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ENERGY PENALTY MODEL FOR FLUE GAS DESULFURIZATION SYSTEMS

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Abstract. *The present work aims to present the modeling of a Wet Flue Gas Desulfurization WFGD system and deliver the amount of required energy to filter SO₂ from coal flue gas streams. The system simulation depends on operational parameters to calculate the amount of expended energy need for sulfur removal from flue gas streams and the process impact on power generation. Simulated results are compared to available data in literature from coal-fired power plants. An Energy Penalty Function EPF is proposed to calculate the work ratio R_w (kJ/m_N³), defined as the ratio of the expended energy for sulfur removal to the flue gas volume. EPF inputs are the inlet and outlet flue gas temperature, SO₂ content and cleaning efficiency. Design of Experiment (Box-Behnken method) is used to build the EPF and to identify main parameter weighting and cross dependencies. The EPF best desulfurization efficiency interval is 97% to 99,5%, witch achieved a relative deviation of 1.86% in respect to the detailed model. The analysis of the control parameters identified that the inlet and outlet temperature variation does not influence the R_w , which led to a simplified of the EPF. The simplified function reached an overall maximum deviation of 1.41% in respect to the detailed model. The EPF can be coupled to any kind of combustion system to quantify the energy penalty related to remove SO₂ from flue gases released to the environment.*

Keywords: *Energy Penalty Function, Flue gas filtration, Flue gas desulfurization, FGD, Design of Experiment.*

1. INTRODUCTION

Brazilian electricity generation from thermal sources jumped from 4% to 23% since 1990. Thermoelectric plants have increased their share of the electricity market due to the lack of water in the reservoirs of hydroelectric plants, and became a strategic backup for high demand periods (Estadão, 2017).

Non-renewable sources are not affected by natural oscillations and remain strategically needed to ensure the country's energy security. Coal fired power plants must operate with some kind of Flue Gas Desulfurization (FGD) systems to mitigating potential environmental impacts, according to the chemical composition of the fuel, but their use impact both capital and operating costs (Brasil Energia, 2014). Operational duties (OPEX) must be considered as they affect the plant overall efficiency and financial success.

Modeling of FGD systems are reported in literature (Brogren and Karlsson, 1997; Kiil and Michelsen, 1998; Olausson, *et al.*, 1993; Warych and Szymanowski, 2001) focused on the chemical reactions in absorbers, sulfur is

captured. Reported penalties on coal fired power plants were found to be of 1.01% (Feng, *et al.*, 2014), 1.13% (Tsumita, 2007), 1.78% (Sargent, 2002), 1.87% (Gansley, 2008) and 0.60% (Wu, *et al.*, 2017).

Poulikkas (2015) detailed several FGD technologies available on the market, with their correspondent capital and operating costs (CAPEX and OPEX). The Wet Flue Gas Desulfurization (WFGD) system was pointed out to be the predominant technology because it delivers gypsum as a marketable by-product, whereas similar technologies require additional final destination to sulfur components.

In an earlier research, Rubin and Nguyen (1978) studied the influence of FGD system parameters on energy consumption using individual sensitivity analyzes for ten control parameters, which caused variations in energy plant consumption from 2.5% to 6.1%.

Literature review pointed out to a lack of specific models able to forecast energy demand for desulfurization processes, based on operational data and fuel characteristics. The objective of the present work was then defined upon the need of developing an Energetic Penalty Function - EPF, easy to link to any kind of simulation platform or assessment routine. Its output parameter is the work ratio, given in joule per kg of SO₂, as a function of the input flue gas temperature and SO₂ content, the output flue gas temperature and the sulfur removal efficiency.

2. THE FLUE GAS DESULFURIZATION SYSTEM - FGD

2.1 System overview

The Flue Gas Desulphurization system - FGD modeled and implemented in the present work was presented in Tomazi, *et al.* (2017) and it is based on wet scrubbing (Perry, 1997), assuming that flue gases were already free of solid particulates.

The general wet scrubbing FGD system is depicted in the next figure, focusing three of its subsystems, dedicated to the reagent preparation, SO₂ absorbing and gypsum dewatering.

