Untypical bromine corrosion in boilers co-firing biomass

A. Hernas [a,*], B. Chmiela [a], B. Szczucka-Lasota [b]

[a] Faculty of Materials Science and Metallurgy, Silesian University of Technology,
ul. Krasińskiego 8, 40-019 Katowice, Poland

[b] Higher School of Labor Protection Management,
ul. Bankowa 8, 40-007 Katowice, Poland

* Corresponding e-mail address: ahernas@polsl.pl

Properties

ABSTRACT

Purpose: The aim of this study was to determine the untypical corrosion resistance of rotary air preheaters in a biomass co-fired power plant.

Design/methodology/approach: The selected results of some components of regenerative rotary air preheaters in a biomass co-fired power plant are presented. The macro and microstructure of corroded components of a LUVO preheater after 3 years of service are presented. The chemical composition of corrosion products was determined by X-ray microanalysis.

Findings: Data concerning toxicity and identification of PBDEs (Polibrominated diphenyl ethers) in the biomass is very limited. The presented research provides information and the proposed corrosion reaction mechanism in environments containing biomass with aggressive compounds like Cl, Si, H and Br.

Research limitations/implications: An extended research on PBDEs in power plant fuels is necessary to give a full assessment of the corrosion mechanism in the presented environment; obligatory classification of the co-fired biomass.

Originality/value: Firstly, an assessment of the danger for boiler materials co-firing biomass containing brominated organic flame and presentation of the reaction during the corrosion process and degradation of power boiler components in the presented environment. Secondly, the highlighting of the problem that there is a need to properly select and determine the chemical composition of the biomass used.

Keywords: Bromine corrosion; Corrosion of heating surface; Corrosion in boilers co-firing biomass

1. Introduction

Directive No. 2009/28/EC [1] and political priorities of the European Union in the scope of the power sector dictate the "3x20" plan, i.e. that to the year 2020:

- the emission of greenhouse gases will be reduced by 20%,
- the energy consumption will be reduced by 20%,
- the share of renewable energy sources will increase by 20%.

The national legislation in the scope of energy from renewable sources, including the Action Plan of the Council of Ministers of 7th December 2010 and numerous ordinances of the Minister of Economy, also regulate activities in this area. One of the methods of implementing the above-mentioned plan is the firing and co-firing of biomass in boilers. The former consists in the oxidation of biomass with excess air and generating hot combustion gases for the production of steam or hot water in power boilers specially

designed to this end. The co-firing of biomass is connected with the replacement of a part of the basic fuel (hard coal or brown coal) in the operated boiler with an addition of biomass, usually in the amount of 5-10 mass %. On one hand, by reducing the emission of greenhouse gases, biomass co-firing is beneficial from the point of view of ecology and in terms of the economic calculation of a power plant. On the other hand, it causes a number of problems of technical and operational nature. The first group of problems is connected to the preparation of biomass and fuel mixture and its delivery to the boiler. The second group of problems concerns the need to properly supervise the technology of the combustion of fuel mixture with biomass and the occurring accelerated processes of the damage and shortening of the durability of heating surfaces of the boiler. Apart from basic damage processes, which depend on temperature, pressure, operation time and generally on the maintenance engineering of the boiler, the action of the increased aggressiveness of the environment, which contains Cl and S compounds as well as heavy alkaline metals, e.g. Na, K, Pb, Zn, Cd and other impurities, dependent on the composition and amount of the used biomass, also takes place. Recent experiments in this area indicate a possibility of the occurrence of damage which is also connected with the presence of bromine in the fuel mixture [2-4].

Biomass co-firing accelerates the degradation of boiler structural components, such as the firebox, steam superheaters, components of the end zone of convection surfaces, water heaters ^{o}I and air preheaters, which are heated by flue gases (Fig. 1) [3-7].
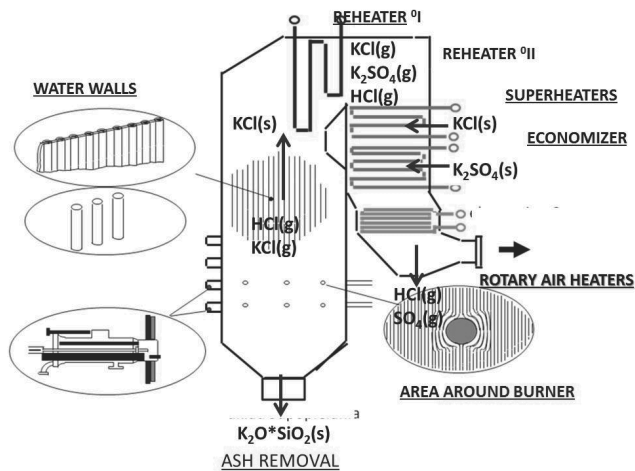

