

CO-COMBUSTION OF COAL AND OIL SHALE BLENDS IN CIRCULATING FLUIDIZED BED BOILERS

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Abstract. *Coal co-firing experiments were conducted in a 250 MW oil shale fired circulating fluidized bed combustion (CFBC) boiler. The objective of the experiments was to test whether adding coal to oil shale would allow the use of the latter with lower heating value. Bituminous coal was mixed with oil shale and fed into the boiler via existing fuel feeding ports. Two test series were accomplished: 11–29% thermal input of coal mixed with 8.4 MJ/kg oil shale (standard fuel), and 12–32% thermal input of coal mixed with 7.5 MJ/kg oil shale. During the experiments, which lasted in total for 15 days, ash samples were collected and flue gas analysis was performed. The boilers were able to continue work with all the fuel mixtures, but a significant increase of nitrogen oxides (NO_x) emissions and heat losses due to unburnt carbon in the bottom and fly ashes were observed. The heat losses can be reduced by upgrading the fuel preparation system, but NO_x emissions limit can be reached only with installation of an additional DeNO_x system. The ash chemical composition remained similar. Sulphur emissions stayed minimal, but a slight increase of carbon monoxide concentration was noticed. Coal co-firing is possible in oil shale CFBC boilers, but the coal must have low fuel nitrogen content and extra attention to the fuel preparation system has to be paid.*

Keywords: *circulating fluidized bed combustion, oil shale and coal blend, co-combustion, emissions.*

1. Introduction

Estonian oil shale (OS) industry has long-term experience in oil shale utilization. Today's industry has implemented circulating fluidized bed combustion (CFBC) technology, older pulverized combustion (PC) power units have been equipped with modern DeSO_x and DeNO_x equipment. Transition

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to the CFBC technology resolved problems with fouling of heating surfaces, and NO_x and SO_2 emission. CFBC boilers have demonstrated fuel flexibility – biomass and peat co-combustion decreased gaseous pollutants, particulate matter and ash emissions [1, 2]. The oil shale CFBC boiler has also been used for utilization of pyrolytic wastewater. Successful combustion tests were conducted, the NO_x and SO_2 emissions remained low [3].

The power industry is trying to decrease its environmental impact while still keeping in use old PC power units although it has been found that PC boilers have even a higher environmental impact during load cycling [4, 5]. Considering this, co-combustion of biomass in a CFBC unit offers a good alternative to impact reduction. A number of biomass co-firing examples are available [6–9], the biomass co-combustion experiments showed positive impact also at oil shale power plants [1]. The motivation for coal co-firing in oil shale boilers enables the use of oil shale with lower heating value in the existing boilers without major retrofits. Calorific value of oil shale depends on oil shale layers and mining locations. Therefore, oil shale beneficiation plants are needed [10].

The use of oil shale in Estonia is limited and is directed towards oil production. One reason for the use of coal in OS CFBC energy units is that power companies try to expand the portfolio of fuels and leave more OS for shale oil production.

Prior to industrial testing, the Department of Energy Technology, Tallinn University of Technology conducted a laboratory test on its own laboratory scale 60 kW CFB combustion test facility. The laboratory test gave the input to industrial tests and enabled choosing a proper methodology and paying attention to possible problems. Laboratory tests revealed there was a problem with nitrogen emissions.

To explore the possibilities of coal and oil shale co-combustion in a CFBC boiler oil shale was mixed with coal. At the second stage, oil shale with a slightly lower heating value was used. The results and conclusions of the experiments are summarised in this paper.

2. Experimental

2.1. The boiler

The firing tests of oil shale and coal blends were carried out in the CFBC double boiler energy unit. The electrical capacity of the energy unit was 215 MW_{el}. The main parameters of the power unit are given in Table 1.

Table 1. Main parameters of the power unit

| Parameter | Value |
|---|----------|
| Steam mass flow (primary/secondary), kg/s | 95/76 |
| Primary/reheat steam pressure, MPa | 12.7/2.4 |
| Primary/reheat steam temperature, °C | 535/535 |

During the tests, the analyses of fuel, ash and flue gas were carried out. The location of ports (1–8) for collecting ash samples is shown in Figure 1. Fuel samples were collected on a daily average basis. The ash was sampled from several extraction ports located in the furnace chamber, super-/reheater (SH, RH), economizer (ECO), air preheater (APH) and from all four fields of the electrostatic precipitator (ESP). Samples of fly ash for determining the mass division (total suspended particulates PM 10/2.5) were collected after ESP. The samples were used for determining the detailed chemical composition of ash.

