Protection of Condensing Heat Exchange Surfaces of Boilers from Sulfuric Acid Corrosion

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Abstract. The method of metal protection of boiler condensing heat exchange surfaces can be successfully used in stationary and ship boilers, which burn fuel oils containing sulfur. The proposed method includes the operation of coating with a protective film against sulfur corrosion of the boiler heat exchange surface at a wall temperature below the dew point temperature of H_2SO_4 vapor. A passive layer of iron oxides is used as a protective film. It is obtained by passing physicochemical processes of passivation over the entire condensing surface from the beginning of sulfuric acid vapor condensation by pretreatment of exhaust gas flow with ionizing electron beams with a capacity of about 1 Mrad, ozone water-fuel emulsion combustion with a water content of about 30%. The metal surface is under the protection of a very thin passive film, which has a reliable connection with the metal at the level of the crystal structure and eliminates direct contact of the metal with the aggressive environment. The protective film constantly occurs naturally under the condition of creating an equimolar ratio of nitrogen oxides NO_2:NO (50:50)% in front of the condensing surface in the gas flow. The protection provides a significant increase in the boiler's efficiency (by 4 to 6%) when sulfur fuels combustion in their furnaces and deeper exhaust gases heat utilization in internal combustion engines and gas turbines (to 70%).

Keywords: Energy efficiency · Industrial innovation · Water-fuel emulsion · Exhaust gases · Boiler · Condensing heat exchange surface · Passivation

1 Introduction

The exhaust gas temperature largely determines the economic performance of auxiliary [1, 2] and exhaust gas boilers (EGB) [3, 4]. Its value is determined not only by the course of heat exchange processes in the elements of boilers, the requirements for their weight and size indicators, which is important for ship boilers but also by the intensity of thermochemical processes, which take place in the exhaust gas flow and on the heat exchange surfaces (HES) [5, 6] with a temperature below the dew point temperature (DPT) of H_2SO_4 vapor. The minimum value of HES temperature t_s determines the

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minimum exhaust gas temperature and, consequently, the economic indicators of their work. It is more difficult to reduce of exhaust gas temperature, since its value (about 160 °C) is determined by the rate of low-temperature corrosion (LTC) [7, 8], which sharply increases at $t_s = 130$ °C and reaches the level of the "corrosion peak" ($K =$ 1.2 mm/year) at $t_s = 110$ °C, reduces the work reliability of HES. Consequently, the thermochemical processes in the gas ducts of boilers and the LTC intensity significantly limit the possibilities of increasing the efficiency of the boiler and the depth of exhaust gases heat utilization of gas turbine (GT) [9, 10], gas engines (GE) [11, 12] and internal combustion engine (ICE) [13, 14].

Therefore, any measures to reduce the level of the "corrosion peak" to an acceptable level (about 0.2 mm/year) will provide reliable work of condensing HES to increase the boiler efficiency and fuel-saving [15, 16]. It is currently impossible to assess the influence of numerous factors on LTC intensity analytically. It is necessary to carry out experimental research of corrosion processes on condensing HES of boilers at t_s in the range of 60–150 °C and thermochemical processes which take place in exhaust gas flow before these surfaces [17].

2 Literature Review

Practically in all works devoted to studies of the H_2SO_4 formation in boilers, the mass flow of acid on the condensing HES, only the contact mechanism of the H_2SO_4 formation is considered. However, the possibility of the NOx influence on this process is also indicated. In [18], the author cites data that confirm the hypothesis of the possibility of interaction between SO_3 and NO with the formation of nitrose. However, work [19] does not provide quantitative estimates of the proposed reactions either from thermodynamic or experimental positions. The weighty circumstance can indicate that with a decrease of the gas temperature, the part of SO_3 is bound by nitrogen oxides, which seems to be confirmed by practice [19], which indicates a slight decrease of SO_3 concentration in exhaust gases.