Fig. 1. Boiler components exposed to chlorosulphuric corrosion

2. Biomass co-firing

Biomass has been fired and co-fired in Polish energy sector since circa 2002. The basic types of biofuels are as follows: wood and wood waste (chips, bark, sawdust, furniture waste), plants cultivated for energy purposes (willow, mallow, polygonum, miscanthus), straw (wheat, rape, buckwheat, soya – straw contains the most chlorine), other agricultural waste (bagasse as well as rice, maize, sunflower and nut hulls), organic materials (municipal as well as pulp and water waste) and agri-food industry waste (meat and bone meal, manure). Biomass has variable physicochemical and technological characteristics (among others circa 40-50% lower calorific value than coal, high moisture content and low content of incombustible parts) which depend on the chemical composition, combustible mass and mineral substance as well as the content of volatile parts and ash. One of the most important problems connected with the operation of conventional biomass co-firing power boilers is the aggressiveness of the environment and the related corrosion processes [8]. The chemical composition of the ash from the biomass which after biomass firing contains SiO_2, CaO, K_2O and Na_2O, has a significant impact on operational problems. High meltability of alkaline oxides causes the biomass ash to melt at a much lower temperature (c. $800^{o}C$) than coal ash, which is the major cause of the forming of deposits covering heating surfaces of the boiler (Fig. 2).

Fig. 2. Deposits on superheaters of the second pass on one of outlet pipes of the convection cage

Corrosion processes

The primary damage process of heating components of boilers in which biomass is co-fired is the high-temperature corrosion connected with the presence of aggressive components of chlorine which is present in the deposits of the chlorides of alkali metals (K and Na), and suphur (Fig. 3).
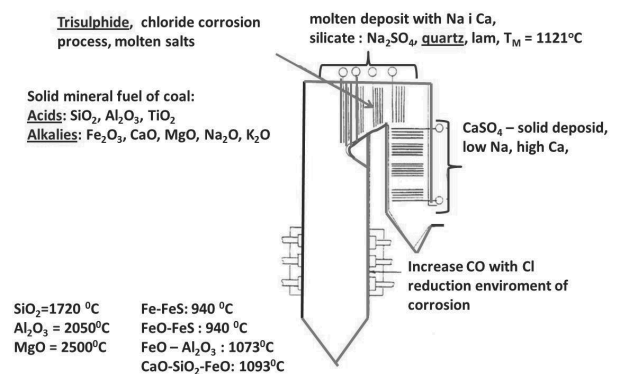

Fig. 3. Chlorosulphuric corrosion in biomass co-fired boilers

2.1. Chloride corrosion

The action of chlorides is particularly intense in a liquid phase due to the forming of low-melting eutectics, for example with heavy metals. The presence of the chlorides of alkali metals (K, Na) in deposits depends on the composition and type of biomass, the proportion between coal and biomass in fuel and on the construction of the boiler [9-12].

Chlorine corrosion starts as a result of HCl oxidation in accordance with the reaction (1):

$$2HCl + 0.5O_2 = Cl_2 + H_2O \tag{1}$$

or condensation reactions on pipes between potassium and sodium chlorides with iron oxides in the deposit (2):

$$2(K, Na)Cl + Fe_2O_3 + 0.5O_2 = (K, Na)_2Fe_2O_4 + Cl_2 \tag{2}$$
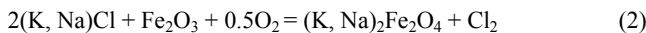
Molecular chlorine, released due to reactions (1) and (2), diffuses through deposits to the metal and reacts with it, in accordance with (3):

$$Fe + Cl_2 = FeCl_2 \tag{3}$$

Chlorides forming on the surface of the metal have high vapour pressure at the temperature of c. 500°C, which causes them to change into gas, making their identification difficult.

Chlorides diffuse through a magnetite layer, damage it and oxidise in oxygen-rich areas in accordance with the reactions (4) and (5).

$$2FeCl_2 + 3/2O_2 = Fe_2O_3 + 2Cl_2 \tag{4}$$

$$3FeCl_2 + 2O_2 = Fe_3O_4 + 3Cl_2 \tag{5}$$

The reconstructed iron oxide does not have any protective features because it is not a compact layer, and the oxidised surface is porous.