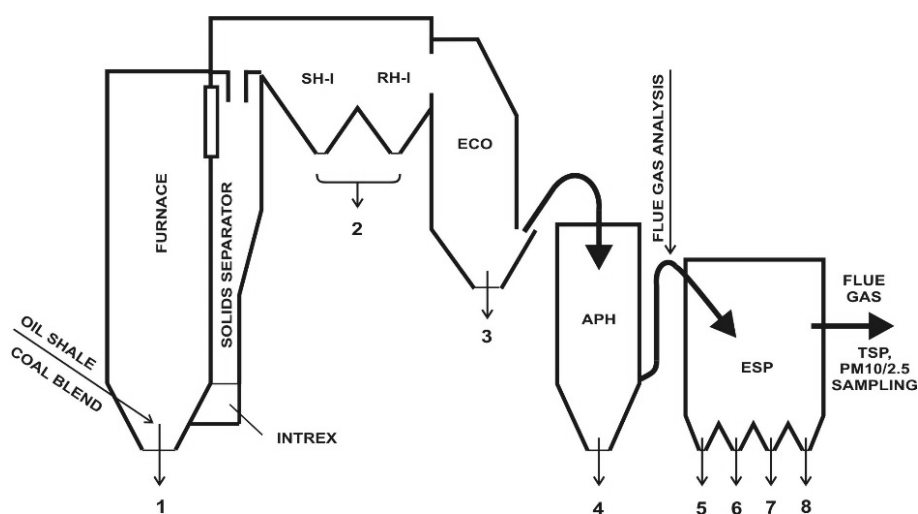


Fig. 1. CFBC boiler sampling points.

The results of analyses were averaged to reach a representative estimate. During the tests, the major process parameters of the boiler and energy unit were recorded with the plant data acquisition system. The temperature and composition of flue gas were measured before ESP. The composition of flue gas and the flue gas moisture content were determined applying a Fourier transform-infrared spectroscopy (FTIR)-type analyser for wet gas, at a temperature of 180 °C.

2.2. Fuels – oil shale and coal

Table 2 and Table 3 present the results of proximate and ultimate analysis of the fuels.

Carbon was present in both the organic and mineral matter of oil shale, the same applied to coal. During oil shale combustion the mineral $(CO_2)_m$ is released as a result of decomposition of carbonate minerals. The average extent of carbonate minerals decomposition was around 70%, varying strongly in different ash flows [11]. The main element in the oil shale ash was calcium. Calcium oxide content in the coal ash was considerably lower (Table 4).

Table 2. Proximate analysis of fuels

| Parameter | Symbol | Unit | Coal 19.7 MJ/kg | Oil shale 7.5 MJ/kg | Oil shale 8.4 MJ/kg |
|---------------------------------------|-----------|-------|--------------------|------------------------|------------------------|
| Lower heating value, as received fuel | Q_{i^r} | MJ/kg | 19.7 | 7.5 | 8.4 |
| Moisture content | W_r | wt% | 19.0 | 14.8 | 11.2 |
| Ash content | A_r | wt% | 12.2 | 43.8 | 43.7 |
| Volatiles | V_r | wt% | 30.9 | 39.6 | 40.2 |
| Fixed carbon | FC_r | wt% | 37.8 | 2.1 | 3.3 |

Table 3. Ultimate analysis of fuels, wt%

| Element | Coal 19.7 MJ/kg | Oil shale 7.5 MJ/kg | Oil shale 8.4 MJ/kg |
|------------------------------|--------------------|------------------------|------------------------|
| Carbon | 62.1 | 27.3 | 29.6 |
| Hydrogen | 4.04 | 2.74 | 2.79 |
| Nitrogen | 1.75 | 0.08 | 0.06 |
| Sulphur | 0.53 | 1.41 | 1.45 |
| Total inorganic carbon (TIC) | 0.29 | 5.16 | 5.62 |

