RESEARCH REGARDING DESULPHURIZATION OF FLUE GASES RESULTED FROM THE ELECTRIC THERMAL POWER PLANT HALÂNGA
CERCETARI PRIVIND DESULFURAREA GAZELOR DE ARDERE EMISE LA CET HALÂNGA

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Abstract: **This paper presents the need for the reduction of the pollutant's concentration in the flue gases from the steam boilers of the Halânga electric thermal power plant (CET). By comparing these results with the legal limit values of the emissions, the necessity to consider very efficient and urgent measures for flue gases desulphurization is more than evident. In order to depict the most appropriate method and technology, series of comparative experimental tests have been organized by the authors. A lab pilot plant working in fluidized bed - from the Laboratory of Renewable Energy Resources of the POLITEHNICA University of Timişoara** http://energieregen.mec.upt.ro/**- was used for research to test the different methods for the sulphur dioxide retention, and not only. The achieved results were essential for selecting and planning a new desulphurization plant using calcium carbonate, which will equip the large combustion facilities from the power plant Halânga, before 2010.**

Key words: **Flue gases, Pollutant emissions, Pollution control, Desulphurization**

Descriptori: Gaze de ardere, Noxe poluante, Controlul poluării, Desulfurare

Contribuţii

Contribuţia autorilor se referă la măsurătorile în situ pe cazanele funcţionale de la CET Halânga, experimentarea soluţiilor de desulfurare pe staţia pilot şi interpretarea rezultatelor. Experimentările s-au realizat în comun la Universitatea POLITEHNICA din Timişoara, iar măsurătorile de calitate a arderii în baza cărora s-au realizat calculele generale, s-a identificat strategia şi soluţia de desulfurare au fost realizate pe cazanele, în funcţiune, de la RAAN CET Halânga.

1. Prologue

Humankind living and further development under comfortable and healthy conditions require more and more heat power and electricity. A large amount is generated through fossil fuel combustion, such as brown coal, which is a low-grade coal, with high ballast content (water and ash) i.e. low calorific power (Table 1). Brown coal utilization to generate energy raises many difficulties regarding both ignition and flame stability and the complex environmental impact: air, water, ground, and fauna. A large amount of pollutants is generated, such as: SO_x, NO_x, CO, CO_2, HCl and FCl vapours, in addition ash, unburned matter and soot [1]. The flue gas filtering installation is capturing only ash. The slag is discharged mainly in the furnace, typically using a hydraulically system to the slag dump. Every-so-often, the dust and the other emitted pollutants released during coal transportation and storage, as from the exhaust of the flue gases through the chimneys, as well as the one originating in the ash dump affect the air quality in the neighborhoods [2].

The currently operating coal-fired thermal power plants from Romania were designed and built 25-35 years ago when no environmental protection legislation was effective. The only measures for this purpose provided two-step electrostatic precipitators to dedust flue gas, respectively a very high smoke stack (240-280 m), to disperse pollutants, such as NOx, SO2, non-removed particles etc., on a wide range. These solution are nowadays not acceptable and more, by no means [3].

The situation is similar also in the Power Plant (CET) Halânga which is involved in the generation of 16 and 38 bar industrial steam required by the Heavy Water Plant (ROMAG PROD), electric power delivered to the National Power System (NPS, and not at last, heat power as hot water required by the district heating system of Drobeta Turnu Severin municipality. CET Halânga consists mainly of two large combustion plants (IMA-1 and IMA-2), each fitted with three boilers having as steam parameters a flow of D_n = 420 t/h, a pressure of p_n= 13,7 MPa and a temperature of t_s= 420^OC [4]. They are powder brown coal fired combustion facilities, which provides up to 94% of the heat necessity required by the steam generation, the rest being assured by a support of hydrocarbons. Table 1 features the main characteristics of the brown coal used: amounts of ash (A^{rap}), humidity (W_t^{rap}) and sulphur (S_c^{rap}). For ignition, flame stability and operation under reduced loads, the low sulphur fuel oil is normally used (Si <1%) is used, which provides the rest of the input heat necessity.