The fact that biomass ash usually melts at a much lower temperature than coal ash is also important from the point of view of the course of chlorine corrosion. Chlorine corrosion becomes more intensive when deposits containing potassium and sodium present on pipes are in a liquid phase. The melting point of potassium and sodium chlorides is high, but during biomass firing dangerous low-melting eutectics form with chlorides of other metals with much lower melting points. That is why during the co-firing of biomass containing chlorine, alkalis and heavy metal the risk of corrosion is present already at the temperature of 250^0C and includes for example the firebox walls. An example of chlorine corrosion of waterwall pipes of a biomass co-fired boiler is shown in Fig. 4.
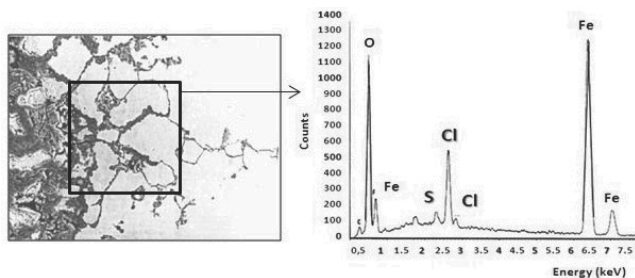

Fig. 4. Chloride corrosion at the boundaries of ferrite grains and EDS of waterwall pipes

It can be stated that KCl and NaCl intensify corrosion when they occur at the surface of a metal in the solid state.

2.2. The share of chlorine and sulphur

If there is sulphur in the system, a conversion of chlorides into sulphates also takes place: K_2SO_4 and $NaSO_4$ in accordance with the reaction (6):

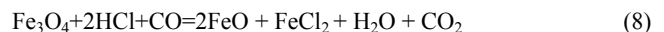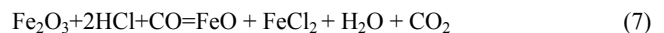$$2(K, Na)Cl + SO_2 + 0.5O_2 = (K, Na)_2SO_2 + Cl_2 \tag{6}$$
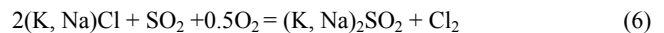
At the temperature of c. 500°C, sulphates are still stable and less corrosive than chlorides. Therefore, chlorine released according to the reaction (6) is formed on the deposit surface and flows to combustion gases. Only its small fraction penetrates into the deposit. Sulphurisation of chlorides occurs significantly faster with the participation of SO_3 than with the participation of SO_2.

The action of sulphur, which reduces the chloride corrosion, consists in the replacement of KCl and NaCl chlorides in the deposits with more stable sulphates. In theory, it is assumed that with the mole ratio between sulphur and chloride of more than 2.2 chloride corrosion ceases to be dangerous. According to literature data, in practice this does not apply to all combustion systems and the mechanism itself depends also on other factors.
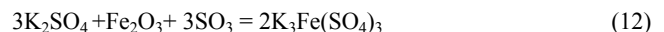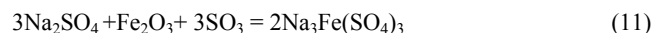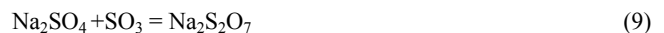
Co-firing of biomass containing chlorine and coal

In the case of co-firing biomass with fossil fuel (coal), the decomposition of HCl can be transcribed a follows:

$$Fe_2O_3 + 2HCl + CO = FeO + FeCl_2 + H_2O + CO_2 \tag{7}$$

$$Fe_3O_4 + 2HCl + CO = 2FeO + FeCl_2 + H_2O + CO_2 \tag{8}$$

2.3. Sulphuric corrosion

The other type of corrosion in the discussed boiler co-firing biomass is sulphuric corrosion [11]. Na_2SO_4 and K_2SO_4, which condense on the surface of pipes, are not directly aggressive due to high melting point. Na_2SO_4 melts at 884°C and K_2SO_4 at 1069°C.

However, pyrosulphates forming in the presence of SO_3 according to reactions (9) and (10) and trisulphates forming in accordance with reactions (11) and (12) are aggressive in relation to the metal.

$$Na_2SO_4 + SO_3 = Na_2S_2O_7 \tag{9}$$

$$K_2SO_4 + SO_3 = K_2S_2O_7 \tag{10}$$

$$3Na_2SO_4 + Fe_2O_3 + 3SO_3 = 2Na_3Fe(SO_4)_3 \tag{11}$$

$$3K_2SO_4 + Fe_2O_3 + 3SO_3 = 2K_3Fe(SO_4)_3 \tag{12}$$

The melting point of $Na_2S_2O_7$-type pyrosulphates is 389°C and of $K_2S_2O_7$-type pyrosulphates – 404°C. The damaging corrosion action of pyrosulphates also consists in the degradation of the protective oxide layer of a steamer, in accordance with (13):

$$3(K, Na)_2S_2O_7 + Fe_2O_3 = 2(K, Na)_3Fe(SO_4)_3 \tag{13}$$
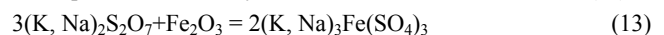
An example of a distribution of sulphides on boiler waterwall pipes is shown in Fig. 5.
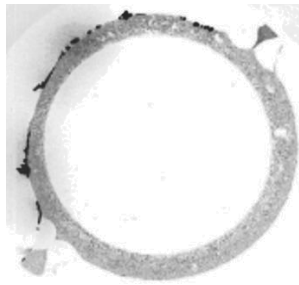
A. Hernas, B. Chmiela, B. Szczucka-Lasota