In addition to the main process of H_2SO_4 formation by the contact mechanism in sulfuric acid production, the process of H_2SO_4 obtained by the nitrous mechanism is also used. The appropriate conditions are created: adsorbed sulfur dioxide SO_2 with nitrogen dioxide NO_2 in the presence of water is oxidized to sulfuric acid with the formation of nitrosylsulfuric and nitric acids and the release water. In this case, sulfuric acid's best absorption of nitrogen oxides occurs at an equimolar ratio of NO and NO_2 in exhaust gases [20]. Well-cooled (~100 °C) sulfuric acid, which contained 78% H_2SO_4, is used to obtain nitrose [21]. The same acid concentration is obtained during H_2SO_4 vapors condensation on boilers condensing HES, indicating the possibility of a nitrous mechanism for H_2SO_4 formation in the H_2SO_4 condensate on the condensing HESs.

The intensity of the acid and metal interaction is determined by the wall temperature and the acid concentration, the intensity of H_2SO_4 vapors mass flow to the surface, the protective properties of corrosion products, and the passive state of the metal, which can significantly limit the corrosion rate, despite the amount and concentration of acid [22, 23]. It should be noted that according to [24], the absorption of nitrogen oxides by a sulfuric acid solution improves the service conditions of steel equipment due to the passivation of the steel surface at a temperature of 70 °C. The passivation process lasts 12–20 h.

Exhaust gases of power boilers consist of 5% of NO_2 in NO_x [25], of auxiliary boilers - 12% [26, 27]. With an increase of SO_2 concentration in exhaust gases, the absorption rate of SO_2 by nitrose increases. It also increases with an increase of O_2 concentration in the gases since the rate of O_2 absorption by nitrose increases, enhancing NO to NO_2 in the liquid phase [28, 29].

During radiolysis of a gas flow with an increase in the water vapor content in exhaust gases content at the same radiolysis energy, a faster increase of NO_2 content occurs. For obtaining an equimolar mixture is required not very high radiation energy (about 1 Mrad) [30]. It can be assumed that similar ionic reactions can occur in exhaust gases under the influence of acoustic waves energy arising under the influence of "microexplosions" of the water-fuel emulsion (WFE). Adsorption processes occurring in the pollution layer must necessarily affect the chemical processes in the layer and the concentration of toxic ingredients in exhaust gases at the exit from the boiler.

The presence of nitrogen oxides in a sulfuric acid solution should lead to the appearance of the passivation process of the metal surface. In the theory of passivation, an important role is played by adsorption O_2 and oxide layers formation, which form a passivating protective layer. The O_2 source involved in the formation of passivating layers may be HNO_3.

$$SO_2 + NO_2 + H_2O \rightarrow H_2SO_4 + NO$$

$$2NO + O_2 \rightarrow 2NO_2$$

$$H_2SO_4 + 2NO_2 \rightarrow HNSO_5 + HNO_3$$

$$NO + NO_2 \rightarrow N_2O_3$$

$$2H_2SO_4 + N_2O_3 \rightarrow 2HNSO_5 + H_2O$$

Anions that form insoluble salts with metal or oxides can facilitate passivation. The appearance of a salt layer on the surface of iron may be preceded by the oxide passivation of iron in acidic solutions. In particular, this applies to H_2SO_4 solutions, where $FeSO_4$ and $Fe_2(SO_4)_3$ layers are formed, i.e., in the presence of Fe^{3+} in solution, reducing the critical passivation current.

$$Fe + H_2SO_4 \rightarrow FeSO_4 + H_2$$

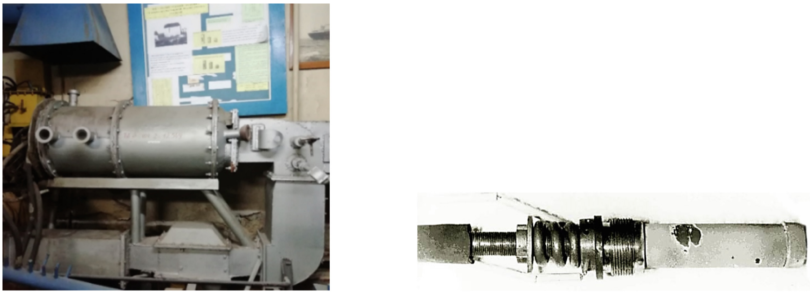
$$2Fe + 6H_2SO_4 \rightarrow Fe_2(SO_4)_3 + 6H_2O + 3SO_2$$

To confirm a significant reduction of LTC intensity in connection with creating a passive protective layer, special studies of this corrosion rate and the processes that confirm this phenomenon of passivation were conducted.

