

# PETROLEUM COKE AND ELECTRODE CARBON FIRING IN CFBC BOILERS

by

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*The Romanian economic and energy situation requires a large-scale implementation of clean and efficient technologies for the utilisation of different residues and wastes from industry. The need for such technologies has become even more acute, since January 1998 when new environmental regulations became law.*

*This is why our research is focused on the clean combustion of industrial wastes, sludge and biomass. The research work initiated aims to establish a circulating fluidised bed (CFB) combustion and desulfurisation technology for these wastes. In the current paper, the results obtained for two types of residues are presented: a solid waste from the production of electrodes for metallurgic furnaces, and the high-sulphur petroleum coke, that is a residue from oil cracking.*

## Introduction

The industrial development registered worldwide in the last decades has led, besides the growth of energy demand, to the accumulation of high quantities of wastes, with a negative environmental impact.

As a consequence, the regulations for environment protection constrained the industrial producers to eliminate their residues or, if possible, to find an alternative use for them. Such examples are paper mill wastes, used tyres, biomass, sludge, petroleum residues. Not long ago, high sulphur petroleum coke was also considered to be among these, because it is not suitable for use in industry, where it should meet certain quality conditions. Once desulfurisation technologies developed, its utilisation as a fuel in power plants became possible. In some countries – such as USA and Denmark – it is even used on a large scale [1].

However, not all desulfurisation technologies provide a competitive price for the energy produced even from a low-price fuel such as delayed coke. A more attractive solution from this point of view is circulating fluidised bed combustion (CFBC), which allows the in-bed limestone desulfurisation.

The data obtained from several Romanian refineries allowed us to estimate the internal production of high sulphur petroleum coke at about 360.000 tons annually. Most of it is currently exported at 40 USD per ton, and the price could further decrease by the end of the year.

In Romania, no power plants use petroleum coke as a fuel. Another potential fuel with no utilisation in Romania is the waste resulting from the production of electrodes for metallurgic ovens. In spite of its high heating value, this waste is not being used because of the difficulties encountered at its combustion in classical furnaces.

Since their utilisation seems to be both necessary and cost-efficient, we performed several combustion and desulfurisation tests with petroleum coke and electrode waste on a 1 MWt CFBC pilot plant to determine the optimum operating parameters.

### Description of the test facility

The experimental pilot plant is the first CFBC plant built in Romania. The fuel it was designed for was lignite with heating value higher than 5000 kJ/kg and size range of 0–10 mm, or oil shale with 3800 kJ/kg and 7% fuel oil support.

A simplified scheme of the boiler is presented in Fig. 1; its main components are:

- furnace;
- cyclone;
- loop seal;
- external fluidised bed heat exchanger (FBHE);
- convective pass;
- air preheater.

The combustion takes place in the furnace, both in the dense bed from the bottom and above it, where the particle density is lower. The start-up of the plant is achieved by means of a fuel oil burner that warms the fluidised bed up to the solid fuel ignition temperature, when the combustion process is initiated.

Two water circuits ensure the cooling of the plant. The main cooling circuit consists of the heat transfer surfaces located in the fluidised bed ash cooler, the furnace and the convective channel. The secondary circuit cools the ash screw conveyer and the fixed bed ash cooler.

The solid fuel is introduced into the fluidised bed by means of screw conveyers. During the fluidising process, fuel particles are mixed with recirculated ash particles, and burn as they are elutriated in the upper part of the furnace. To ensure complete combustion and in order to achieve the desired gas velocity, secondary and tertiary air are introduced at two different levels. The coarse particles that are not carried by the flue gas, are discharged through the drain pipe from the air distributor and removed by means of a water-cooled screw conveyer.