Fig. 5. The distribution of sulphides (Bauman's test) on boiler waterwall pipes on the side of the action of combustion gases

3. The major causes of damage of biomass co-fired boilers

During the exploitation of biomass, a dynamically changing composition of "fuel", its calorific value, consistence and physicochemical parameters influence the instability of firing, which also leads to the faster degradation of the installation (Fig. 6) [13-18].
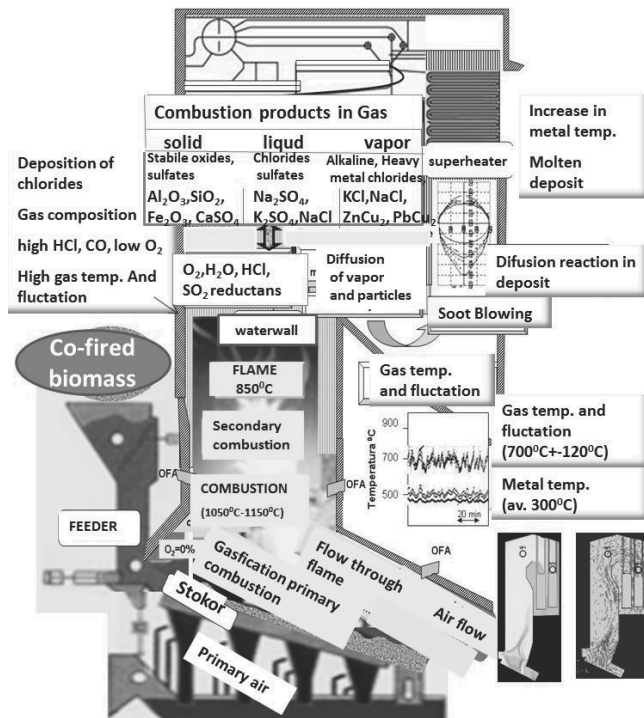

Fig. 6. Degradation process in biomass co-fired power boilers

During the process of the firing of biomass, among others volatile substances of the following types are formed: KCl, $NaCl$, $ZnCu_2$, $PbCl_2$ and melted substances and compounds such as Na_2SO_4, K_2SO_4 or $NaCl$. Dangerous low-melting eutectics are also formed, which lead to slagging and consequently to the increase of the corrosive wear of boiler components and the pipes of steam superheaters (Fig. 6). Biomass ash is often already liquid at the temperature of c. 800°C because the presence of the following types of fusible oxides can be detected in the chemical composition of biomass: CaO, K_2O, Na_2O [13-18].

A solid deposit containing the compounds of oxides and sulphides, e.g. Al_2O_3, SiO_2, Fe_2O_3 and $CaSO_4$, accumulates on the pipe walls. Percentage values of individual substances in the biomass may differ considerably. Fluctuations, trajectory and the type of particles (in particular of the hard ones) affect erosive processes, but this damage mechanism is less frequent [17-20].

In conclusion, the composition and type of the used biomass, apart from chlorosuphuric corrosion, may adversely affect the operation of the boiler causing[13-20]:

- bed defluidisation (ash sintering and agglomeration),
- the forming of deposits on convection surfaces,
- the processes of fouling and slagging resulting from a high content of alkalis and a lowered melting point of ashes,
- erosion caused by the presence of hard particles in some types of biomass, e.g. quartz.

Consequently, this may lead to the decrease in boiler efficiency, dedusting in electrostatic precipitators and fire hazards.

Biomass fuel poses a potentially lower risk of danger to the environment than the majority of coals due to the low content of sulphur (to c. 0.05%), nitrogen (c. 0.3%) and incombustible parts (0.5-3%) [13-17]. Unfortunately, biomass often contains production and post-production waste, e.g. from the furniture industry: wood containing paint, lacquers, antipyrines or formaldehyde glues. This may cause the forming of dangerous compounds containing traces of mercury, arsenic, copper or chromium after the combustion process. However, the process of the combustion of such materials may lead to other corrosion mechanisms that cause damage to boilers than those already described in the literature. A diversified chemical composition of biomass, the moisture content and the presence of chemical compounds of the POP-type (Persistent Organic Pollutants) may make the course of corrosion processes unusual. One example is an untypical bromine corrosion whose results are presented later in this paper.

4. Scope

In the initial period of the implementation of biomass co-firing in two domestic power plants, an accelerated corrosive wear of sheets and heating baskets of rotary air preheaters was observed.

This applied in particular to the so-called "cold end" of the baskets where the condensation of combustion gas takes place due to reaching the "dew point" temperature (Fig. 7).