Table 4. Chemical composition of fuels ashes, wt%

| Component as oxide | Oil shale 7.5 MJ/kg | Oil shale 8.4 MJ/kg | Coal 19.7 MJ/kg |
|--------------------------------|------------------------|------------------------|--------------------|
| Na ₂ O | 0.2 | 0.1 | 1.2 |
| MgO | 5.4 | 2.4 | 3.0 |
| Al ₂ O ₃ | 10.3 | 1.8 | 27.5 |
| SiO ₂ | 31.8 | 8.3 | 45.9 |
| P ₂ O ₅ | 0.2 | | 1.1 |
| SO ₃ | 3.9 | | 3.6 |
| K ₂ O | 3.8 | 0.6 | 2.3 |
| CaO | 39.7 | 54.5 | 8.0 |
| TiO ₂ | 0.5 | | 1.4 |
| MnO | 0.1 | | 0.1 |
| Fe ₂ O ₃ | 4.1 | 3.5 | 5.6 |

The results of thermogravimetric analysis (TGA) of coal and oil shale are shown in Figure 2. The figure reveals that the coal ignition occurred at a rather low temperature (< 200 °C) and the combustion was finished before the temperature reached 650 °C. No later char burning could be noticed, there was only a small mass loss due to decomposition of carbonate minerals. Based on the data no problems with fuel burnout were predicted.

The fuel mixtures for full-scale experiments (Table 5) were prepared on the fuel conveyor belt. The experiments lasted for 15 days. Each fuel mixture was fired for at least 24 h. During the experimental period, a large number of samples were collected.

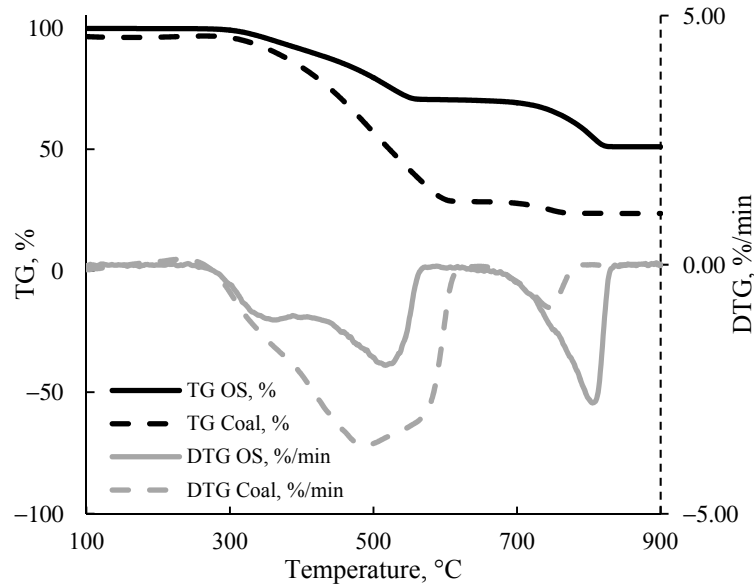


Fig. 2. Thermogravimetric analysis of oil shale (dashed line) and coal (black solid line) combustion in the air at a heating rate of 10 K/min.

Table 5. Fuel test matrix of co-combustion tests

| Fuel blend | Symbol | Wt% | % thermal input | Lower heating value, Q_{f}^r |
|-------------|--------|-------|-----------------|--------------------------------|
| OS 8.4 | A | 100 | 100 | 8.4 |
| OS 8.4/coal | B | 95/5 | 89/11 | 9.0* |
| OS 8.4/coal | C | 90/10 | 79/21 | 9.5* |
| OS 8.4/coal | D | 85/15 | 71/29 | 10.1* |
| OS 7.5/coal | E | 95/5 | 88/12 | 8.1* |
| OS 7.5/coal | F | 90/10 | 77/23 | 8.7* |
| OS 7.5/coal | G | 85/15 | 68/32 | 9.3* |

* LHV of fuel blend

3. Results and discussion

3.1. Temperatures of bed, furnace, separators and air preheater

Coal co-firing experiments were conducted in the oil shale fired industrial CFBC boiler. Overall the experiments were successful, no emergency shut downs occurred. The plant data acquisition system provided temperature readings from various locations. To illustrate changes in the CFBC boiler some chosen temperatures are shown in Figure 3. The temperature above the dense bed increased with the increased share of coal. In contrast, the flue gas temperature in the entrance of the solid separator and after the air preheater was stable. The average temperature values support this notice (Fig. 4). The increased temperatures may lead to the fouling and sintering of ash on heating surfaces, which were typical problems in oil shale PC boilers [9].