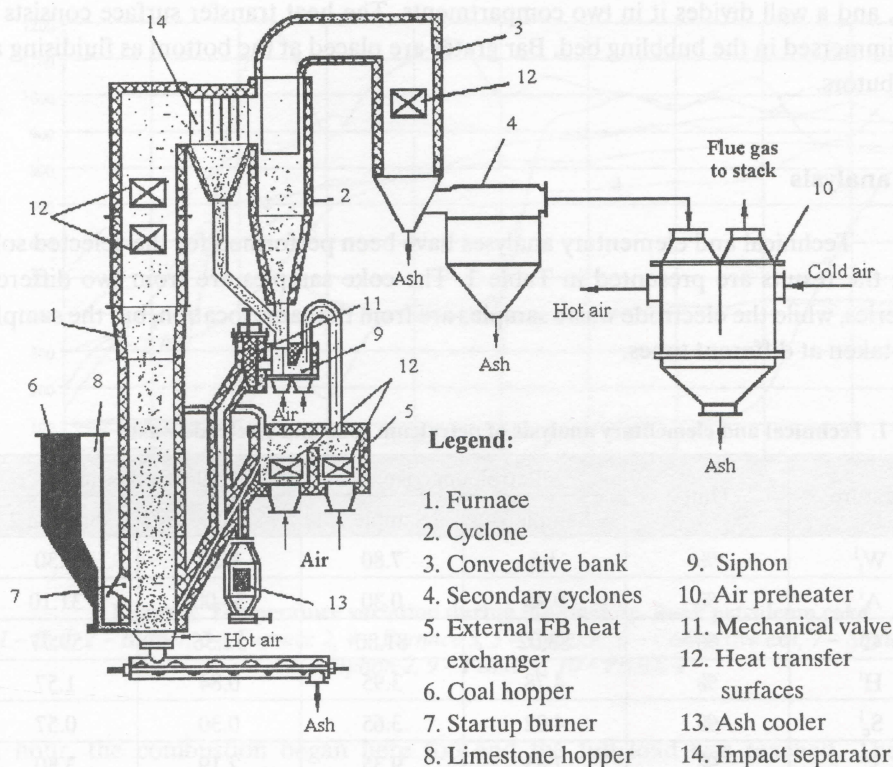


Figure 1. The 1 MWt CFBC pilot plant

The test rig is equipped with a tangential entry reverse flow single cell cyclone, where the coal and ash particles are centrifugally separated from the flue gas and fall through a duct into the siphon. The hot flue gas are evacuated at the upper part through a 90° turn and sent in the convective pass. An impact separator was fitted between the furnace and the cyclone for evaluation purpose, but only for a short period.

The siphon consists of a mechanical valve and two compartments split by a dividing wall. The movement of the solids in the non-mechanical valve is facilitated by air, which is introduced through a bar grate similar to that from the furnace.

Apart from its sealing function, this device makes possible the control of the fraction of ash directed into the external FBHE, by modifying the air flow in the two compartments. A strict control of the furnace temperature is obtained by diverting the solids either directly into the oxidising zone of the furnace, or into the external fluidised bed heat exchanger.

The external fluidised bed heat exchanger (FBHE) is used to supplement the furnace heat transfer surfaces, also making the boiler more fuel flexible. It is refractory

lined, and a wall divides it in two compartments. The heat transfer surface consists of coils immersed in the bubbling bed. Bar grates are placed at the bottom as fluidising air distributors.

## Fuel analysis

Technical and elementary analyses have been performed for the selected solid fuels; the results are presented in Table 1. The coke samples are from two different refineries, while the electrode waste samples are from the same location, but the samples were taken at different times.

**Table 1. Technical and elementary analysis of petroleum coke and electrode waste**

Feature	Unit	Petroleum coke		Electrode waste	
		Sample 1	Sample 2	Sample 1	Sample 2
$W_t^i$	%	1.6	7.80	9.30	4.30
$A^i$	%	0.45	0.30	37.00	31.10
$C^i$	%	88.02	81.80	52.56	59.77
$H^i$	%	4.78	3.95	0.84	1.57
$S_c^i$	%	4.02	3.65	0.30	0.57
$V^i$	%	7.85	9.35	2.19	3.80
$V$ (d.a.f.)	%	8.01	10.17	4.08	5.88
$Q_i^i$	kJ/kg	34338	31219	17958	21524