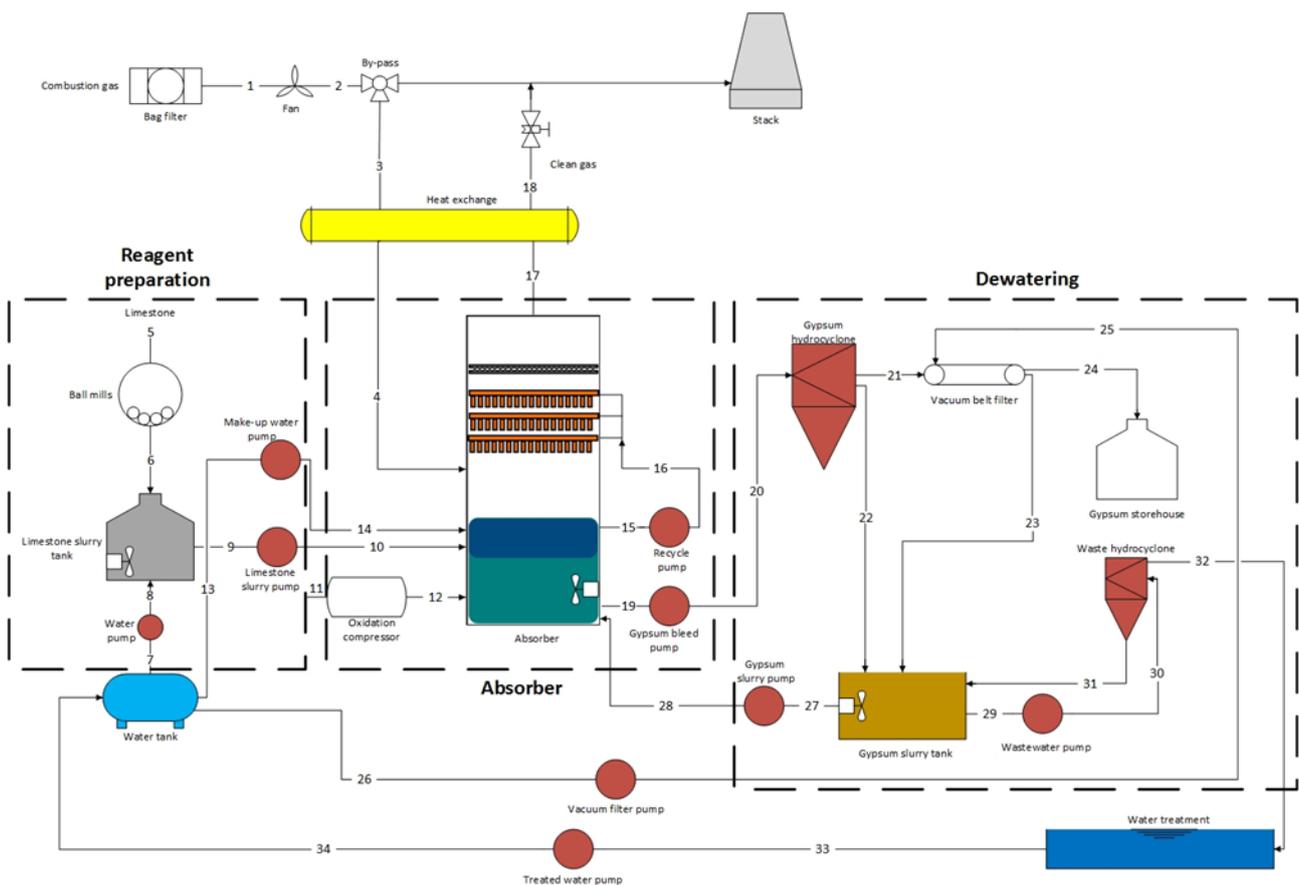
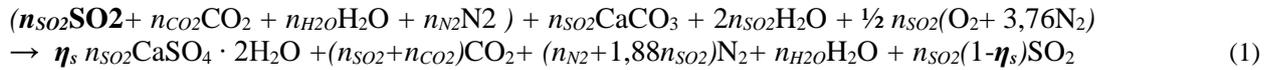


Figure 1. Flue gas desulfurization FGD plant flow diagram with three subsystems: reagent preparation, SO₂ absorbing and gypsum dewatering.