4.1. Purpose of research

The purpose of the research conducted by the authors was to analyse the results and causes of the premature intensive corrosive wear of the sheets of baskets of rotary air preheaters.
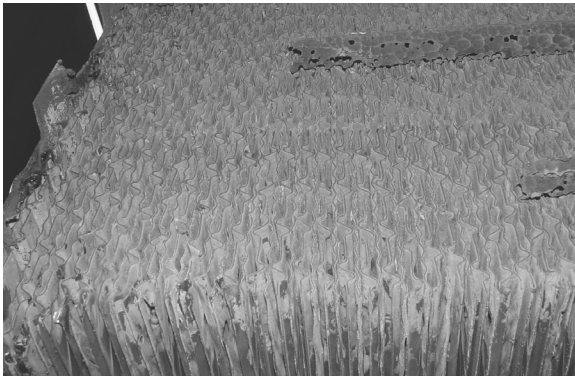
Fig. 7. The view of the sheet pack of the heating basket in the area of the "cold end". Destroyed flat stiffening sheets and the corrosion of lower edges of the pack

4.2. Materials for research

The materials for research comprised the sections of sheets made of CORTEN-type rust-resistant steels, collected from the "cold end" after c. 3 years of the operation of the facility where the co-firing of coal with biomass took place [5, 7]. Macro- and microstructure research with a particular emphasis on X-ray microanalysis of the chemical composition of corrosion products was conducted.

5. Research results

Example macro photographs of corrosively damaged sheets of the "cold end" are shown in Figs. 8a and b.

Corrosion-related changes were observed along the length sheets from non-damaged areas of the "hot end", through the transition area of pitting and uniform corrosion to the areas with weakly connected corrosion products and considerable material loss. Oxides on the surface from which external layers have broken away have complex, multi-layer structure and varied phase/chemical composition, which manifests itself in a strong contrast of those microregions in secondary electrons (white, light and dark microregions). Corrosion products are weakly connected to the substrate. Furthermore, separate particles with distinct phase boundaries, weakly connected and incorporated into the solid layers of iron oxides, are visible in those layers. A typical morphology of corrosion products outside the "cold end" area, in the uniform corrosion area, is shown in Fig. 9, and of pitting corrosion – in Fig. 10. An X-ray phase analysis of that area indicated the presence of Fe_3O_4 iron oxides and SiO_2 ash component.

A microanalysis of the chemical composition from the area not affected by intensive damage (as in Fig. 9) showed the presence of the following ash components: Si, Ca, K, Al, sulphur and selenium (Table 1). The X-ray phase analysis showed that apart from Fe3O4 iron oxide and SiO_2 ash components, there is FeSO4 iron sulphate present in corrosion products, which is connected to the porosity of oxides and accelerated corrosion (Fig. 11).
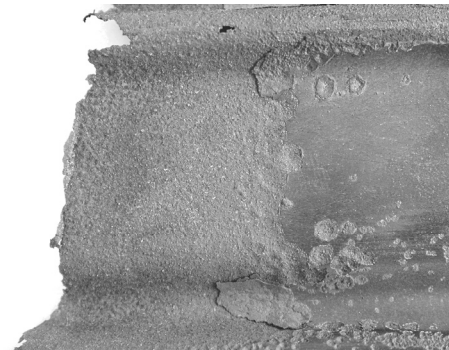
a)

b)

Fig. 8. a, b). Sections of the examined sheets of rotary air preheaters in the area of the "cold end"

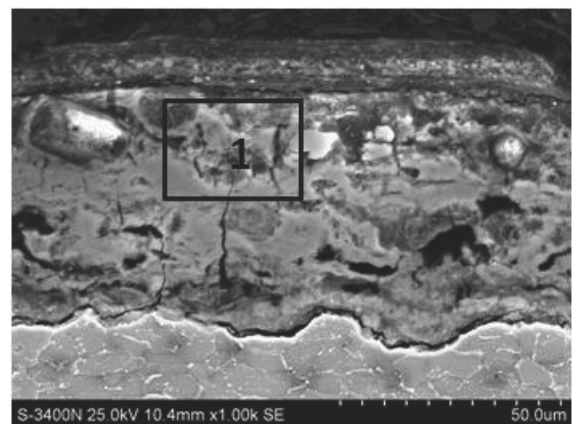Fig. 9. Microstructure of the surface layer of a flat sheet of the basket housing with corrosion products and deposits

