The calculated rate of sediments of corrosion products on the heat exchange surface SG in the ammonia-hydrazine mode is 1.48 $\text{mg}/(\text{m}^2\cdot\text{h})$, while in the ethanolamine mode this value decreased to 0.6 $\text{mg}/(\text{m}^2\cdot\text{h})$. Estimated calculation of heat transfer characteristics in SG at different (hydrazine-ammonia and ethanolamine) water-chemistry at the end of a one-year campaign showed that, under operating modes, power limitations begin for ethanolamine water chemistry – on day 192, and for hydrazine-ammonia – on day 83, which indirectly confirms the formation of sediments on the surface of HTT [41]. It should be noted that such a change in the water-chemistry allows you to increase the period between washing the SG.

Thus, the process of sediments formation is based on the phenomenon of solid phase separation from a supersaturated solution of salts in SG water. The intensity of the formation of salt sediments is due to their content in the water.

2.3. ESTIMATION OF OPERATING STRESSES DURING THE OPERATION OF SG

In HTT, internal stresses can arise because of technological operations at the manufacturing stage, after thermomechanical processing, as well as during welding, installation and repair. In some cases, the magnitude of internal stresses can be comparable to the yield strength of steel. In this case, the alloy cannot be used in places where stresses occur that exceed the yield strength from the combined influence of internal and external stresses. Table 3 [42] shows the main parameters of the PGV-1000 SG, which affect the occurrence of stresses in the HTT.

Table 3
Basic data on heat transfer tubes of WWER-1000 SG and parameters of the primary and secondary circuits, according to regulatory guide [44]

Parameter	Meaning
Number of tubes, pcs	10978
Average tube length, m	14
Tube size, mm	16x1.5
Stress in metal, MPa	87
Temperature drop in the tube wall (maximum), °C	45.2
Steam capacity, t/h: nominal	1470
maximum allowable	1573
Maximum values in the primary circuit: pressure, MPa	16
temperature, °C	232.7
Maximum values in the second circuit: pressure, MPa	6.4
temperature, °C	278.5

It is of interest to calculate the sum of stresses that may arise in the cross section of the HTT wall during normal operation of the SG [42–46]:

- residual stresses resulting from manufacturing technology, welding, installation;
- operating stresses that arise from the effect of pressure difference between the primary and secondary circuits;
- thermal stresses arising from the difference in thermal expansion of the inner and outer walls of the HTT.

It should be noted that in a thin layer of the outer surface of the pipe ~ 0.1 mm after grinding, residual stresses $\sim 150\ldots 200$ MPa can occur [45]. Residual stresses arising in the HTT during their manufacture are almost completely removed at the austenite temperature. The total relaxation of stresses after austenite temperature of Kh18N10T steel can be $\approx 30\ldots 50$ MPa.

Operating tension arise in the HTT under the action of a pressure difference between the I and II circuits. Stresses due to internal pressure (σ_p) can be determined according to the “classical” concepts:

$$\sigma_p = \Delta P \cdot R / \delta = 46.4 \text{ [MPa]}, \quad (1)$$

where ΔP is the difference in water pressure in the 1st and 2nd circuits; R – the average radius of the tube, $R = 7.25$ mm; δ – tube wall thickness; $\delta = 1.5$ mm.

Thermal stresses (σ_T) in HTT arise because of the difference in thermal expansion of the inner and outer walls of the heat exchange tube. The maximum thermal stresses can be written as:

$$\sigma_T = (\lambda \cdot E \cdot \Delta T) / (2 \cdot (1 - \mu)) = 109 \text{ [MPa]}, \quad (2)$$

where λ is the coefficient of linear expansion, for steel 08Kh18N10T; $\lambda = 1.7 \cdot 10^{-5} \text{ deg}^{-1}$; E – Young's modulus, $E = 2 \cdot 10^5$ MPa; ΔT – temperature difference across the wall thickness; μ – Poisson's ratio, 0.3.

Proceeding from this, the total design stresses that can arise in the cross section of the HTT wall during SG operation can reach $\approx 185\ldots 240$ MPa. The conducted studies [46] showed that the stress arising in the HTT is ≈ 250 MPa. At the same time, it should be noted that more than 80% of the total damage occurs in steels with high values of residual stresses [47].