2.2 Absorber

At the heart of the process, a counter flow absorber tank based on a vertical tower gathers the sulfuric acid flue gas stream from flue gases (stream 4) with the alkaline limestone slurry (stream 10) and some extra atmospheric air (stream 12) to produce raw calcium sulfate dihydrate (stream 19). The scrubbing process is described as follows:



with solution approach was focused on the SO₂ capture from flue gases, whose input quantity is highlighted in the first term in parentheses, allowing to calculate the correspondent contents of limestone, water and oxygen reactants. The stoichiometric coefficients n were calculated considering complete combustion of coal with stoichiometric air quantity, and are displayed in the first four terms in parentheses of Eq. (1). The desulphurization efficiency η_s was introduced to represent an incomplete conversion of SO₂ into CaSO₄ · 2H₂O, typical from every FGD equipment, in the place of a more generic kinetic reaction set of equation. The absorber tank was modeled to operate under steady state conditions and its mass balance (Eq. 2) is given as follows, where each parameter is labeled and numbered according to Fig.1:

$$\begin{cases} \sum_{i=in} \dot{m}_i = \sum_{j=out} \dot{m}_j \\ \sum_i \dot{m}_i = \dot{m}_{flue\ gas-in(4)} + \dot{m}_{water(14)} + \dot{m}_{limestone\ slurry(10)} + \dot{m}_{air(12)} + \dot{m}_{gypsum\ slurry(12)} \\ \sum_j \dot{m}_j = \dot{m}_{gypsum(19)} + \dot{m}_{flue\ gas-out(17)} + \dot{m}_{wastewater} \end{cases} \quad (2)$$

Power plant flue gases (stream 3) are cooled down before admission in the absorber (stream 4), where they react with limestone slurry (stream 10), added by an air injection (stream 12). In addition, cooling water (stream 14) aids to control the inside tank temperature and some residual calcium sulfate dihydrate is recovered from the gypsum slurry tank to enhance its concentration (stream 28). The absorber outputs are calcium sulfate dihydrate, or gypsum, and filtered or purified flue gases, according to the equipment SO₂ removal efficiency.

Absorber tank energy balance (Eq. 3) is built to find the cooling water mass flow rate $\dot{m}_{water(14)}$:

$$Q = 0 = \xi \Delta H^0_{reaction}(T_0) + \sum_{i=in} \int_{T_i}^{T_0} \dot{m}_i c_{pi} dT_i + \sum_{j=out} \int_{T_0}^{T_j} \dot{m}_j c_{pj} dT_j \quad (3)$$

Model considered the absorber tank surfaces to be adiabatic ($Q=0$). The adopted strategy was to force the process to operate at controlled tank bulk temperature T_0 and fixed cleaned flue gas flow output temperature $T_{flue\ gas-out(17)}$. The scrubbing process (Eq. 1) is exothermic, and though its enthalpy of reaction $\xi \Delta H^0_{reaction}$ is positive, with ξ the extent of reaction ($\xi=1$ for full sulfur conversion).

Reagent preparation - Calcium carbonate CaCO₃ (limestone) is admitted (stream 5) at the ball mill to be crunched and then send to the limestone slurry tank to become the scrubbing agent, injected in the absorber tank (stream 10). Water to that process is recovered from the gypsum dewatering treatment (streams 26 and 34).

Dewatering - Calcium sulfate dehydrate (stream 19) is pumped from the absorber tank to the hydrocyclone (stream 20) to start the 1st step towards the gypsum drying, which ends at stream 24. Hydrocyclone underflow (stream 21) is processed at a vacuum filter to be then (stream 24) send to its final destination, the gypsum storehouse storage. The rejected product from the vacuum filter (stream 23) joins the same class of waste rejected from the hydrocyclone overflow (stream 22).

3. FGD ENERGY PENALTIES AND MODEL PARAMETERS

FGD contribution to energy expenditure on a power plant comes from mechanical work from pumps, fans, mills, and other auxiliary equipment, as listed in Tab. 1.