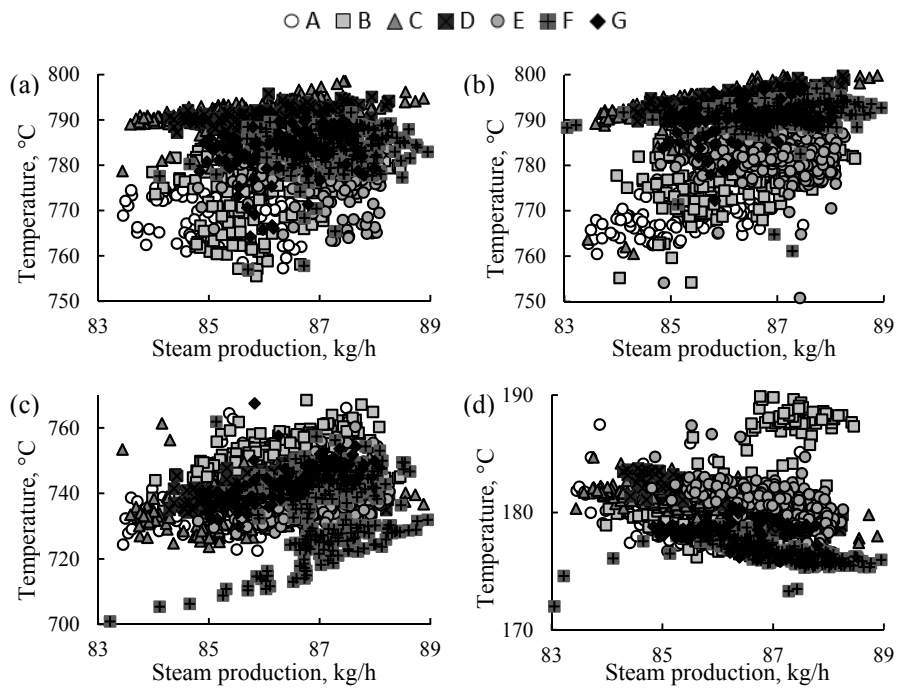


Fig. 3. The temperatures: a) in the bed nearby the front wall, b) in the furnace dilute zone, c) in the entrance of the solid separator, d) after the air preheater. The darker colour represents more coal in the fuel blend.

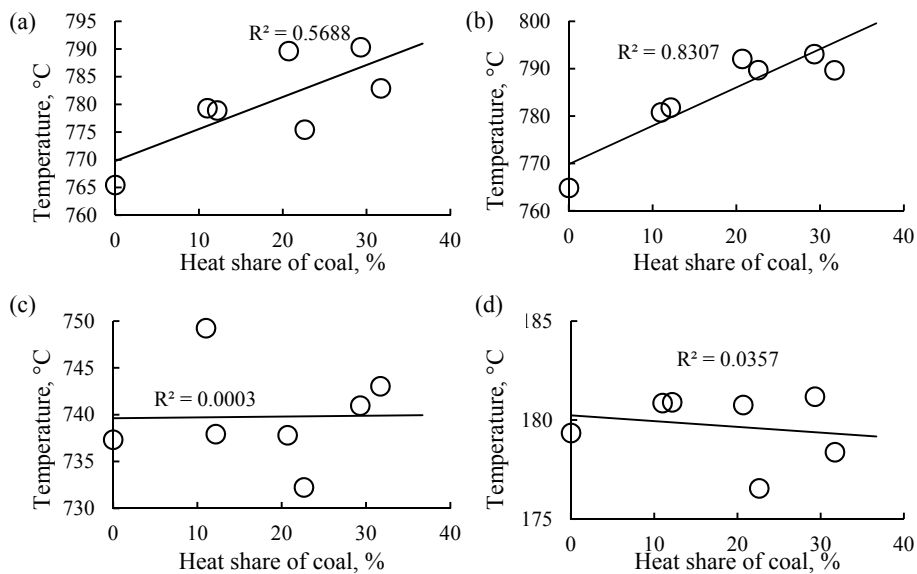


Fig. 4. The average temperature trends depending on the heat share of coal: a) in the bed nearby the front wall, b) in the furnace dilute zone, c) in the entrance of the solid separator, d) after the air preheater.