In [8, 15], based on the analysis of experimental data, an empirical dependence was obtained to estimate the operating time of the HTT before CC:

$$\lg \tau_{\kappa.p.} = 4.4 - 0.11\sigma, \quad (3)$$

where σ is the applied mechanical stress, MPa. This formula is valid for steel grade 08Kh18N9, where the stress level σ was not more than 280 MPa.

At present, it is possible to draw general conclusions, which are that:

1) CC can manifest itself even in the presence of small external tensile stresses, in the presence of sufficiently large residual stresses.

2) With increasing tensile stresses, the susceptibility to CC increases.

2.4. EFFECT OF TEMPERATURE ON DAMAGE

In almost all studies, an increase in the sensitivity to CC of stainless steel with increasing temperature is

noted. According to the data of [8], CC proceeds most intensively at temperatures of $150\ldots 250$ °C, slows down strongly at temperatures below 100 °C, and only slightly decreases in the range of $250\ldots 300$ °C.

The authors of [8], based on laboratory tests, propose the empirical dependence of the time to CC on temperature to be calculated by the formula:

$$\lg \tau_{\kappa.p.} = A + B/T, \quad (4)$$

where A and B are constants obtained after processing the test results.

According to the data of [18], in an environment with an oxygen concentration of ~ 0.1 mg/kg and a chloride ion concentration of 300 mg/kg, the dependence is valid:

$$\lg \tau_{\kappa.p.} = -11.25 + 6.5/(T \cdot 10^{-3}). \quad (5)$$

A similar experimental dependence was obtained in [15]:

$$\lg \tau_{\kappa.p.} = -11.4 + 5/(T \cdot 10^{-3}). \quad (6)$$

3. PROCESSES OF METAL DAMAGE HTT

Currently, several processes have been proposed for the occurrence and development of damage to HTT metal [8, 13, 15, 23, 25, 36, 38]. The proposed CC hypotheses, due to the presence of many factors, are sometimes based on opposite ideas. Most assumptions are based on SCC, which is associated with electrochemical phenomena. The most recognized of them is the destruction of the oxide surface layer, which is observed in a corrosive environment in the presence of stresses. In this system, the unprotected metal surface acts as the anode, the oxide film is the cathode, and the anode dissolves in the electrically conductive medium under such conditions. The resistance of austenitic steel to corrosion damage largely depends on the corrosive environment. An analysis of studies [1–22, 31, 32] showed that, in general, corrosion damage to HTT occurs with the simultaneous occurrence of several processes:

- chloride-oxygen CC;
- formation of corrosion pits due to the presence of copper in sediments;
- hydrogen embrittlement, decrease in pH.

In this case, CC is the final stage of HTT destruction under the combined influence of the above processes. An analysis of the experimental data on the crack propagation rate in different alloys under load showed almost the same value of at least 10^{-4} cm/h [12, 13], which indicates a long incubation period before crack propagation. In this case, the time of the incubation period is determined by the properties of the steel, the corrosive environment and the magnitude of the resulting stresses.

3.1. OXYGEN-CHLORIDE SCC

The surface of Kh18N10T steel is covered with a strong continuous film, which mainly consists of iron and chromium oxides. The protective film consists of two layers: the outer magnetite (Fe_3O_4) and the inner one, spinel of the $\text{Cr}_2\text{O}_3 + \text{FeO}$ type. Under the continuous oxide film there is a layer consisting mainly of chromium oxides. On the surface of the HTT in sediments, chloride ions accumulate and the pH of the

water decreases, which prevents the restoration of the protective layer of the austenitic steel of the HTT. During long-term operation of the SG at the interface between the HTT surface and the aggressive environment, the protective layer of the HTT is destroyed and the reaction proceeds with the formation of CrCl_3 and the release of oxygen. It should be emphasized that the displacement of oxygen by the adsorbed chloride ion occurs at chloride ion concentrations $C_{\text{Cl}^-} = (35 \dots 35) \cdot 10^{-3} \text{ mg/kg}$ [22]. According to the current regulations WCR [49], the permissible limit concentration is $C_{\text{Cl}^-} = 50 \dots 100 \text{ } \mu\text{g/kg}$. Consequently, there is always enough chlorides in the SG water for the occurrence of corrosion damage in austenitic steel. Literature analysis [8, 9, 26, 27] showed that the combined presence of chloride ion (activator) and oxygen (oxidant) causes a significant increase in the rate of corrosion pits on the HTT surface during SG operation.