The research aims to confirm the passivation of metal on boiler condensing HES when WFE combustion.

3 Research Methodology

Studies of LTC intensity were carried out when fuel oils and WFE with a water content of 10, 15, 20, 30% were burnt. The research was carried out at a special experimental setup [29], where it is possible to ensure the stability of all parameters. This increases the reliability of the obtained results. The general view of the experimental setup and tube sample are presented in Fig. 1.



(a) (b)

Fig. 1. General view of the experimental setup (a) and tube sample (b).

In addition to determining the corrosion rate, analyzes of deposits composition were performed to determine the content of nitrogen oxides (in terms of HNO_3) and ions Fe^{3+}. Their content is largely defining the passivation possibility of the metal surface.

The nitrous mechanism for producing sulfuric acid is based on the absorption of SO_2 and NO_x. Therefore, it is possible to confirm the presence of this process in pollution on condensing HES when WFE combustion. For this, it is necessary to determine the change in the content of the SO_x and NO_x in exhaust gases before and after these HES with an increase of water content in WFE.

Taking into account the data of direct measurements of nitrogen oxides content and the relative content of Fe^{3+} in pollutions, experimental data on the intensity of LTC when WFE combustion [29], it can be argued that in this case, there is an additional passage of the nitrous mechanism of sulfuric acid formation in pollutions at the condensing HES at temperatures below the DPT of H_2SO_4 vapor. In addition, direct measurements of the content of SO_2, NO, and NO_2 in exhaust gases when WFE combustion and the obtained ratio NO_2:NO will give reasons to assert the possibility of a nitrous process in condensate on these HES.

4 Results

The dependences of the exhaust gas composition before the condensing HES, the corrosion rate, and the processes confirming the creation of the passivity of the metal surface were obtained.

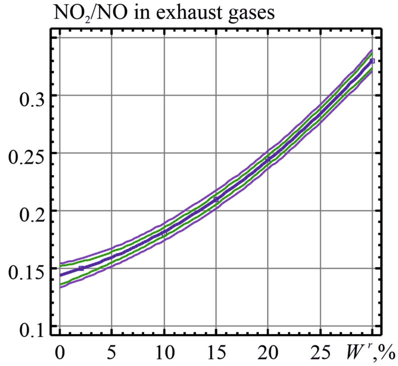


Fig. 2. Dependence of nitrogen oxides NO_2:NO ratio in exhaust gases before HES on water content W^r in the emulsion.

An analysis of exhaust gas composition showed (Fig. 2) that when the water content of emulsion W^r is increased, the NO_2:NO ratio approaches the equimolar mixture.

The polynomial equation of nitrogen oxides NO_2:NO ratio in exhaust gases before condensing HES on water content W^r in the emulsion was determined ($R^2 = 0.9998$):

$$NO_2/NO = 0.1438 + 0.0026W^r + 0.00012(W^r)^2 \quad (1)$$

Figure 2 shows the calculated values for NO_2/NO with the prediction (violet line) and confidence intervals (green line).

To confirm the passivation of metal on boiler condensing surfaces, the dependence of content of absorbed NO_x (as calculated on HNO_3) in acid condensation on condensing HES from the metal surface temperature t_s was obtained (Fig. 3).

The level of content of NO_x in condensate on condensing HES corresponds to sulfuric acid production with a nitrose mechanism. In the technological scheme of this production, to reduce the corrosion of sulfuric acid coolers, the phenomenon of passivation of carbon steel metal surface is provided at the expense of absorbed nitrogen oxides. In the course of the research, the dependences of corrosion rate (Fig. 4a) and the Fe^{3+} content (Fig. 4b) from the temperature of metal HES t_s are obtained.