Experiments carried out by Parve et al. [12] showed that sintering of Estonian oil shale ashes became an issue when the temperature exceeded 900 °C. The temperatures measured in the boiler were well below the dangerous zone, but there were no measuring points in the centre of the boiler where the temperatures may be higher. Investigations carried out by Al-Otoom et al. [13] have shown that firing of Jordanian El-Lajjun oil shale would pose minimal operational problems related to sintering and agglomeration if the operating temperature is between 800 °C and 900 °C.

3.2. The chemical composition of ash samples

Typically, there is no problem with oil shale burnout, the organic carbon content in the boiler ashes is below 0.1% [1]. The results of chemical analysis of ash show relatively high levels of organic carbon, C_{org} (Table 6). When obtaining the ash samples, an unusual picture was taken of the bottom ash conveyor (Fig. 5). Orange glowing particles and black particles were seen. Unburnt particles were visible to the naked eye. This encouraged in depth fuel preparations control. As seen in Figure 6, oversized coal particles

Table 6. Chemical composition of ashes (85/15 oil shale/coal blend), wt%

| Ash | CO ₂ | C _{CO2} | TC* | C _{org**} | SO ₃ | SiO ₂ | R ₂ O ₃ | CaO | CaO _{fr} | MgO | K ₂ O | Na ₂ O | Cl |
|------------|-----------------|------------------|------|--------------------|-----------------|------------------|-------------------------------|-------|-------------------|------|------------------|-------------------|------|
| Bottom ash | 14.93 | 4.07 | 4.70 | 0.63 | 9.41 | 14.03 | 5.65 | 49.88 | 20.68 | 5.06 | 1.8 | 0.33 | 0.16 |
| ESP1 | 3.33 | 0.91 | 1.15 | 0.24 | 5.33 | 34.00 | 14.16 | 29.6 | 13.49 | 6.1 | 2.44 | 0.24 | 0.21 |
| ESP2 | 3.15 | 0.86 | 1.11 | 0.25 | 5.25 | 36.16 | 13.93 | 31.38 | 12.71 | 6.03 | 3.55 | 0.48 | 0.37 |

* TC – total carbon

** C_{org} = TC – C_{CO2}



Fig. 5. Coal pieces still burning on the bottom ash conveyor.



Fig. 6. Oversized coal pieces in the fuel blend.

were found in the fuel. Particle size analysis showed that a higher share of coal in the fuel mixture resulted in larger particles (Fig. 7). The maximum particle diameter allowed for the boiler by the design was 15 mm. The fuel preparation system was not able to crush coal as efficiently as required because Oil Shale Hard Groove Index (HGI) is considerably lower when compared with coal. The mineral composition of ash is presented in Table 7. The composition of ash flows, i.e. bottom ash (BA) and ashes of electrostatic precipitator field 1 (ESP1) and field 2 (ESP2), is similar to that of ash from pure oil shale combustion. The calcite content is typically higher in BA, dolomite exists only in BA and is absent in fine ash fractions (ESP1 and ESP2). The anhydrite (CaSO_4) content is the highest in the ESP2 ash, which indicates the binding of sulphur to finer ash fractions.

Chemical analysis of the ash samples showed a measurable organic carbon content (Fig. 8). Based on on-site observations, unburnt organic carbon was expected in the bottom ash, but fly ash contained also some unburnt organic carbon. The carbon content and share of coal in the fuel mixture correlated. Heat losses due to unburnt fuel during experiment D (29% heat from coal) were 0.79%. This value is low compared to the typical values for coal boilers. The heat losses due to unburnt fuel when working with oil shale are as low as 0.02% [1].