i = initial

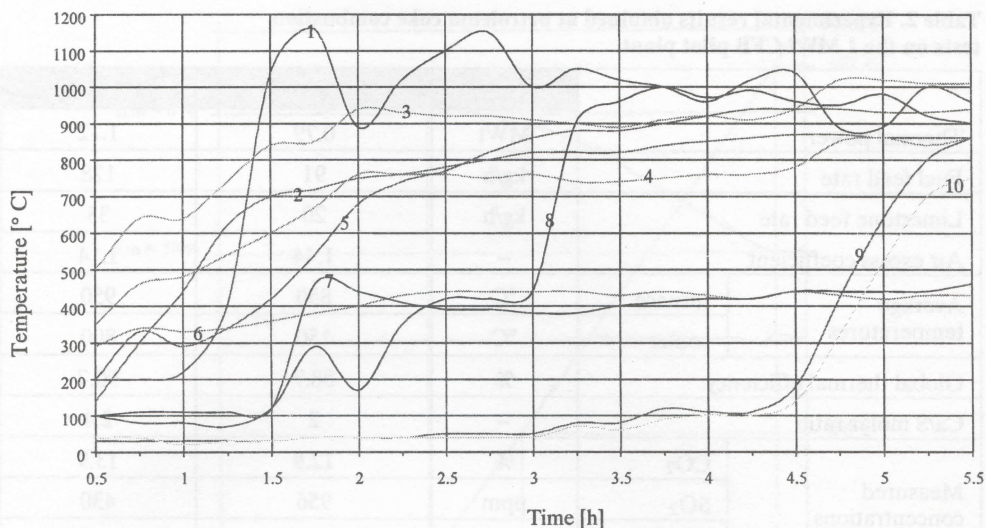
## Petroleum coke experiments

The results of the petcoke combustion experiments are presented in Table 2. Since the petroleum coke has practically no ash, during the preliminary combustion tests sand was introduced in the furnace as inert material. In the final tests, the limestone added for desulfurisation served as bed material.

The particle size distribution of the petcoke for all the experiments was between 0 and 10 mm, with mean diameters between 1.9 and 3 mm, while the size range of the sorbent was between 0 and 1 mm, with mean diameters between 92 and 500  $\mu\text{m}$ .

A detailed presentation of the flue gas temperatures from test no. 2 can be seen in Fig. 2. The bed temperature was above 1000  $^{\circ}\text{C}$  as long as the external fluidised bed heat exchanger (FBHE) was by-passed. The direct recirculating valve was then gradually closed, so a larger fraction of the solids was recirculated through the FBHE. After about





**Figure 2. Temperature variation during the start-up. Fuel: petroleum coke**

1 – Bed, 2 – Burner, 3 – Furnace 2, 4 – Furnace 3, 5 – Cyclone, 6 – Convective exit, 7 – Siphon 1, 8 – Siphon 2, 9 – FBHE 1, 10 – FBHE 2

an hour, the combustion began here too and the full load was reached. The bed temperature stabilised at 980 °C.

The combustion efficiency was over 99%, in spite of the low air excess. In test no. 1, because the combustion was taking place only in the furnace, the bed temperature was between 1100–1200 °C. This fact, correlated with the high air excess, has led to a high  $\text{NO}_x$  emission – about 300 ppm.  $\text{SO}_2$  concentration was also high, because of the low temperatures from the upper furnace and cyclone, that prevented the sulfation of the CaO particles.

A better desulfurisation efficiency was obtained in test no. 2, when the temperatures have been in the optimum range of 850–1050 °C [2] all along the recirculation loop: furnace-cyclone-FBHE-furnace. The analysis of solids proved that the sulfation is higher in fine particles, due to their higher active surface, in spite of the shorter residence time in the reaction zone. The fraction of calcium carbonate decomposed into oxide was around 98%. However, considering the relatively high Ca/S molar ratio (2.4), the 80% efficiency that was obtained is still low. This is mainly because of the small size of the limestone particles – 50% less than 63  $\mu\text{m}$ , with an average diameter of 92  $\mu\text{m}$  – that were not recirculated, leading consequently to a very short contact time between CaO particles and the flue gas. Better results are expected in the case of industrial plants with greater furnace height.

**Table 2. Experimental results obtained at petroleum coke combustion tests on the 1 MWt CFB pilot plant**

		Unit	Test 1	Test 2
Thermal power		MWt	0.79	1.12
Fuel feed rate		kg/h	91	128
Limestone feed rate		kg/h	20	35
Air excess coefficient		–	1.44	1.14
Average temperatures	furnace	°C	850	950
	cooler	°C	150	800
Global thermal efficiency		%	88.7	90.7
Ca/S molar ratio		–	2	2.4
Measured concentrations at 6% O <sub>2</sub>	CO <sub>2</sub>	%	12.9	13.9
	SO <sub>2</sub>	ppm	956	430
	NO <sub>x</sub>	ppm	302	142
	CO	ppm	111	65
Desulfurisation efficiency		%	53	80

## Electrode waste experiments

Its particular characteristics, and the fact that this fuel has not been thoroughly studied until now, made the thermogravimetric analysing essential for electrode waste. One of the diagrams is presented in Fig. 3, and it confirms once more the low reactivity of this fuel: up to 380 °C, the only phenomena that occur have a physical nature, and complete combustion is only obtained at 950 °C.