It is of interest to estimate the time to destruction of HTT from the degree of chloride ion concentration in the sediments. At a temperature of $\approx 573 \text{ K}$, the dissolution of chloride ions in vapor is low, so the concentration of chloride ions in the sediment channels will increase [6, 16, 24, 27]. Some researchers, based on the processing of experimental data, derived empirical dependencies for estimating the time to destruction of HTT. These formulas lack many factors that affect damage to HTT, but they can be used to estimate the time to destruction of steel of the Kh18N10T type.

Thus, the authors of [50] presented a formula for estimating the destruction time of AISI-304 steel (analogous to 08Kh18N9) on the degree of concentration of chloride ions at a temperature of $366 \dots 433 \text{ K}$:

$$\lg \tau_{\text{к.р.}} = 4.824 + C_{\text{Cl}^-} (0.028 - 3.3 \cdot 10^{-4} T) - 0.0916T, \quad (7)$$

where $\tau_{\text{к.р.}}$ – time to failure, h; C_{Cl^-} is the chloride ion concentration, mg/kg; T is temperature, K.

A similar formula was presented by other researchers [13]:

$$\lg \tau_{\text{к.р.}} = 2.6 - 0.9 \lg C_{\text{Cl}^-}. \quad (8)$$

In [13], based on the processing of experimental data, an empirical dependence was proposed for estimating the time to fracture, using two factors, the chloride ion and oxygen concentrations:

$$\lg \tau_{\text{к.р.}} = \lg \tau_0 - 0.5 \lg C_{\text{O}_2} - 1.5 \lg C_{\text{Cl}^-}, \quad (9)$$

where τ_0 – coefficient, $\lg \tau_0 = -8 + 6.5/(T \cdot 10^{-3})$; $\tau_{\text{к.р.}}$ – time to cracking, h; C_{Cl^-} is the chloride ion concentration, mg/kg; C_{O_2} – oxygen concentration, mg/kg; T is temperature, K. Based on this, it becomes possible to evaluate the effect of one of the concentrations of C_{Cl^-} or C_{O_2} on the time until the destruction of HTT. The main disadvantage of this formula is the lack of influence of residual and applied tensile stresses.

An analysis of the test data calculated according to the formula for Kh18N10T steel under load in the yield strength region at $T_{\text{test}} = 300 \text{ }^\circ\text{C}$ in “pure” water for 3500 h showed that CC is not observed (Fig. 2).

According to the regulatory guide of the water-chemistry regime for SG, the concentration limit for chlorides is $C_{\text{Cl}^-} = 0.1 \text{ mg/kg}$ and for oxygen –

$C_{\text{O}_2} = 0.01 \text{ mg/kg}$. Increasing the oxygen concentration to 2 mg/kg ($C_{\text{Cl}^-} \leq 1 \text{ mg/kg}$) leads to CC after 2500 h. An increase in the concentration of chlorides to $C_{\text{Cl}^-} = 10 \text{ mg/kg}$ ($C_{\text{O}_2} = 0.2 \text{ mg/kg}$) CC is observed in less than 1000 h. From the analysis of the figure, it follows that CC increases with increasing concentrations of chlorides and oxygen in the temperature range of $250 \dots 300 \text{ }^\circ\text{C}$. In this case, chlorides have a stronger effect on CC than oxygen.