Table 7. Mineral composition of ashes (85/15 oil shale/coal blend), wt%

| Ash \ Compound | CaCO_3 | CaO | CaSO_4 | MgO | $\text{CaMg}(\text{CO}_3)_2$ | KAlSi_3O_8 | Fe_2O_3 | $\text{Ca}_2\text{Al}[\text{AlSiO}_7]$ | SiO_2 | KCl |
|----------------|-----------------|--------------|-----------------|--------------|------------------------------|----------------------------|-------------------------|--|----------------|--------------|
| Bottom ash | 44.69 | 8.57 | 3.62 | 3.41 | 2.63 | 1.34 | 0.2% (< 0.5) | 0.00 | 3.06 | 0.03 |
| ESP1 | 8.50 | 16.98 | 6.38 | 3.70 | 0% (< 0.5) | 6.25 | 1.01 | 2.18 | 8.30 | 0.43 |
| ESP2 | 8.01 | 16.65 | 7.22 | 3.88 | 0% (< 0.5) | 5.11 | 0.78 | 2.03 | 6.49 | 0.3 |

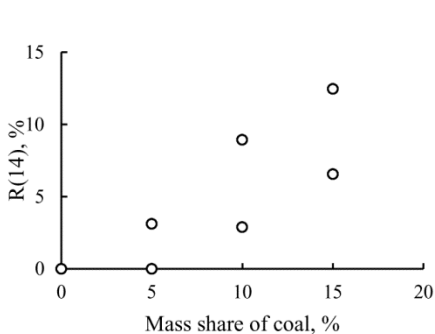


Fig. 7. Share of particles with a diameter over 14 mm, %.

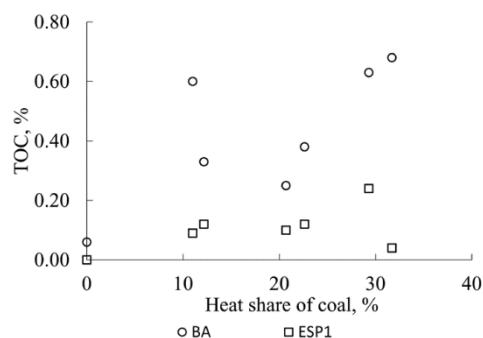


Fig. 8. Total organic carbon content in the bottom ash and ESP1 depending on the heat share of coal in the fuel mixture.

Ash samples were collected and analysed, but no major changes in its chemical composition were noticed. The ashes were similar to typical oil shale CFBC ashes described by Plamus et al. [11]. The insignificant influence of coal additive on ash chemical composition could be explained by the relatively low coal ash content (15%) compared to oil shale's (55%). Nevertheless, some trends were still observed in the bottom ash: manganese had a positive correlation and chloride a negative correlation with coal mass share. The higher share of coal in the fuel mixture resulted in an increased content of sulphur, sodium, phosphorus, manganese, magnesium, aluminium and titanium in the fly ash. The content of potassium in the fly ash decreased. Although a number of trends were noticed, the changes were minor and should not alter the physical or chemical properties of the ashes.

3.3. Flue gas and emissions

The values of average concentrations of major gas emissions are presented in Table 8. The flue gas did not meet environmental requirements ($\text{NO}_x > 200\text{mg/Nm}^3$ at 6% O_2) due to the high nitrogen oxides concentration (Fig. 9).

The data analysis showed that nitrogen oxides concentration correlated with nitrogen content in the fuel mixture (Fig. 10). So the excess NO_x was formed due to additional nitrogen in the fuel. The temperature in the boiler was around 800 °C, thus the nitrogen in air did not contribute to the NO_x formation. The nitrogen content in oil shale was low (< 0.1%). Adding any fuel with a higher nitrogen content necessitated the use of a De NO_x solution.

Sulphur dioxide emissions remained nearby the quantification limit of a standard FTIR gas analyser (5 ppm). The coal sulphur content was rather low and in addition, oil shale contained a lot of calcium that binds sulphur well in the CFBC boiler. It is possible to co-fire oil shale in the CFBC boiler with coals containing more sulphur without extra sulphur removal devices.

Table 8. Average concentration of main pollutants in flue gas (dry, 6% O_2), mg/Nm^3

| Fuel | Heat share of coal, % | CO | NO_x | SO_2 |
|---------------|-----------------------|-------|---------------|---------------|
| A | 0 | | | |
| B | 11 | 31 | 260 | 10 |
| C | 21 | 36 | 356 | < 5 |
| D | 29 | 47 | 404 | 7 |
| E | 12 | 20 | 296 | < 5 |
| F | 23 | 45 | 372 | < 5 |
| G | 32 | 60 | 455 | < 5 |
| OS + peat [2] | Peat 30% | 60 | 165 | 9 |
| OS + BIO [1] | Biomass 15% | 20–30 | 140–200 | < 5 |
| OS 8.5 [14] | | 35 | 120 | 15 |
| PC OS [15] | | 30 | 220 | 3000 |