Table 1. FGD mechanical drivers and their position on the plant according to Fig. (1)

#	Equipment	Upstream	Downstream
1	Flue gas fan	1	2
2	Ball mill	5	6
3	Water pump	7	8
4	Make-up water pump	13	14
5	Limestone slurry pump	9	10
6	Oxidation compressor	11	12
7	Recycle pump	15	16

8	Gypsum bleed pump	19	20
9	Gypsum hydrocyclone	20	22
10	Vacuum filter pump	25	26
11	Waste hydrocyclone	30	31-32
12	Waste water pump	29	30
13	Gypsum slurry pump	27	28
14	Treated water pump	33	34
15	Limestone slurry stirrer	Limestone slurry tank	
16	Slurry stirrer	Absorber	
17	Recovered slurry tank stirrer	Gypsum slurry tank	

Total power requirement \dot{W}_{FGD} to run the FGD plant could be expressed by the summation of all individual demands described in Table 1, given by (4):

$$\dot{W}_{FGD} = \sum \dot{W}_{pump} + \dot{W}_{fan} + \dot{W}_{mill} + \dot{W}_{stirrer} + \dot{W}_{hydro} \quad (4)$$

All devices are electrically driven, and total power is a negative demand to be deducted from total plant output. As a general relation, electrical power demand \dot{W}_{fm} for fluid machines as pumps and fans was calculated from:

$$\dot{W}_{fm} = \frac{\dot{m} v \Delta P}{\eta_{fm}}, \quad (5)$$

with \dot{m} is the fluid mass flow rate (kg/s), v is the fluid specific volume (m³/kg), ΔP is the flow pressure difference (kPa), and η_{fm} is the conceptual or average equipment conversion efficiency.

Operational data for limestone milling were quite hard to find, and a combination of separate and independent information was taken as a first approximation. Limestone consumption was based on actual data from the Pego power plant in Portugal (Alves, 2013), reported as 75.335 ton/year. The HJ Crusher catalog (Pershing and Wendt, 1977) indicated that their product power demand was 155 kW for a range of 4.5 to 12.0 ton/year. A specific energy consumption e_L was then estimated as 64.8535 kJ/kg, and milling power $\dot{W}_{milling}$ (kW) was then expressed by

$$\dot{W}_{milling} = \dot{m}_L e_L \quad (6)$$

with \dot{m}_L is the limestone mass flow input at stream 5.

Three stirrers are needed to operate the system, placed at the absorber tank, the limestone slurry tank and the gypsum slurry tank. The absorber tank power was modeled as an ideal Continuous Stirred Tank Reactor CSTR, whose volume V_{CSTR} (m³) was defined (Perry, 1997) as:

$$V_{CSTR} = \dot{V}_{fg} x_{SO_2} t_{res}, \quad (7)$$

with \dot{V}_{fg} standard flue gas volumetric flow rate (m³/s) and x_{SO_2} the flue gas molar fraction of SO₂. According to Perry (1977), the reaction residence time t_{res} was calculated to be 12 s. The stirrer power demand \dot{W}_{CSTR} (kW) was then calculated by:

$$\dot{W}_{CSTR} = N_{po} \bar{\rho} N^3 D^5 / 1000 \quad (8)$$

with N_{po} the power number (dimensionless), $\bar{\rho}$ the average fluid density (kg/m³), N the propeller angular velocity (rps) and D its diameter (m). A maritime propeller was selected, with a correspondent N_{po} of 0.35 (turbulent regime) and a propeller to tank diameter ratio of 1/3. The average density $\bar{\rho}$ was calculated after the flue gas, water and slurry density values (Perry, 1977; Alves, 2013).

The limestone and the gypsum slurry tanks were modeled after the definition of the power to volume ratio \dot{W}_{PVR} , assumed to be an average and constant value of 0.1261 kW/m³, collected from industrial data. Tank volumes were estimated based on the residence time of each of the processes.

Hydrocyclone power demand \dot{W}_{hydro} was once more based on private communication with the operational staff of ENGIE¹, which operates these devices with an average downstream to upstream stream mass flow ratio of 23% to 77%. Energy requirement was calculated with the aid of Eq. (5) for both streams.

¹ TRACTEBEL/ENGIE Charqueadas power plant

The FGD model inputs and operational parameters were the flue gas mass flow rate $\dot{m}(4)$, temperature $T(4)$ and SO_2 fraction $x_{\text{SO}_2}(4)$, as key information. The operational bulk temperature in the absorber tank T_0 and the flue gas temperature at the heat exchanger discharge $T(18)$ were also prescribed. Two major outputs were selected, among many others, to characterize the FGD energy penalty to the power plant: the work ratio R_W and the generation impact, given by:

$$R_W = \frac{\dot{W}_{FGD}}{\dot{V}_{fg}(4)} \left[\frac{kW}{m_N^3/s} = \frac{kJ}{m_N^3} \right] \quad (9)$$

$$I_G = \frac{\left(\frac{\dot{W}_{FGD}}{1000} \right)}{C} 100[\%] \quad (10)$$

with C the plant net power output (MW).