Table 1

Brown coal characteristics, as used in CET Halânga*

Coal mine	H_i [MJ / kg]	A^{rap} $\left[\frac{\%}{MJ/kg}\right]$	W_t^{rap} $\left[\frac{\%}{MJ/kg}\right]$	S_t^{rap} $\left[\frac{\%}{MJ/kg}\right]$
BOR	9,529	3,110	2,895	0,159
ROMALEX LOT 2	8,449	3,738	3,474	0,179
LUPOAIA	8,650	2,473	3,393	0,102
KOVIN	9,956	2,110	3,574	0,110
ROMALEX	14,444	1,044	1,997	0,147
GÂRDOAIA	8,306	2,965	4,751	0,045
ROȘIUȚA	8,738	2,029	4,988	0,097
LORFA	8,499	4,150	3,177	0,068
VALEA COPCII	6,833	3,230	6,627	0,184
A.M.W. STYLE	8,813	1,374	5,021	0,152

*Testing Report NB 134/28.03.2007 and NB 489-497/23.05.2007 delivered by SC ICEMENERG SA.

The required steam flow is provided by the simultaneous operation of two or three boilers. As there is no continuous NO_x, SO_2, or CO monitoring system, monthly on line thermodynamics measurement were performed on the operating boilers, starting February 2003 [5]. Based on the numerous data obtained through measurements, Table 2 exemplifies the average concentration amounts of NO_x in dry flue gas with $O_{2.ref}$= 6 %, measured at boiler K1 in the year 2004. Analyzing the results from 2008, one states that not considerable successful clean combustion technologies were applied, meaning the emissions are still of major environmental impact. It is obvious that while (C_{CO}), (C_{NOx}) and dust (C_{dust}) concentration amounts are under the limit allowed by the current legislation, the (C_{SO2}) concentration exceeds over 12-19 times the legal limit, for this range of thermal facilities. The large amounts and fluctuations for C_{SO2} are generated by the coal high content of sulphur, variable from one coal mine to another (Table 1), as well the diverse S content of the support fuel [6]. Table 3 shows the average concentration of the SO_2 for the six steam boilers, respectively the two large combustion plants.

Table 2

Diverse pollutants' concentration average amounts measured for boiler K1, in the year 2004, in reference to $O_{2.ref}$= 6% -dried flue-gas

Month	Flue gas connecting duct to the stack	C_{CO} [mg/m_N^3]	C_{SO2} [mg/m_N^3]	C_{NOx} [mg/m_N^3]	$C_{powd.}$ [mg/m_N^3]	C_{CO2} [mg/m_N^3]
January	Inferior	75,6	6789,1	354,7	75,8	256,1
	Superior	82,5	6925,3	321,0	83,6	274,3
February	Inferior	67,9	7025,4	358,6	94,3	268,9
	superior	69,5	6753,1	401,3	87,3	275,3
March	-	-	-	-	-	-
April	Inferior	81,3	7519,0	362,4	98,9	254,0
	superior	88,7	6264,3	396,5	94,1	249,9
May	Inferior	52,3	5182,3	321,6	77,3	258,2
	Superior	109,0	5948,2	310,7	73,5	272,1
June	Inferior	76,7	6733,7	489,6	96,7	252,1
	Superior	62,4	6651,4	356,5	98,4	250,6
July	Inferior	69,3	5916,5	362,1	90,2	167,3
	superior	67,2	4971,6	372,8	74,3	252,7
August	Inferior	107,0	6873,3	336,0	87,5	252,7
	Superior	120,3	7256,2	352,9	90,9	252,9
September	Inferior	50,4	6424,5	336,2	94,3	253,3
	superior	69,2	5765,8	299,3	76,2	254,0
October	Inferior	64,3	5723,4	286,7	83,1	247,9
	Superior	57,8	6087,3	239,1	79,6	253,6
November	-	-	-	-	-	-
December	Inferior	63,8	6742,0	301,5	91,3	246,7
	superior	72,6	6982,4	299,6	83,4	263,4
Limit concentration		100 1)	400 2)	500 2)	100 2)	-

1) According to the Romanian legislation Order 462/1993 of MAPPM;

2) According to the Romanian legislation HG 541/2003, modified and completed by HG 322/2005.