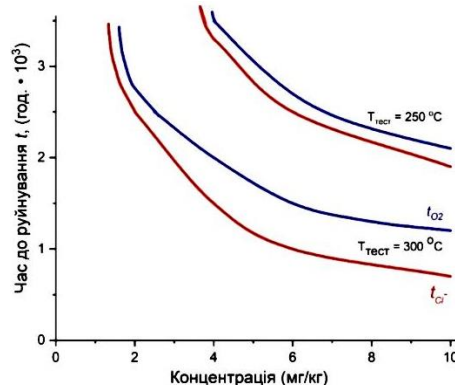


Fig. 2. Effect of C_{Cl^-} or C_{O_2} concentrations on the time to failure of HTT (calculated values) [14, 53]

Also, this steel exhibits an increase in the sensitivity to CC with increasing temperature. Metallographic studies have shown that the development of cracks in Kh18N10T steel has a pronounced mixed fracture pattern. The calculated data were confirmed in experimental studies.

Based on the analysis of the dependencies it can be concluded that the purer the water in terms of chlorides, the higher the oxygen content, the CC begins, and the less oxygen, the higher the concentration of chlorides is permissible. At the same time, the concentration of chlorides during SG operation should be more strictly controlled relative to other characteristics. It follows from the analysis of the data that at normalized concentrations of chlorides and oxygen in the SG water, delayed CC should be observed in Kh18N10T steel.

3.2. FORMATION OF CORROSION PITS DUE TO THE PRESENCE OF COPPER IN SEDIMENTS

Copper and its alloys tend to form complex compounds with ammonia, which is used to raise the pH. Good solubility in such water ensures the transfer of copper ions into the SG volume, where the reduction of metallic copper on the HTT surface takes place. Considering that there is practically no oxygen in the operating SG when water is boiling, in this case copper ions are the strongest oxidant. In the reduction of copper on the surface, the HTT creates a system of a steel-copper microgalvanic cell, in which copper is the cathode, and steel is presented as the anode. During the operation of such a galvanic couple, the steel dissolves, which contributes to the formation of a corrosion pit. It should be mentioned that the accumulation of metallic copper on the HTT surface occurs because of insufficient washing from copper during chemical washings of SG.

To estimate the rate of formation of corrosion pits due to the presence of copper in sediments, we will take the following statements as initial data [46-49]:

- The growth of corrosion pits occurs by the mechanism of anodic dissolution.
- The only possible oxidant in sediments is copper ions, in sufficient quantity.
- The electrochemical reaction is limited to the area of the corrosion pit.
- The surface area of sediments will be taken based on the channel diameter of 10 μm ($\sim 10^{-9} \text{ m}^2$).

Based on these postulates, it is possible to carry out an estimated calculation of the time to the initiation and growth rate of a corrosion pit. Volume (V_R) of a hemispherical corrosion pit with a radius $r = 0.5 \text{ mm}$:

$$V_R = 4\pi r^3 / 6 = 2.617 \cdot 10^{-4} [\text{cm}^3]. \quad (10)$$

In this case, the amount of electricity (Q_p) required to dissolve the metal from a corrosion pit of this volume is:

$$Q_p = I_p \cdot t_p = V_R \cdot \rho_{st} \cdot z \cdot F / M = 2 \cdot 10^{-3} [\text{A} \cdot \text{h}], \quad (11)$$

where I_p is the average current of metal dissolution in the corrosion pit, A ; t_p is the time required for the growth of a corrosion pit of a given size, h ; ρ_{st} is the specific gravity of steel 08Kh18N10T, we accept $\rho_{st} = 7.8 \text{ g/cm}^3$; z is the valency of dissolved metal ions, we take $z = 2$ for steel and copper. F – Faraday number – the amount of electricity required to dissolve 1 g equivalent of metal, $F = 26.8 (\text{A} \cdot \text{h})/\text{g}$; M is the weight of 1 g mole of the dissolved metal, for steel 08Kh18N10T we take $M_{Fe} = 56 \text{ g}$, for copper $M_{Cu} = 64 \text{ g}$.