4. FGD MODEL VALIDATION

Three types of coal were selected to test the model performance, presented in Tab. 2.

Table 2. Coal description

	Relative coal composition, dry base (%)						Flue gases, wet base (kmol)			
	C	H	O	S	N	Ashes	CO2	H2O	N2	SO2
Coal 1 (Fallavena, 2011)	35.29	2.20	1.46	10.56	0.60	49.89	2.468	1.805	11.920	0.277
Coal 2 (Marreco, <i>et al.</i> , 2006)	45.05	2.97	8.50	6.34	0.65	36.49	3.151	2.126	13.980	0.166
Coal 3 (Kenney, <i>et al.</i> , 2011)	54.81	3.74	14.52	0.28	0.79	25.86	3.833	2.447	15.96	0.007

Coal type 1 is a low rank, with high sulfur and ashes contents, from South Brazil (Fallavena, 2011), where most of the coal power plants are placed in the country. Coal type 3 is a high rank, low sulfur and ashes contents, reported by (Kenney, *et al.*, 2011). Coal type 2 was chosen due to its intermediate composition (Marreco, *et al.*, 2006).

Table 3 shows the FGD model outputs when submitted to four input scenarios. Coal quality was changed for the first three scenarios, according data on Tab. 2, while the last one considered differences on equipment efficiency. The flue gas flow rate was calculated using the ideal gas relations, and plant capacity was calculated for 100 kg/s fuel consumption.

Table 3. Work ratio R_W and generation impact I_G for different operational scenarios, with $\eta_{FGD} = 98\%$, flue gas input temperature $T(4) = 140.0^\circ\text{C}$ and output temperature $T(18) = 62.5^\circ\text{C}$

	Scenarios			
	1	2	3	4
Plant net power C (MW)	405	510	740	740
Flue gas flow rate (m^3/h)	1.33E+06	1.57E+06	1.79E+06	1.79E+06
Coal type – Tab. 2	1	2	3	3
Equipment efficiency (%)*	65.00	65.00	65.00	85.00
\dot{W}_{FGD} (kW) (Eq. 4)	8710	8475	7586	5580
R_W (kJ/m^3) (Eq. 9)	23.58	19.43	15.26	11.68
I_G (%) (Eq. 10)	2.15	1.66	1.02	0.75

*pumps, fans, stirrers and compressor

Values for R_W and I_G followed an expected and logical behavior, as both decreased from low to high quality coal types, and efficiency improvement. Next validation step was performed by comparing data from five coal fired power plants with WFGD systems, presented in Tab. 4.

Table 4. Output data from coal fired power plants with WFGD systems

	Tsumita, <i>et al.</i> , 2007	Sargent, 2002	Gansley, 2008	Wu, <i>et al.</i> , 2017	Feng, <i>et al.</i> , 2014
	Plant 1	Plant 2	Plant 3	Plant 4	Plant 5
Plant net power (MW)	600	500	600	1200	600
Flue gas flow rate (m ³ N/h)	2.44E+06	2.97 E+03	3.57E+06	-	2.11E+06
Flue gas input temperature $T(4)$ (°C)	127.0	148.8	-	-	-
Flue gas output temperature $T(18)$ (°C)	-	54.4	-	-	-
Sulfur content	-	high	High	-	-
Equipment efficiency (%)	-	85	87 - 90	-	-
FGD efficiency η_{FGD} (%)	-	98	98	-	-
FGD total power demand (kW)	6840	9056	11464	7204	6150
Work ratio R_W (kJ/mN ³)*	10.09	10976.97	11.57	13.84	10.48
Generation impact I_G (%)*	1.14	1.81	1.91	0.60	1.02

* Calculated by the authors of the present work according to Eq. (9) and Eq. (10).

Data are irregularly available and trustful, but the ones selected in Tab. 4 allowed to calculate an average work ratio R_W of 11.49, excluding Plant 2 due to data inconsistency, which is approximate to the values calculated by the model for high ranked coals in Tab. 3. The average generation impact I_G was found to be around 1.3, whereas the one calculated by the model was near to unity for the same kind of coal.

These numbers point out that the FGD model answered in a logical way with values that can be compared to the ones from literature for high ranked coals.