Table 1.
Chemical composition of corrosion products from area 1, Fig. 9

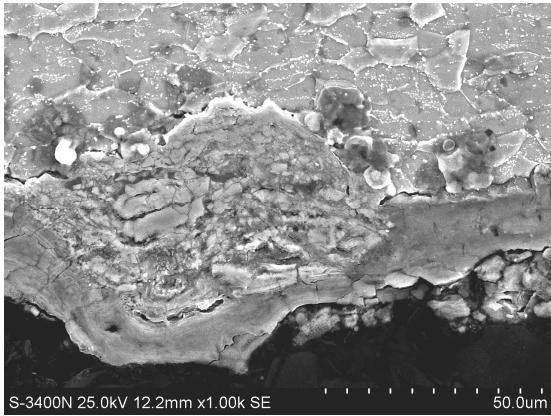|	Chemical composition (% mass.)						
Fig. 9:	Al-K	Si-K	S-K	K-K	Ca-K	Ti-K	Fe-K	Se-K
area 1	4.77	19.61	16.49	0.28	16.68	1.89	32.52	7.75

Fig. 10. An example of sub-deposit gutting corrosion

In the products of intensive corrosion, in particular from corrosion layers that loosely adhere to the substrate (Fig. 8b) in the area of considerable corrosion damages it revealed the presence of a relatively large quantity (about 20-25 mass %) of bromine (Fig. 12, Table 2) and chlorine (about 17-24%). Corrosion products formed on the basis of iron oxides have inhomogeneous structure. They contain separate particles, mainly ash-based SiO_2, which show high porosity and numerous cracks. Such morphology of corrosion products may be explained with intense action of chlorine and bromine (Figs. 12 and 13).

Both chlorine and bromine by diffusing through a layer of magnetite damage it in oxygen-rich areas. In accordance with (4) and (5), oxidation reactions make the reconstructed iron oxide porous and devoid of protective properties. Porous and uneven surface with a high degree of development is more exposed to degradation processes than a scale which constitutes a compact layer.
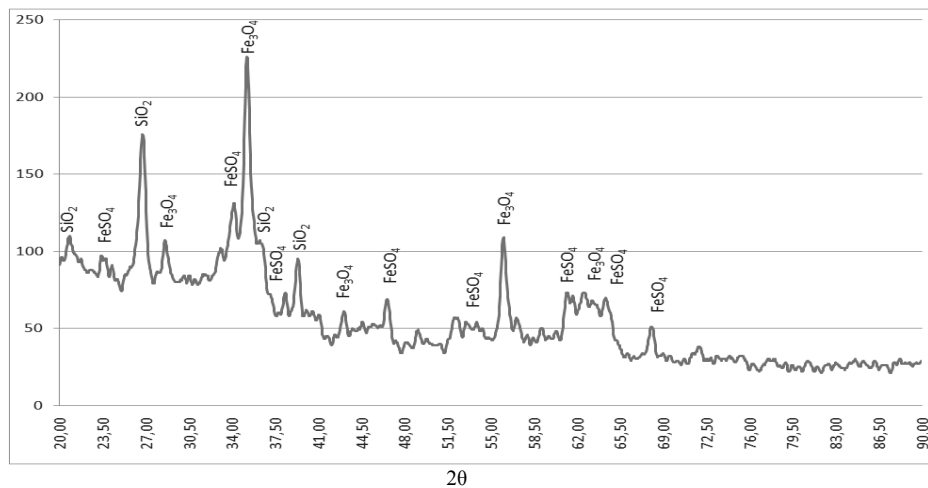

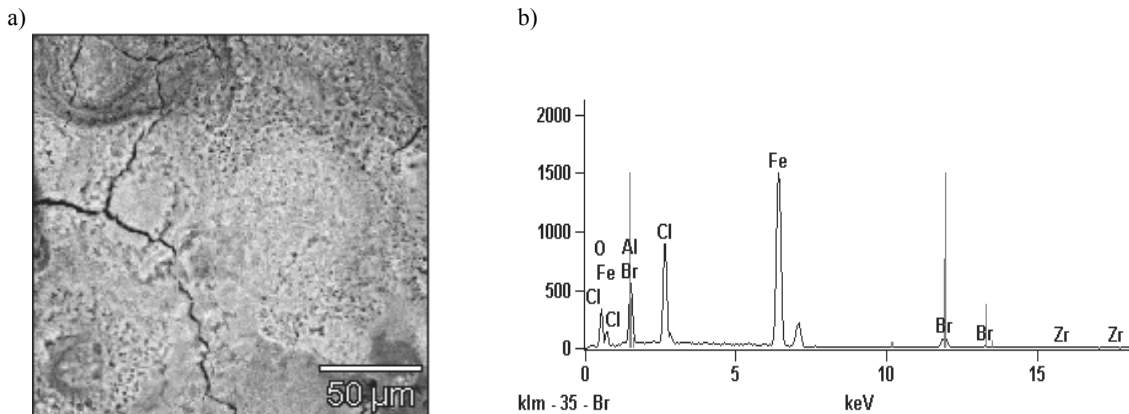Fig. 11. An X-ray phase analysis of corrosion products

a)

b)