The weight of copper G_{Cu} sedimented on the surface of the HTT from Cu^{2+} ions with the release of the required amount of electricity $Q_p = 2 \cdot 10^{-3} [\text{A} \cdot \text{h}]$ can be determined by the formula:

$$G_{Cu} = Q_p \cdot M_{Cu} / (z \cdot F) = 2,3 \cdot 10^{-3} [\mu\text{g}]. \quad (12)$$

In this case, the time t of the growth of a corrosion pit to a depth of 0.5 mm will be:

$$t_p = G_{Cu} \cdot n / g_{Cu} \approx 150000 [\text{h}], \quad (13)$$

where g_{Cu} is the value of the copper flux brought to 1 m^2 of the HTT surface in 1 h. Can be calculated using the formula:

$$g_{Cu} = C_{Cu} \cdot D_{\text{steam}} = 1250 [\mu\text{g} / \text{m}^2 \cdot \text{h}], \quad (14)$$

where D_{steam} is the specific steam capacity of PGV-1000, $D_{\text{steam}} = 250 \text{ kg/m}^2 \cdot \text{h}$; C_{Cu} is the maximum allowable value of copper in the SG blowdown water, according to regulatory guide, $C_{Cu} = 5 \mu\text{g/kg}$.

Consequently, the average growth rate of a corrosion pit at the initial stage of development will be:

$$V_p = h_p / t_p \approx 3 \cdot 10^{-6} [\text{mm/h}]. \quad (15)$$

It is obvious that at such a growth rate of a corrosion pit during SG operation (with the schedule of chemical flushing after ~ 35 thous and h), the maximum depth of the corrosion pit will not exceed 0.12 mm. It is likely that a corrosion pit of this size will not be detected by eddy current testing methods. Thus, under the accepted postulates, the presence of copper in sediments can only contribute to the formation of damage in the HTT.

In [7, 51], a somewhat different approach was proposed for the influence of copper on the development of corrosion processes in HTT SG. The approach is since the total corrosion current of the steel-copper pair is proportional to the mass of copper in sediments to the power of 2/3. Based on these ideas, it is possible to calculate the approximate time until the destruction of the HTT. The relative thinning of the HTT wall thickness on the date of measurement can be written $h_1(t)/h$, where $h_1(t)$ is the depth of the corrosion pit during operation (t) of the SG, can be measured by eddy current testing, or by other methods, we take $\approx 0.5 \text{ mm}$; T – thickness of the HTT wall, 1.5 mm. Consequently, the relative depth of the corrosion pit will grow in proportion to the relative specific contamination of HTT with copper to the power of 2/3 and can be written as:

$$h_1(t) / h = [M_{Cu}(t) / M_{Cu}^{\text{MAX}}]^{2/3}. \quad (16)$$

The relative specific contamination of HTT with copper can be measured experimentally or can be calculated using the assumptions described above. The value of the flow of copper brought to 1 m^2 of the HTT surface in 1 h. Can be calculated using the formula:

$$g_{Cu} = C_{Cu} \cdot D_{\text{steam}} = 750 [\mu\text{g}/(\text{m}^2 \cdot \text{h})], \quad (17)$$

where D_{steam} is the specific steam capacity of PGV-1000, $D_{\text{steam}} = 250 \text{ kg}/(\text{m}^2 \cdot \text{h})$; C_{Cu} is the maximum allowable value of copper in the SG blowdown water, according to regulatory guide, $C_{Cu} = 3 \mu\text{g/kg}$.

Therefore, for 25,000 h (t_H) of operation, the contamination of HTT with copper will be:

$$M_{Cu}(t) = g_{Cu} \cdot t_H = 18.75 [\text{g}/\text{m}^2]. \quad (18)$$

Experimental data [52] showed that for 25,000 h (t_H) of operation, the contamination of HTT with copper was 5 g/m^2 ; chemical washing was possibly carried out. For 50,000 h of SG operation, the measured concentration of copper in sediments at the HTT was 25 g/m^2 , the calculated concentration was 37.5 g/m^2 . From the analysis of the calculated data, it follows that in 50,000 h the depth of the corrosion pit can reach 0.94 mm, experimentally measured $\approx 0.5 \text{ mm}$. Consequently, the growth rate of the corrosion pit is $\approx 0.19 \cdot 10^{-4} \text{ mm/h}$, which is slightly higher than the calculated parameters presented earlier. In this case, the predicted value of the residual HTT resource can be $\sim 80,000 \text{ h}$. Thus, considering the estimation approach in the calculations, this representation does not strongly contradict the experimental data.