The first scenario in Tab. 3 was chosen to present a more detailed view of the equipment energy demand predicted by the FGD model, displayed in Tab. 5.

Table 5. Equipment energy demand for Scenario 1, presented in Tab. 3

Equipment	Energy demand (kW)	Relative impact (%)
Pumps	4341.50	49.84
Fans	1606.00	18.44
Air compressor	1255.00	14.41
Absorber tank stirrer	1211.00	13.90
Mill	275.20	3.16
Gypsum stirrer	12.51	0.14
Limestone slurry stirrer	9.22	0.11
Total:	8710.43	100.00

Results on Tab. 4 were ranked by importance, and its easy to be noticed that the summation of the power demand from 10 pumps reached almost 50% of the total system charge. Air transport by fans and compressed air corresponded to almost 33%, followed by the absorber tank stirrer.

5. CONSTRUCTION OF A FGD ENERGY PENALTY FUNCTION (EPF)

A Design of Experiment (DoE) approach (Montgomery, 1997) was chosen to investigate the model behavior submitted to given range values of selected independent inputs, called control parameters. Table (6) presents the FGD model control parameters with their ranges.

Table 6. FGD model control parameters with their ranges selected for the DoE project

	High bound	Intermediate	Low bound
Flue gas input temperature $T_{in}=T(4)$ (°C)	120.0	140.0	160.0
Flue gas output temperature $T_{out}=T(18)$ (°C)	45.0	62.5	80.0
FGD efficiency η_{FGD} (%)	97.0	98.5	100.0
SO ₂ molar fraction $x_{SO_2}(4)$	0.0003	0.0085	0.0168

A Box-Behnken procedure (Lorenzen and Anderson, 1993) was used to design a controlled sequence of simulations, willing to capture the model capacity to respond to that range of input control parameters. The objective was to build an algebraic function that enables to calculate both the work ratio R_W and the generation impact I_G , that would substitute the original FGD model. Table (7) presents the operational details calculated and selected for the Box-Behnken procedure.

Table 7. Box-Behnken operational details for the FGD assessment

Number of factors k	4	Replication	1
Number of essays	27	Total number of essays N	27
Number of blocks	1	Central points C_o	3

The designed number of essays N was calculated by (Ferreira, *et al.*, 2007):

$$N = 2k(k - 1) + C_o \quad (11)$$

with k the number of factors and C_o the number of central points. The lack of uncertainty data of the control parameters lead to a unitary number of replications, and a total number N equal to the number of essays. Box-Behnken analysis were performed with the aid of (MINITAB), and the first assessment was performed to identify the behavior of the work ratio R_W in respect each of the controlled parameters, as displayed in Fig. 2.