Under these conditions, considering the basic consumer ROMAG PROD and the operation hours of each boiler, one concluded that IMA-2 should be, immediately, fitted with a wet flue gas desulphurization installation, while IMA-1 will be fitted with a desulphurization system before December, 31, 2010.

In order to depict the most appropriate technical solution, from all known possibilities offered by the literature, as well industrial achievements all over EU, experiments on a lab pilot installation were conducted. Selecting the best desulphurization procedure for the coal combustion currently used at CET Halânga and flue gas desulphurization by means of various concentrations of the reagents were performed in accordance to economic and technical possibilities of the power plant.

Table 3

SO_2 average emission, versus the legal limits, in [mg/m_N^3]

Boiler	Year						Average amount	Average amount IMA
	2003	2004	2005	2006	2007	2008		
K1	6520,7	6422,2	5221,1	5105,9	4985,4	5042,5	5549,6	5469,8
K2	6098,1	6022,4	5158,9	4899,5	4685,5	5011,5	5312,6	
K3	6883,3	6737,7	5095,5	4793,7	4788,5	4985,5	5547,3	
K4	5367,0	5560,0	5521,0	5100,8	5221,2	5108,5	5313,0	4644,7
K5	6360,3	5895,5	5188,9	4391,9	4668,8	5012,2	5252,9	
K6	-	-	5117,2	5082,2	5014,5	4995,8	3368,2	

2. Pilot Installation Description

The experiments were conducted in the "Politehnica" University of Timişoara, Fuel and Ecologic Investigation Laboratory pilot station, whose diagram is shown at Figure 1. Basically is made up of a stationary fluidized bed combustion furnace, convective section, cyclone, scrubber, neutralization reactor, air, water supply system, flue gas discharge system, instrumentation and control [7].