The analysis carried out allows us to conclude that the presence of copper in the sediments is a significant, but not a determining factor in the main chain of corrosion damage to the HTT.

3.3. HYDROGEN EMBRITTLEMENT

It should be noted that hydrogen embrittlement is a separate type of damage and does not apply to CC. The difference between them lies in the fact that CC occurs because of the destruction process at the anode of the metal, while hydrogen embrittlement occurs because of the formation of hydrogen at the cathode [53]. In many cases, the CC process and hydrogen embrittlement exist

simultaneously. In stress CC, hydrogen is usually responsible for the occurrence of cracks in the metal and their propagation, easily penetrating the metal in the form of atoms or ions. CC is accompanied by the formation of hydrogen, which is adsorbed by the metal, which can lead to embrittlement of the alloy. Some researchers believe that alloys are destroyed due to alternating processes: CC and hydrogen embrittlement [54]. Others believe that the main process depends on the type of alloy, some alloys are mainly subject to anodic corrosion and CC, and others to hydrogen embrittlement. In structural alloys, the absorbed hydrogen can be in three states: in the atomic state, it is dissolved in the lattice; in the molecular one, it is localized and is in irreversible traps (pores, vacancies, grain boundaries); hydrogenation of alloy components.

Based on the literature sources [55], the hydrogenation of austenitic steel in a SG occurs at low steam velocities, high thermal loads, and elevated metal temperatures. As a result of electrochemical reactions, the hydrogen atom released from water is adsorbed on the surface and then diffuses into the metal. In a metal, atomic hydrogen, getting into a micropore, combines into molecules. It should be noted that hydrogen penetrates the metal only in atomic form; molecular hydrogen cannot penetrate the metal, much less diffuse in it [47]. The more atomic hydrogen penetrates the metal and the more it is in molecular form. Also, atomic hydrogen can interact with alloy components, creating gaseous products and high pressure in irreversible traps. As a result, the mechanical properties are reduced and the probability of crack formation in the alloy increases. It has been experimentally determined that there is several times more hydrogen around the cracks than in the depth of the metal [33]. In the process of further hydrogenation, embrittlement of the metal is observed [47, 53–56]. The phenomenon of embrittlement in steels already manifests itself at hydrogen concentrations of 0.8 cm^3 per 100 g of metal under normal conditions.

An analysis of the structure of austenitic steel showed that, during hydrogenation, the high-angle boundaries remain free of hydrogen ions until the appearance of secondary phases [57]. Before they appear, the hydrogen ion is localized mainly on dislocations inside the grain and on non-metallic inclusions (Fig. 3,a). Only after the appearance of secondary phases the intensive hydrogen saturation of the grain boundaries is observed (see Fig. 3,b).

Based on this, it can be assumed that during long-term operation, diffusing carbon contributes to the formation of nanosized precipitates of the secondary phase at the grain boundaries, mainly carbides. Grain-boundary precipitates of secondary phases are traps for hydrogen and contribute to the creation of local internal stresses, which can ultimately lead to the formation of cracks. In this case, due to the formation of carbides, the protective surface layer of chromium is depleted. Under the action of an aggressive medium, anodic dissolution of the metal occurs at such boundaries, which can lead to intergranular brittle fracture of the metal during SCC.

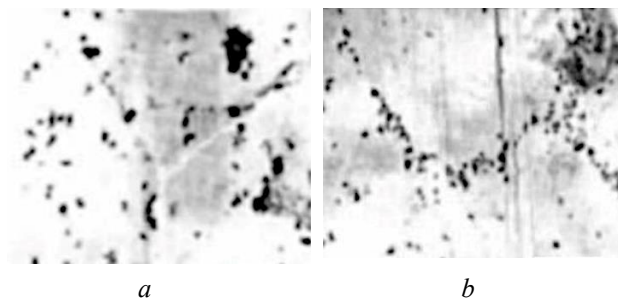


Fig. 3. The structure of austenitic steel after saturation with hydrogen [57]

The results of studies of the structure and mechanical properties of Kh18N10T steel after long-term operation [18, 20, 22] have shown that insignificant secondary phase precipitation is observed. Therefore, there will not be many accumulations of molecular hydrogen at the interfaces. In this case, slight changes in mechanical properties are observed, and the plastic properties of steel have undergone the greatest changes.