The polynomial equation of absorbed nitrogen oxides content in sulfuric acid condensate HNO_3/H_2SO_4 on HES temperature t_s (Fig. 3) was obtained ($R^2 = 0.9474$):

$$HNO_3/H_2SO_4 = 48.4257 - 0{,}8868t_s + 0{,}0041(t_s)^2 \quad (2)$$

The polynomial equation of corrosion rate K on HES temperature t_s (Fig. 4a), was determined ($R^2 = 0.9568$):

$$K = -2866.51 + 143.504t_s - 2.8298(t_s)^2 + 2.75 \cdot 10^{-2}(t_s)^3 - 1.3141 \cdot 10^{-4}(t_s)^4 + 2.476 \cdot 10^{-7}(t_s)^5 \quad (3)$$

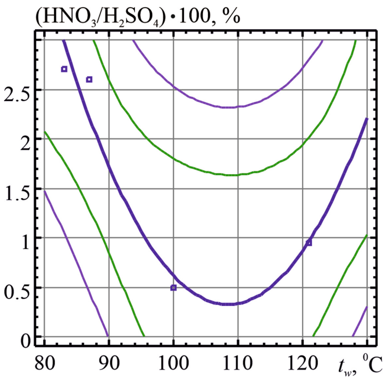
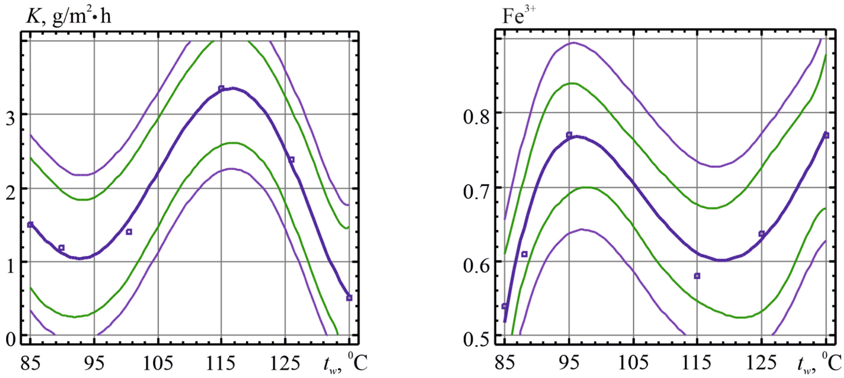


Fig. 3. Dependences of the content of absorbed NO_X in sulfuric acid condensate on HES temperature t_s.



(a) (b)

Fig. 4. Dependences of corrosion rate K (a) and Fe^{3+} content in sulfuric acid condensate (b) on HES temperature t_S.

The polynomial equation of Fe^{3+} content in sulfuric acid condensate on HES temperature t_s (Fig. 4b), was selected ($R^2 = 0.9113$):

$$Fe^{+3} = -106.059 + 3.6901 t_s - 4.7121 \cdot 10^{-2} (t_s)^2 + 2.6336 \cdot 10^{-4} (t_s)^3 - 5.4346 \cdot 10^{-7} (t_s)^4 \quad (4)$$

This equations are obtained for the following characteristics of nitrogen oxides NO_2:NO ratio, corrosion rate K and Fe^{3+} content: t_s = 85–135 °C and W^r = 10%. Figure 3, 4a,b shows the calculated values with the prediction (violet line) and confidence intervals (green line).

There is a minimum amount of Fe^{3+} at t_S = 110 °C ("peak" of LTC) and at t_S = 60 °C (second maximum). On the contrary, the maximum values of Fe^{3+} content are found at t_S = 80 °C and 130 °C, where there is a minimum corrosion rate. This is a sign of the growth of the passivation phenomenon of the metal surface. The obtained correlation of Fe^{3+} content in sulfuric acid condensate on condensing HES on W^r in the emulsion also confirms the significant increase of metal passivation (Fig. 5a). Comparing the dependences of Fe^{3+} amount (Fig. 5a) and corrosion rate (Fig. 5b) from W^r in emulsion at metal surface temperatures 110, 115, 120 °C is confirmed the indicated position.