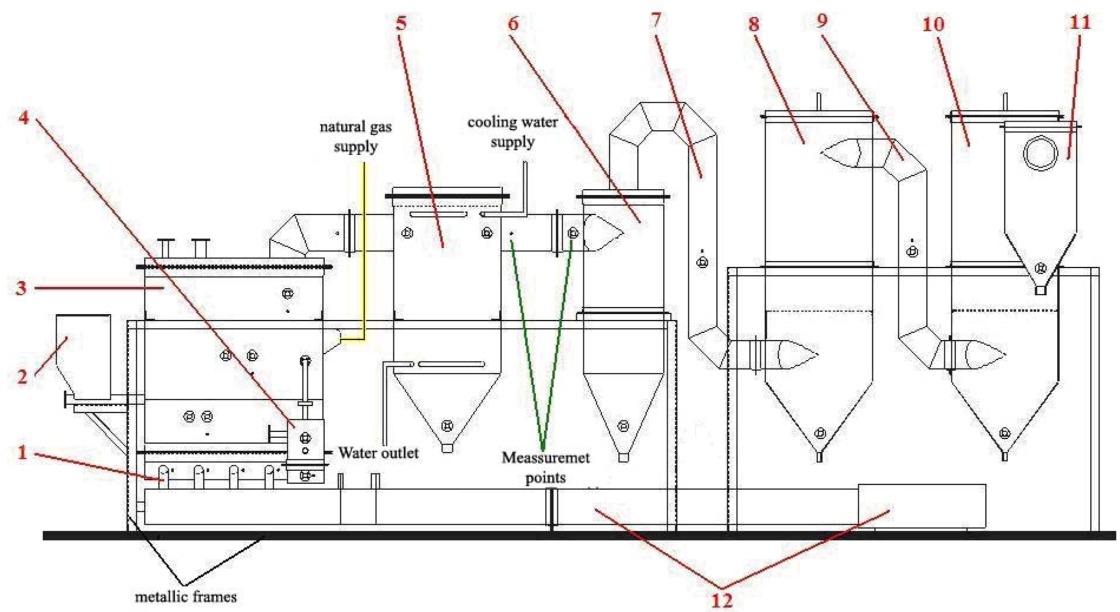

Fig. 1. Fluidized bed combustion pilot station. 1-Air distributor, 2-Solid fuel supply system, 3- Furnace, 4-Ash cooler 5-Convective section, 6-Cyclone, 7-Cyclone - scrubber connecting pipe, 8-Scrubber, 9-Scrubber - reactor connecting pipe,10-Neutralization reactor, 11-Drops separator, 12-Air supply system.

The furnace is fitted with two solid fuel supply systems, each provided with one adjustable rotating helical conveyer. The air, required by the bed fluidization and fuel particles combustion, is supplied by a fan and largely introduced in the distributor erected under the furnace and partially in the ash cooler. For the cold start-up and dioxides self-annihilation as well, the furnace is fitted with a gas burner, mounted on the upper side, at the furnace end, allowing a level of high temperature window at this point.

The hot flue gas leaving the furnace gets cooled in the convective section provided with pipe serpentines through which circulates the water originating in cooling tower. Flue gas dedusting occurs in the cyclone and gets completed in the scrubber where water gets sprayed in counter current with gas movement direction. The neutralization reactor, provided on the bottom side with Rasching rings, desulphurizes flue gas by spraying water solution with a certain amount of reagents. Drops are seized from the discharged gas in the drops separator.

To control the processes within, the pilot station is provided with apparatus and instrumentation to measure automatically and record simultaneously, water and natural gas temperatures, pressures and flow, etc. The heating system is connected with the acquisition plate by way of leads provided with copper cover to protect the signal released by these ones. The data furnished are fed in the database of a computer through the connection unit fastened to the computation system cover. The data acquisition is performed through Lab View program. Gaseous SO_2, CO, NO_x concentrations are determined through TESTO 350 XXL gas analyzer, while dust concentration is set via STRŐHLEIN apparatus using standard methods [8].

3. Methodology Used. Results

The Pilot station start-up complied with the instructions of the operating procedures, that provide mainly the following stages: facility completeness ventilation and utilities existence verification (electric power, water, gas, coal, reagents to treat flue gas), furnace coal and air feed kick-off, start-up the cooling system pump and air compressor required by the membrane pumps, introducing the scrubbing liquid in the scrubber and reactor, gas burner start-up etc. When the temperature in the furnace reached a level of 600-650 ^{o}C, the support burner flame is cut down while the coal bed fluidizing airflow is gradually increased, in order to stabilize the ignition and combustion [9].

Once the operation conditions were stabilized, the flue gas was scrubbed with clean water, sprayed in the scrubber and in the reactor, having various concentrations such as 1,5 %, 2,5 % and 5 % $Ca(OH)_2$, 1,5 % and 2,5 % NaOH. All % are indicated by mass. The exhaust flue gas was analyzed, sampled prior and post flue gas treatment system (SEGA). The results of these measurements, based on which the desulphurization degree was calculated, are shown in Table 4. One notices that the highest desulphurization efficiency degree of 97,5 % is achieved during the experiments refers to the use of scrub from Valea Copcii brown coal, fired with gas, using a 2,5 % $Ca(OH)_2$ concentration water solution injection in the flue gases. In this case, the ultimate concentration of the SO_2 in the $O_{2.ref}$= 6 % by volume, in dried flue gas, decreased in the necessary amount, meaning under the limi (measurements attested C_{SO2}=125,9 mg/m_N^3).