The limited heat amount released by the volatile matters at temperatures over 380 °C is not sufficient to start the burning of fixed carbon, meaning that in an industrial plant, the furnace temperature should be increased to at least 700 °C using liquid fuel, so the start-up procedure will last longer. These conclusions were also supported by the many trials performed on the pilot plant for the autothermal combustion of the wastes.

At the preliminary tests, it came out that the combustion conditions are very difficult to achieve. After more attempts to run the CFBC plant on electrode waste only, it was judged to be impossible to maintain adequate temperature conditions; in consequence, a mixture containing 60–70% electrode waste and 30–40% lignite was fed. With this mixture as a fuel, the operation became steady and the first combustion tests have been performed.

The combustion efficiency achieved was with up to 4% lower for the combustion of electrode waste and lignite mixture than in the case of petroleum coke. This can be explained by the non-uniform size distribution (the electrode waste having 22.5% under



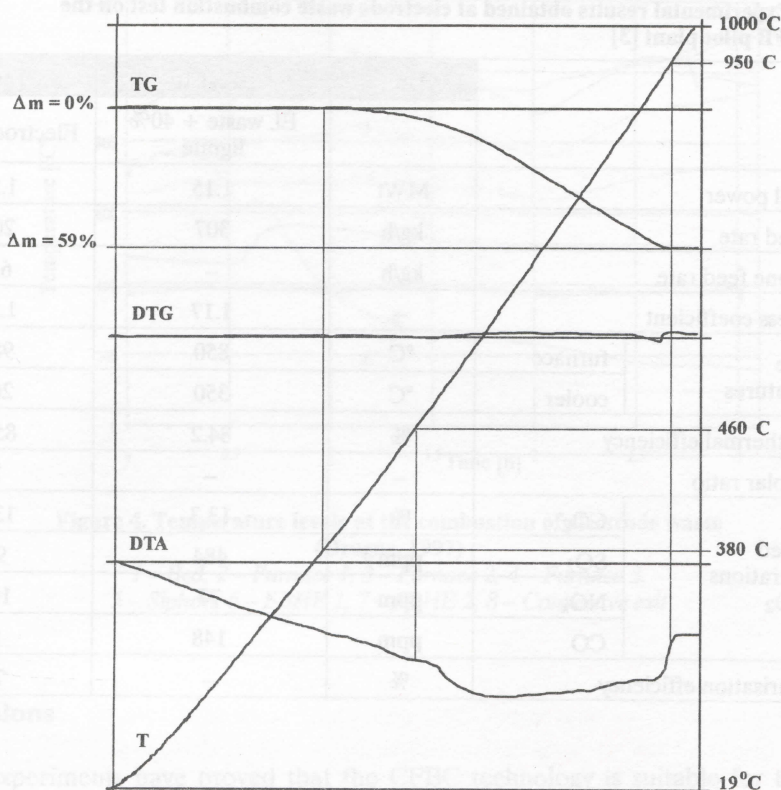


Figure 3. Thermogravimetric analysis of the electrode waste [3]

200  $\mu\text{m}$ , compared to 10% for the lignite from the mixture). Thus, a large fraction was not recirculated and had an insufficient time for combustion, the average carbon content from ash being higher (11.6%). Limestone was not introduced, but the measured  $\text{SO}_2$  emissions were with 10–15% lower than the theoretical concentration, due to the self-desulfurisation process.