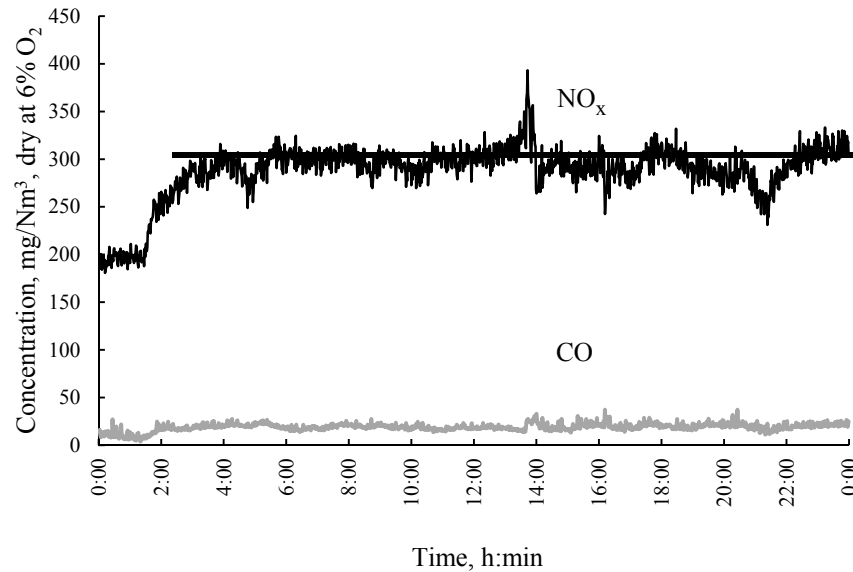


Fig. 9. Nitrogen oxide and carbon monoxide concentrations in the flue gas: oil shale: 7.5 MJ/kg, coal: 19.7 MJ/kg, fuel blend E (95/5).

A slight increase of carbon monoxide concentration was noted (Fig. 11) when the share of coal was increased in the fuel mixture. A similar behaviour was observed when peat was co-fired with oil shale [2]. When firing peat, the temperature fluctuations in the boiler were noticed, but this is not the case herein. It seems that the temperature is not sufficient to ensure a 100% burnout of the volatiles.

The burning experiments under oxy-fuel conditions have exhibited similar or low SO_2 and NO_x levels in flue gases compared to the conven-

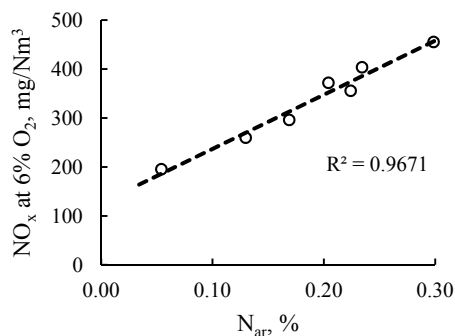


Fig. 10. Nitrogen oxides concentration in the dry flue gas compared with the nitrogen content of the fuel mixture.

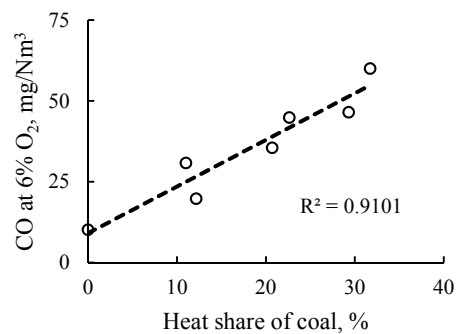


Fig. 11. Carbon monoxide content in the dry flue gas.

tional CFB combustion [16]. The nitrogen conversion ratio to NO_x was found to be up to 24% and the emissions were rather low [16]. Investigations by Al-Makhadmeh et al. [17] have shown that in case of Jordanian El-Lajjun oil shale the NO_x emissions are lower under oxy-fuel conditions than in air-firing mode.