Table 4

Flue gas wet desulphurization measurement results

No.	Brown coal mine or mixture	Injected matter		C_{SO2} [mg/m_N^3] ref. to O_{2ref}		Capture efficiency SO_2 [%]
		Scrubber	Reactor	Prior SEGA	Post SEGA	
1	Roşiuţa	Water	-	3143,6	2601,6	17,2
2	Roşiuţa	Water	2,5% $Ca(OH)_2$ solution	3335,5	226,2	93,2
3	Roşiuţa	Water	5% $Ca(OH)_2$ solution	3343,2	169,9	94,9
4	Gârdoaia	Water	-	1501,0	1228,7	18,1
5	Gârdoaia	Water	1,5% $Ca(OH)_2$ solution	1490,4	424,5	71,5
6	Gârdoaia	Water	1,5% NaOH solution	1564,6	204,3	86,9
7	Gârdoaia	Water	2,5% NaOH solution	1607,1	151,1	90,6
8	Valea Copcii	Water	-	4089,7	3298,9	19,3
9	Valea Copcii	Water	1,5% NaOH solution	6021,3	777,5	87,1
10	Valea Copcii	Water	2,5% $Ca(OH)_2$ solution	4975,6	125,9	97,5
11	40 g $CaCO_3$ to 1 kg Roşiuţa lignite	Water	-	2951,0[1)]	1466,1[2)]	50,3
12	40 g $CaCO_3$ to 1 kg Roşiuţa lignite	Water	-	2951,0[1)]	553,6[3)]	81,2

[1)] No reagent;

[2)] With 40 g $CaCO_3$ to 1 kg brown coal;

[3)] With 40 g $CaCO_3$ to 1 kg brown coal and water injection in the scrubber.

To meet legal provisions [10], besides the option previously mentioned, one can consider any other option that results in a final concentration in the dry flue gases of C_{SO2} < 400 mg/m_N^3. As example one indicates the Gârdoaia brown coal-combustion gas scrubbing with 1,5% NaOH water solution. One specifies that, in terms of operation cost, gas scrubbing with water containing 2,5% $Ca(OH)_2$ is the most economical of all options.

Still Table 4 features good results of the achieved experiments to cut SO_2 concentration in flue gas, originating in Roşiuța brown coal combustion, also by way of the intra-combustion technology [11], [12], [13], respectively LIFAC [12], [14] procedures. Introducing 40 g $CaCO_3$ to 1 kg coal (corresponding to a molar ratio Ca/S = 1,5), one may perform with a primate and attested desulphurization efficiency of 50,3 %, while applying the technology with a post injection of water in the scrubber a value of 81,2% was obtained.

4. Conclusions

"On-line" thermodynamic technical measurements performed monthly, over more than a period of five years for the CET Halânga boilers, delivered a functional and representative database, concerning the evolution and real situation of the pollutant emissions. It is remarkable that such an interest exists, and might be an example for further units, to perform their individual measurements, in accordance to national legislation and tendencies [15]. Based on these data, significant conclusions are issued, regarding the cleaner possible operation of all combustion installations, respectively; actual technical steps are set up to fix the present deficiency, and planning the rehabilitation of electrostatic precipitators and coal dust preparation installations, burners etc.

Very large amounts of C_{SO2} emission (over 12 times as the present legal limit $C_{SO2\ lim}$ = 400 mg/m_N^3 with respect to 6 % O_2) were repeatedly attested by in situ on line measurements. Studies concerning real data emissions and referring to the NO_x dispersion from the Halânga power plant to the neighborhoods depicted possible air quality damaging amounts in the endangered areas, historical areas and Yugoslavia border included. Thus both NO_x and SO_2 exhaust level must be reduced by simultaneous or specific methods. Fluidised bed combustion is used as basic technology, allowing thus the limestone addition. Further second level cleaning technologies are tested and the results are comparative presented.

Good results on wet desulphurization of gas originating in currently used brown coal fluidized bed combustion are actually selected for industrial scale applications, through the wet desulphurization planned facilities to be erected to CET Halânga by the year 2010. These facilities were designed on data resulted from theoretical research and experiments carried out extensively by the authors of this paper, as well.

LIFAC procedure application to flue gas desulphurization from CET Halânga fails to meet the current legislation yet it is an option to consider for IMA-1, assuming its operation for 20.000 hours by 2010, when it is to be decommissioned. After this date, only IMA-2 is to operate as being fitted with a wet desulphurization station.

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