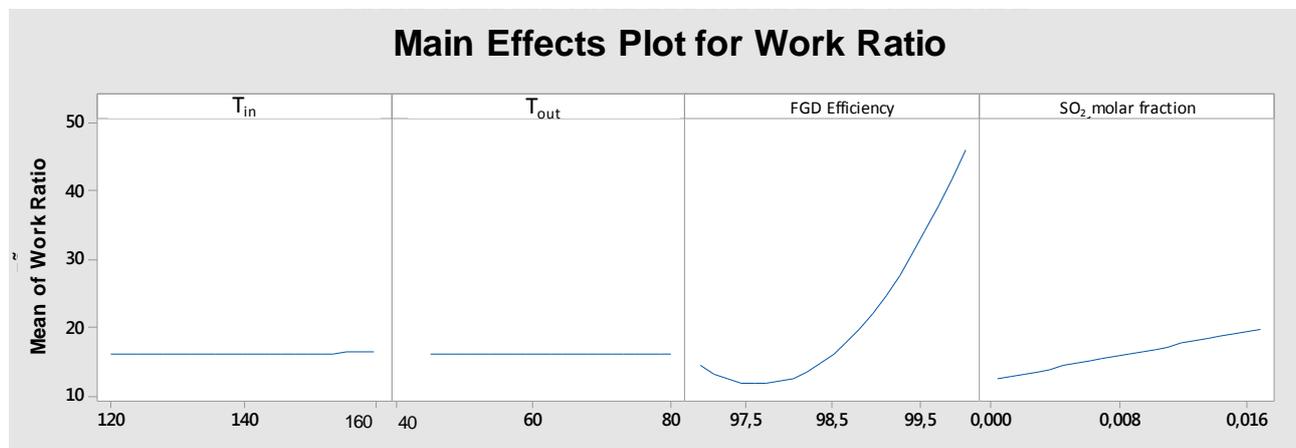


Figure 2. Main effects of the controlled parameters on the Work Ratio R_W

The work ratio R_W varies significantly with sulfur removal efficiency. As already observed before on the present work, the increase on sulfur removal efficiency leads to a greater amount of required power by volume of flue gases. It was also noted that R_W is linearly dependent on SO₂ fraction. Flue gas temperatures do not affect the work ratio R_W , and their contribution will be transformed in constant values for the next assessments. Combined effect analysis showed some iteration among sulfur removal efficiency and sulfur content, but without any significant value for the other combination of parameters, which won't be presented in this paper.

Next step of the DoE assessment was to build the FGD Energy Penalty Function EPF, an algebraic expression for R_W as a function of its controlled parameters (Eq. 12), based on data from Tab. 6.

$$R_W = 59751.2 + 0.01175 T_{in} - 0.0048 T_{out} - 1223.54 \eta_{FGD} + 247.7 x_{SO_2} + 0.000019 T_{in} * T_{in} + 0.000004 T_{out} * T_{out} + 6.26444 \eta_{FGD} * \eta_{FGD} - 165.3 x_{SO_2} * x_{SO_2} - 0.000083 T_{out} * \eta_{FGD} + 2.02 \eta_{FGD} * x_{SO_2} \quad (12)$$

Results from Eq. (12) for R_W were compared to the ones obtained from the FGD model, and deviations are displayed in Fig. 3.

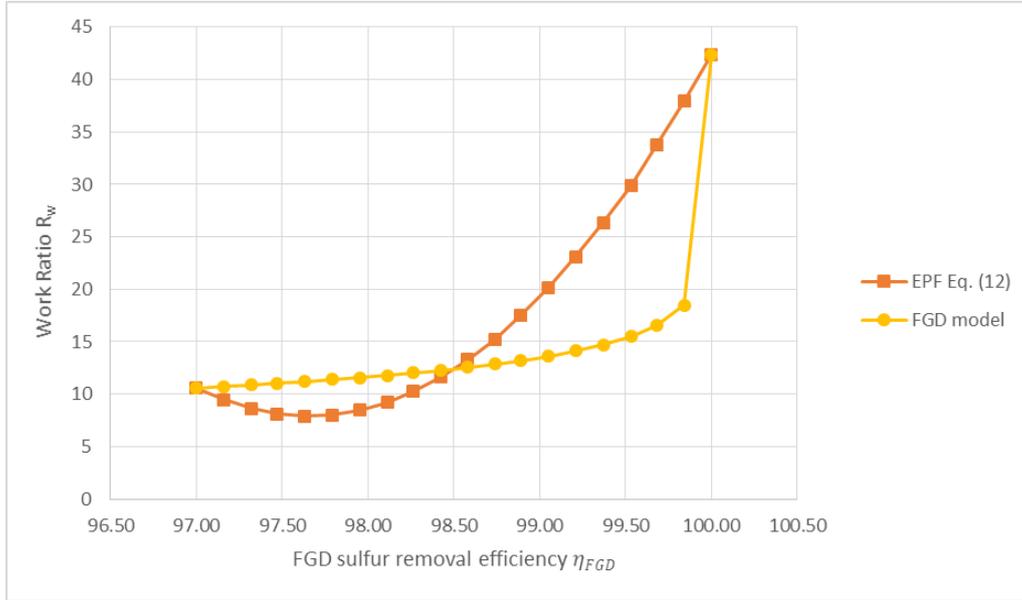


Figure 3. Predicted work ratio R_W from the FGD model and by the EPF Eq. (12).