Estonian oil shale has a molar Ca/S ratio of 8–10 and no further addition of lime is necessary for the complete binding of sulphur. The sulphur content of El-Lajjun oil shale is nearly four times higher than that of Estonian oil shale. CaO and ash contents are comparable in both fuels. Al-Makhadmeh et al. [18] have found that a Ca/S molar ratio of 3 is sufficient to obtain the desulphurisation efficiencies up to 93–100%. To bind the sulphur it is necessary to add limestone.

The content of total suspended particles (TSP) remained within the same limits, 30–40 mg/Nm^3 , independently of fuel blends. The content of the finest particles of fly ash together with its mass division (PM10/2.5) after ESP is an important indicator of flue gas composition. The mass division of the finest fly ash after ESP was determined for three coal and oil shale blends: 5/95, 10/90 and 15/85 (Fig. 12). It can be concluded that the distribution of fine fly ash does not depend on the fuel blend.

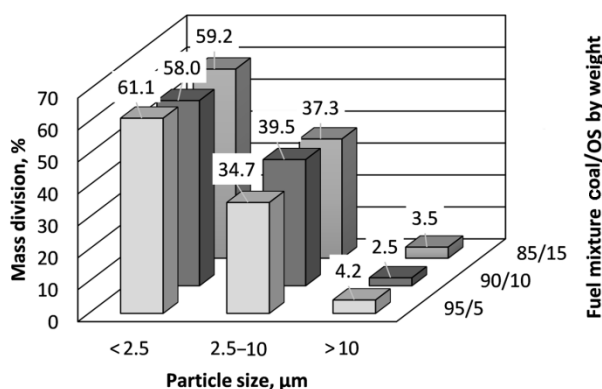


Fig. 12. Fly ash mass distribution after ESP.

3.4. Issues and solutions

The most important problem was the high nitrogen oxides concentration in the flue gas. There are no cheap ways to solve it. If it is necessary to use more nitrogen containing fuel in the power plant, then the De NO_x equipment must be installed. In the CFB boiler, the temperatures are considerably lower than in the PC boiler, but the technology requires temperature between 760 °C and 1090 °C. From Figure 3 it can be seen that the temperatures in the boiler stayed below the required values. It might be possible to increase the temperature of the gases, but it would be a technical challenge. There are

no easy ways for reduction of nitrogen oxides emission for this type of technology.

When co-firing coal and oil shale, two further problems were related to the presence of unburnt combustibles in the ashes and increased carbon monoxide concentration in the flue gas. Both increased the boiler heat losses, but were not as crucial as excess nitrogen monoxides in the flue gas. The TGA (Fig. 2) and proximate analysis (Table 2) suggested that there should be no problem with fuel burnout since coal had a high volatile content and its ignition and burnout temperatures were low. Coal was basically similar to oil shale; the only major difference was its considerably lower contents of ash and carbonate. However, the fuel-handling system for OS was not able to prepare the coal fuel. It resulted in oversized particles in the fuel feed and increased unburnt carbon content in the bottom ash. Upgrading the fuel handling system can reduce unburnt carbon in the bottom ash. Increasing the temperature in the furnace and balanced fuel feeding may decrease the losses due to unburnt fuel in the fly ash and carbon monoxide in the flue gas.

4. Conclusions

Coal co-firing with oil shale in an industrial CFBC boiler was performed. The maximum heat share of coal in the fuel blend was 31% and maximum mass share 15%. During the experiments the boiler maintained a stable load of 250 MW_{th} (83–89 kg_{steam}/s). The flue gas emission did not meet the environmental requirements due to increased NO_x concentration. The NO_x concentration in the flue gas correlated strongly with the nitrogen content in the fuel mix. This indicates that the formation mechanism of NO_x did not change. The average SO₂ concentration in the flue gas remained close to the detection limit. The fuel preparation system could not provide the fuel of required quality, it contained oversized coal particles. This resulted in unburnt carbon in the ash and increased heat losses. The average CO concentration correlated with the coal share in the fuel mix, but remained in all cases below 70 mg/Nm³. The ash produced was similar to the typical calcium rich oil shale ash.

The chemical analysis of the ash samples showed a measurable organic carbon content especially in BA, but also in the ESP ash. The highest TOC content was 0.6%. Typically, there was no problem with oil shale burnout, the organic carbon content in the boiler ashes was below 0.1%.

Coal and oil shale co-firing fuel preparation should be continuously improved and the coal used should contain minimal organic nitrogen.

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