It is known that alloying with chromium, nickel, and titanium significantly reduces the hydrogen permeability of Kh18N10T steel [58–60]. In this case, the accumulation of hydrogen will occur at an insignificant rate and, as a result, during operation can only lead to additional tensile internal stresses in the steel. Therefore, it can be argued that, compared with the process of hydrogen embrittlement, CC is still the leading process of damage to HTT metal, but it is hydrogen embrittlement that determines the mechanism and kinetics of metal fracture under SCC conditions.

4. INFLUENCE OF LONG-TERM OPERATION ON THE METAL OF SG HEAT TRANSFER TUBES

It is known that the corrosion resistance of Kh18N10T austenitic stainless steel significantly depends on the heat treatment parameters, grain size, and residual stresses in the HTT metal. The presence of secondary phases in austenitic steel can significantly accelerate the rate of local corrosion initiation [57, 61–66]. It is known that, during plastic deformation during manufacture, welding, and installation in austenitic steels, a martensitic transformation of the type $\gamma \rightarrow \alpha$ or $\gamma \rightarrow \epsilon \rightarrow \alpha$ occurs. During annealing, the reverse transformation should occur, but, according to the data in the table, in Kh18N10T steel, such transformations do not proceed to the end. Studies, at room and operating temperature of the SG, of the mechanical properties of the initial and after long-term operation of the HTT showed overestimated values of the strength properties, while the elongation is at the level of the RG requirements. Nevertheless, the overestimated values of the HTT metal indicate the presence of residual stresses and structure heterogeneity in the HTT metal. At the same time, some researchers note that the corrosion resistance of Kh18N10T steel specimens after static deformation is higher than that of undeformed specimens [67, 68]. Based on this, further research is needed in this direction.

Table 4

Mechanical properties of HTT (steel Kh18N10T) before/after operation

Temperature test, °C	Defined parameter	Original metal	Long-term operation	Our standard*	Requirements PNAE-G-7-002-86
20	σ_b , MPa	745	715	545	510
	$\sigma_{0.2}$, MPa	451	431	290	216
	δ , %	53	53	53	35
300	σ_b , MPa	—	470	468	412
	$\sigma_{0.2}$, MPa	—	363	230	176
	δ , %	—	49	31	26

*HT: annealing 1020 °C / 0.5 h; stabilization 720...800 °C/1...3 h.

The microstructure of the steel samples is formed from equiaxed austenite grains with the precipitation of carbides along the grain boundaries. As a rule, in austenitic steels, precipitation of chromium carbides along the grain boundaries leads to intergranular corrosion. It is known that with a decrease in the grain size, the corrosion resistance of Kh18N10T steel increases. After annealing 800 °C/10 h, an increase in the density of secondary titanium carbonitrides is noted, both at the boundaries and inside the grains (Fig. 4). Some authors note that TiC inclusions (up to 1 μ m) do not significantly affect the corrosion resistance of Kh18N10T steel [63, 65]. The formation of stable TiC solid phases retains Cr in the solid solution, which contributes to the protection of the metal from ICC and possibly leads to dispersion strengthening of the metal

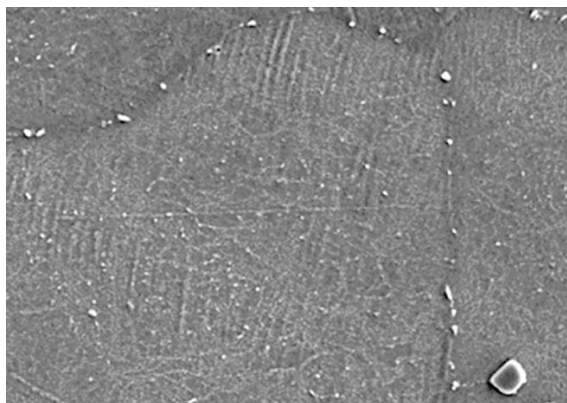


Fig. 4. Precipitation of carbides in the body and along grain boundaries [69]

CONCLUSIONS

1. The duration of SG HTT operation is determined by the simultaneous development of several damaging processes. Based on the data analysis, the main influence is exerted by the processes associated with an increase in the concentrations of chloride ion, oxygen, hydrogen, a decrease in the pH values of water, and the presence of internal and external stresses in the HTT. Additionally, the concentration of copper affects the rate of formation of corrosion pits.