Fig. 12. Morphology (a) and chemical composition (b) of corrosion products on the surface of rotary preheater materials

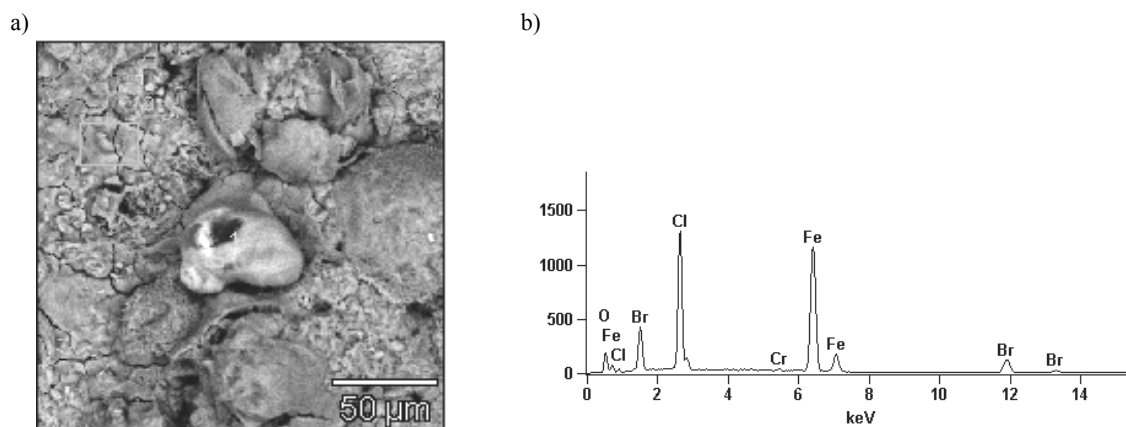a)   b)

Fig. 13. Morphology (a) and average chemical composition (b) of corrosion products on the surface of rotary preheater materials

An analysis of the chemical composition did not show the presence of sulphur compounds in corrosion products being formed (Tables 2 and 3). Therefore, it can be assumed that the mechanism described in chapter 2.2 does not occur.

Table 2.
Chemical composition of corrosion products on the surface of rotary air preheaters (LUVO)

Chemical composition (% wt.)					
Fig. 12:	Al-K	Cl-K	Fe-K	Br-K	Zr-L
area 1	0.12	16.89	62.56	20.28	0.05

Table 3.
Chemical composition of corrosion products on the surface of rotary air preheaters (LUVO)

Chemical composition (% wt.)					
Fig. 13:	Al-K	Cl-K	Cr-K	Fe-K	Cu-K	Br-K
area 1	-	24.65	0.44	49.79	-	25.11

The examined surface of rotary air preheaters is exposed to intense action of chlorine and bromine, which constitute the main cause of degradation. This thesis is highlighted by the fact that chlorine, which is difficult to identify when it is not the main cause of corrosion, has been identified in the examined material (Tables 2, 3). This is because chlorine to a large extent diffuses from the surface of oxides to the gaseous environment. For these reasons the X-ray phase analysis of corrosion products did not reveal the presence of chemical compounds, neither chlorine nor bromine.

Similar corrosion mechanisms, such as those with chlorine, take place in an environment that contains bromine (mainly in the form of hydrogen bromide), which intensifies damage processes.

6. Bromine corrosion

An analysis of the results described in chapter 6 makes it possible to ascertain the presence of the bromine corrosion,

which is one of the main causes of the premature destruction of rotary air preheaters of the LUVO type. The occurrence of the bromine corrosion in a conventional biomass co-fired power boiler is surprising and unusual. According to the authors, it is connected to the chemical composition of fuel, specifically to the presence of polybrominated diphenyl ethers (PBDEs) and bromine combustion retardants (Fig. 14).
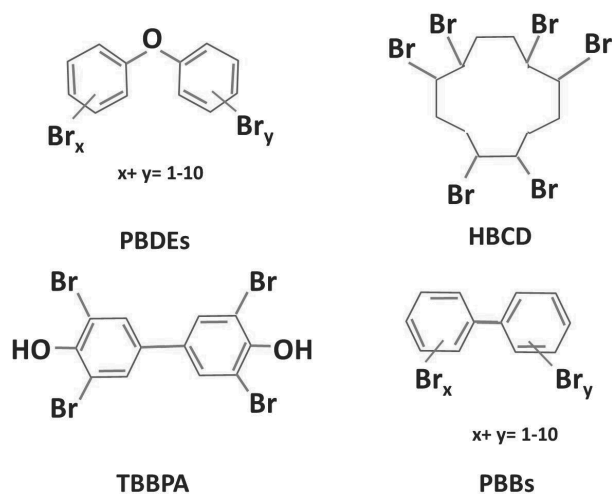