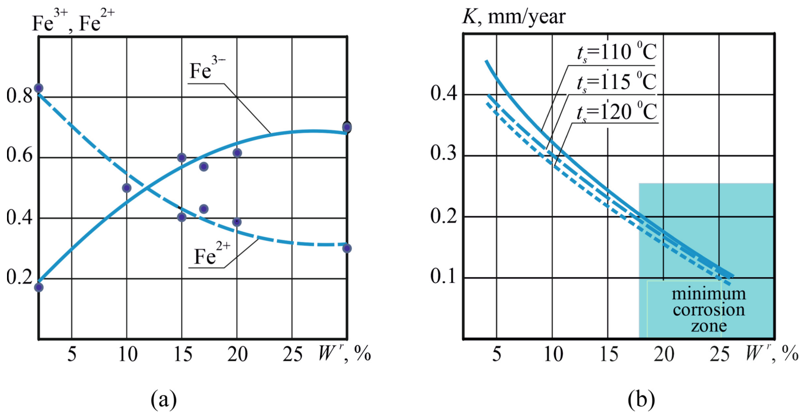


Fig. 5. Dependences of Fe^{3+}, Fe^{2+} content in sulfuric acid condensate (a) and corrosion rate K (b) on water content W^r in the emulsion.

The comparison of the dependence of corrosion rate on the water content W^r in WFE (Fig. 5b) and the dependence of Fe^{3+} content in the corrosion products on W^r (Fig. 5a) indicates that with increasing of water content in the emulsion W^r the corrosion rate decreases and reaches a minimum value at $W^r = 30\%$. The content of Fe^{3+}, which is a sign of passivation of metal surface, increases accordingly and reaches a maximum value at the same value $W^r = 30\%$. This means that the most passive state of the metal surface is achieved (Fe^{3+} – maximum, Fe^{2+} – minimum) when $W^r = 30\%$.

An integrated indicator of a sharp decrease of LTC intensity due to surface passivation is indirect measurements and obtained dependences of LTC on wall temperature (Fig. 6). The research results (Fig. 6) show when WFE is burnt with excess air factor $\alpha = 1.45$ the LTC intensity is at the level of 0.25 mm/year in the range of wall temperatures 70–130 °C in the absence of "corrosion peak".

Therefore, the main factor contributing to a significant reduction of LTC when WFE is burnt with a water content of WFE more than 20% (and especially at 30% water) is the occurrence of metal passivation.

The minimum corrosion rate values at a level of 0.25 mm/year is provided at the wall temperature up to 70 °C. Comparison of the results in Fig. 6 with data [18] at $\alpha = 1$–1.05 and water content of WFE W^r showed a reduction in the corrosion intensity with a decrease in α and increase of W^r and coincidence with curves with an accuracy of 10%.

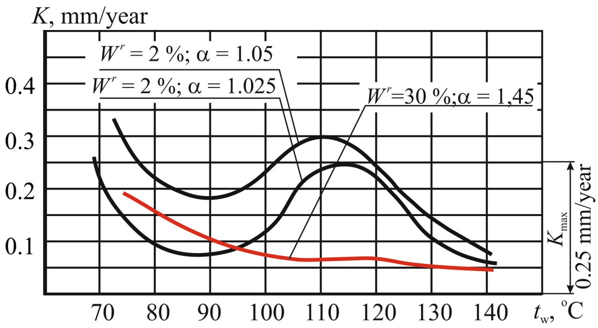


Fig. 6. Dependences of corrosion rate K on wall temperature.

5 Conclusions

This method of protecting metal from LTC provides resistance to thermal and dynamic deformations on the metal surface due to a very thin passive film with a thickness of about 50 A. This film has a reliable connection with the metal at the crystal structure and eliminates direct metal contact with an aggressive environment.

Improving the boiler efficiency in the protection of metal of condensing HES in this direction is achieved by increasing the stability and operation duration of the metal due to the constant automatic (natural) creation of a passive layer due to the occurrence of physicochemical processes in contact with the exhaust gas flow at metal surface temperatures below DPT of H_2SO_4 vapor (130 °C) and to 70 °C.

The wall temperature range of condensing HES safe operation is determined, revealing the opportunities for deep utilization. It makes it possible to reduce the temperature of the exhaust gases to 80 °C, thus significantly increasing the efficiency of boilers.

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