2. Based on the analysis of the data, an attempt was made to estimate the service life of HTT made of steel 08Kh18N10T depending on the concentration of chloride ion, oxygen concentration, operating temperature of the metal, and the magnitude of tensile stresses. Calculations showed that the HTT resource decreases by an order of magnitude with an increase in

base. In contrast to TiC, the presence of a secondary TiN phase 5...7 μ m in Kh18N10T stainless steel contributes to the formation of pitting corrosion due to possible galvanic coupling between TiN and the metal base [69]. Microvoids are observed around TiN inclusions, which are mainly found in undeformed samples with an increased grain size [18, 20, 69-72]. Based on this, until the microvoid between the TiN particle and the matrix surrounding it is filled with an aggressive medium and a galvanic connection is not established, the metal will not dissolve, and corrosion pits will not develop. Thus, it can be assumed that the formation of stable TiC solid phases (dispersion hardening of the metal base) and the presence of non-metallic TiN inclusions can affect the rate of pitting corrosion initiation and the development of transgranular CC.

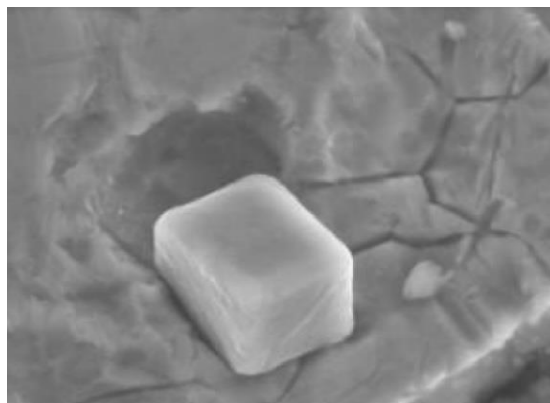


Fig. 5. Microvoid around non-metallic inclusion TiN [20]

the chloride ion concentration and the presence of oxygen. Based on the calculation results, it can be argued that chloride ions have a stronger effect on the HTT resource than oxygen. Based on this, the concentration of chloride ions during SG operation should be more strictly controlled in relation to other operational characteristics. In this case, it becomes possible to increase the HTT resource.

3. The main reason for the appearance and development of damage to HTT is the process of CC. Now data analysis allows us to draw only general conclusions on this problem. Calculation estimates show that if the water chemistry standards are observed (according to regulatory guide), the process of CC of Kh18N10T steel will proceed at a slow pace.

4. In our opinion, the calculation of time between failures is not entirely correct. Since in austenitic steel after 200 thousand hours of operation there are no

significant changes in the structure and mechanical properties. There is local CC of HTT. Therefore, it is necessary to pay attention to improving the “purity” of austenitic steel and to comply with the requirements of water chemistry standards more strictly.

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ОГЛЯД: ДОСЛІДЖЕННЯ ПРОЦЕСІВ ПОШКОДЖЕННЯ ТЕПЛООБМІННИХ ТРУБ ПАРОГЕНЕРАТОРІВ ВВЕР-1000

О.Г. Руденко, С.В. Гоженко

Проведено аналіз експлуатаційних пошкоджень теплообмінних труб у парогенераторах енергоблоків ВВЕР-1000. Розглянуто процеси забруднення поверхонь теплообмінних труб парогенераторів ПГВ-1000М. Проаналізовано процеси зародження та зростання корозійних виразок на поверхні теплообмінних труб. Запропоновано процес зародження корозійних виразок та розвитку тріщин у металі теплообмінних труб, який включає спільну дію корозійно-активного середовища та неметалевих включень у теплообмінних трубах.