Fig. 14. Structural formula of brominated organic flame retardants: PBDEs (polibromated diphenyl ethers), (HBCD) hexabromocyclododecane, (TBBPA) tetrabrominated diphenyl-A, (PBBs) polibrominated bifhenyl [21]

The research results presented in the literature confirm the presence of polybrominated diphenyl ethers in the majority of the components of the aquatic and land environment, among others in the air, water, deposits, fish, birds, mammals, tissues (including human ones) and plants. Polybrominated diphenyl ethers (PBDEs) reach the environment due to the lack of chemical bonds with the matrix of the materials to which they are added. At the same time they show relatively high durability. Antipyrines used in scrapped furniture, chipboards, plywood and paint are also a source of

bromine in biomass. The content of PBDEs in paints, lacquers and polymers may reach even 30%. The analysis of the results of research presented in chapter 6 suggests that the main fraction of the biomass co-fired in the power boiler comprised furniture waste containing bromine combustion retardants [21].

During the co-firing of biomass in the power boiler, high temperature maintained thanks to basic combustible materials is conducive to the decomposition of antipyrines and the release of hydrogen bromide and bromide radicals (Br). These substances are very chemically active, which facilitates the occurrence of many reactions with other combustion products as well as organic and non-organic biomass components. For example, a reaction with the compounds of alkali metals and sulphur oxide (14) proceeds as follows:

$$4KBr + 2SO_2 + O_2 + H_2O \rightarrow 2K_2SO_4 + 4HBr \tag{14}$$

Additional quantities of hydrogen bromide (HBr), which causes intense steel corrosion, are formed in the reaction.

Chlorine corrosion starts as a result of HBr oxidation in accordance with the reaction (15):

$$2HBr + 0.5O_2 = Br_2 + H_2O \text{ [compare with formula 1]} \tag{15}$$
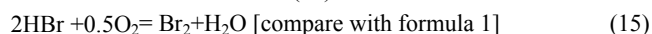
Bromine is also released in this reaction. The course of the corrosion mechanism is therefore similar to chlorine corrosion described in chapter 2.

7. Conclusions

The firing and co-firing of biomass is one of the ways to meet the requirements of the "renewable energy development strategy" through acquiring energy and reducing the harmful emission of greenhouse gases. As shown in the study, the co-firing of biomass entails a number of operational and technical problems. One of the main causes of degradation is the high-temperature corrosion, determined by the chemical composition of the biomass. The intensity of the degradation of conventional power units using alternative fuels, including biomass, depends on the type of the biomass, the quantitative ratios of individual components and, as demonstrated by the studies, on the chemical composition of the biomass.

A suitable chemical composition may slow down the corrosion processes, e.g. the presence of significant amounts of sulfur in the fuel enables avoiding the formation of chlorides in favour of the conversion of chlorides to sulphates. At the same time, the presence of other reactive substances, such as, for example, polibrominated diphenyl ethers, significantly enhances the corrosion. In spite of introduction of many limitations and prohibitions in using brominated organic flame retardants, the materials are in continuous use. They are in polymers, paints, varnishes and in the textile materials produced before the year 2007. They can be found in selected foodstuffs, so they are in the communal waste, too. PBDEs (polibrominated diphenyl ethers) have high persistence. The combusted fuel containing PBDEs accelerates the corrosion process in the power plant. PBDEs make the environment more aggressive. Chemical reactions with bromine lead to the destruction of power plant installations.

The selected research results presented here allow affirming that:

- The main factor behind the high-temperature corrosion of rotary air preheaters during combustion of biomass and coal is chlorine, due to the presence of alkali metal chlorides K and Na) in the deposits;
- In the analysed cases, the process of degradation of rotary air preheaters is intensified by the presence of bromine, whose sources are polybrominated diphenyl ethers present in the biomass. In the presence of oxides and water at an elevated and high temperature, hydrogen bromide, HBr, can be formed from KBr, which intensifies the corrosion process.

This allows formulating a conclusion that in order to ensure the safe operation of boilers in the case of co-firing of biomass and waste, it is necessary to determine the chemical composition of the alternative fuel used. As resulted from the analysis of the experiments carried out in the early stages of implementation of biomass co-firing, the selection of biomass was a bit random, with no objective documentation of the quality of the biomass used. The research results have led to the elimination of biomass without the relevant certificates and to admission to co-firing of qualified biomass only.

The current approach to reducing corrosion degradation of heatable boiler surfaces comes down mainly to monitoring the risk of corrosion in the following ways:

- Directly: through control of KCl share in the flue gas at the furnace outlet, neutralisation of KCl in the flue gas, examination of the flue gas composition at heatable surfaces;
- Indirectly: through examination of specimens for metallographic analysis, examination of the chemical composition of deposits and measurements of wall thickness;
- Through the application of protective coatings: this solution does not eliminate the problem and treats the effects instead of the causes.

Acknowledgements

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