Another combustion test has been performed using a different size distribution, with the fraction under 200  $\mu\text{m}$  of only 15%, but with 35% between 10 and 15 mm. Average results for this test are presented in Table 3 – test 2. The electrode waste was introduced in the lignite-fired furnace at a temperature of over 800 °C (see Fig. 4). The feeding with electrode waste continued for more than three hours; with the external fluidised bed heat exchanger not being used, the furnace temperature reached 1000 °C. On the temperature diagram, the moments when the direct recirculating valve was opened and the air flow was reduced in the siphon are clearly marked by the temperature

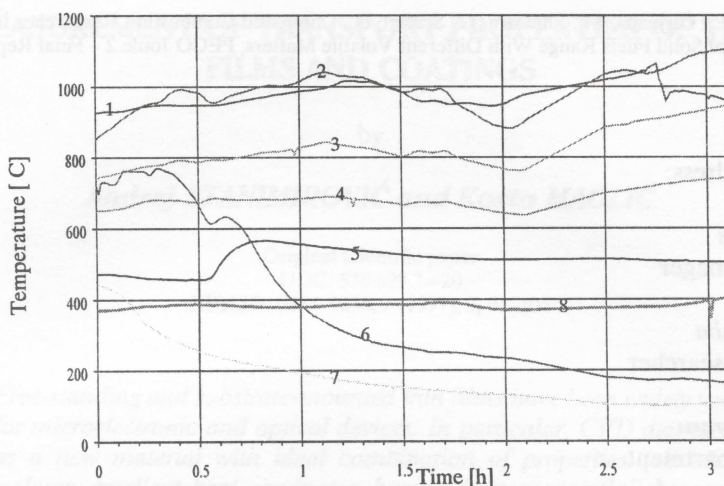
**Table 3. Experimental results obtained at electrode waste combustion test on the 1 MWt CFB pilot plant [3]**

		Unit	Test 1	Test 2
Fuel		–	El. waste + 40% lignite	Electrode waste
Thermal power		MWt	1.15	1.21
Fuel feed rate		kg/h	307	200
Limestone feed rate		kg/h	–	6.8
Air excess coefficient		–	1.17	1.20
Average temperatures	furnace	°C	850	980
	cooler	°C	350	200
Global thermal efficiency		%	84.2	85.9
Ca/S molar ratio		–	–	2
Measured concentrations at 6% O <sub>2</sub>	CO <sub>2</sub>	%	13.3	13.4
	SO <sub>2</sub>	ppm	484	99
	NO <sub>x</sub>	ppm	77	108
	CO	ppm	148	0
Desulfurisation efficiency		%	–	79

variation – a decrease in the external FBHE and an increase in the siphon. The high furnace temperature ensures good combustion and no CO was detected in the flue gas. However, the short residence time of the fines has again led to a high carbon content in the ash, but due the fact that no lignite was added, it can be considered that the electrode waste combustion efficiency is much better than in the previous experiments. The small height of the pilot plant should also be considered, since in the case of a higher capacity industrial plant, with a much greater furnace height, the unburned carbon content in the fines ought to significantly decrease due to the longer residence time.

Limestone was introduced at a Ca/S molar ratio of 2, obtaining a 79% desulfurisation. The relatively low value of the desulfurisation efficiency is due to the lower sulphur content of the fuel, giving thus a lower SO<sub>2</sub> partial pressure, but also due to the temperatures above 1000 °C, that are higher than the optimum temperature range of the process. Increasing the Ca/S ratio to 3 did not prove to be practical, not only because the desulfurisation efficiency improved by just 5%, but also since SO<sub>2</sub> emissions are low enough even without limestone addition.





**Figure 4. Temperature levels at the combustion of electrode waste (Dragos, 1997)**

1 – Bed, 2 – Furnace 1, 3 – Furnace 2, 4 – Furnace 3,  
5 – Siphon, 6 – FBHE 1, 7 – FBHE 2, 8 – Convective exit

## Conclusions

- the experiments have proved that the CFBC technology is suitable for the clean combustion of both the high sulphur petroleum coke and the electrode waste;
- combustion efficiencies over 99% can be achieved in the case of petroleum coke with very low air excess;
- by a proper adjustment of the air staging, low  $\text{NO}_x$  emissions can be obtained for both fuel types tested;
- for an efficient desulfurisation, the temperatures should be above 800 °C all along the recirculation loop, and the average size of limestone particles – under 100  $\mu\text{m}$ ;
- the very low volatile percentage gives the electrode waste a very low reactivity, in spite of its high heating value;
- electrode wastes can burn autothermally, but at higher temperatures, that the plant will have to ensure;
- an industrial plant using electrode waste as fuel should have bigger start-up burners and a greater furnace height.

## References

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