The magnitude of the deviations from the EPF results in comparison to the FGD reference model were far too high, making it impossible to use the function. Based on the knowledge and experience with regression functions, a further assessment was made based on the restriction of the upper limit values of the controlled parameters. Many crossed combinations were performed with upper limits of efficiency and SO_2 fraction to arrive to a final EPF based on sulfur removal efficiency η_{FGD} ranging from the original 97.0% up to 99.5%, instead of 100%, displayed in Tab. 6. The new EPF is presented in Eq. (13):

$$R_W = 5762.73 - 118.898 \eta_{FGD} + 232.01875 x_{SO_2} + 0.6144 \eta_{FGD} * \eta_{FGD} - 183.7 x_{SO_2} * x_{SO_2} + 2.182 \eta_{FGD} * x_{SO_2} \quad (13)$$

Flue gas temperatures T_{in} and T_{out} were condensed in the first term of Eq. (13), as the EPF is insensitive to their variation. The sulfur removal efficiency η_{FGD} limited to 99.5% allowed for a much better fit of the penalty function, with an maximal deviation of 1.86% along the complete range of values. Figure 4 presents the successive fitting trails performed with η_{FGD} upper limit of 100.00%, 99.99% and 99.50%.

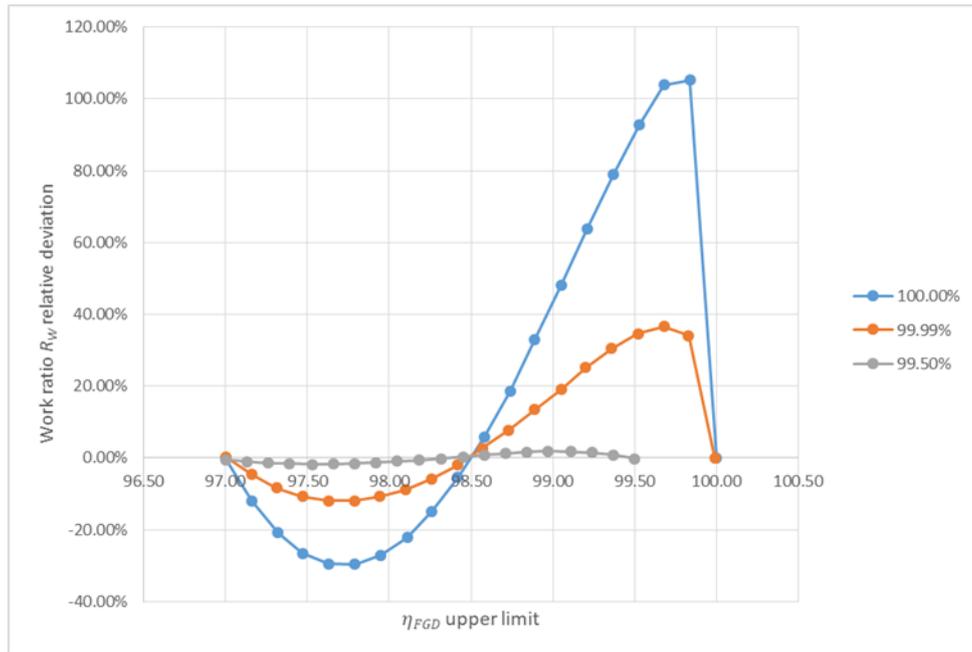


Figure 4. Work ratio R_w relative deviation calculated by the Energy Penalty Function EPF in respect to FGD model results for η_{FGD} upper limit of 100.00%, 99.99% and 99.50%

The intermediate EPF prior to Eq. (13) were quite similar in their essence, but the last one was only one capable to copy the FGD model results, with a narrow uncertainty or deviation, besides its shorter number of elements. Equation (13) can be used independently of the original FGD model in any kind of coal fired system to predict the power to volumetric flue gas flow ratio, as long as the limits of the controlled parameters are respected.

6. CONCLUSION

The present work complements the prior one from Tomazi, *et al.* (2017), which developed a Wet Flue Gas Desulfurization WFGD model based on mass, species and energy balances. That model can be run as a stand-alone program or linked to a larger simulation system written for the Engineering Equation Solver EES software. An alternative option was proposed here in order to develop an independent routine to be used in any kind of simulation platform allowing to calculate the amount of energy needed to remove sulfur from flue gas streams from combustion processes. That independent routine was called the Energy Penalty Function EPF.

The Box –Behnken method was chosen to build the EPF capable to summarize the WFGD process in a simple and easy to use expression. The method also helped to identify the relevant input parameters of the desulfurization process that enabled to calculate the energy penalty. First exploration pointed out the role of flue gas input and output temperatures, sulfur content and filtering efficiency. Further exploration showed that these temperatures were of little relevance, and the final Energy Penalty Function was proposed for the sulfur content ranging from 0.03% to 1.68 % in molar basis and filtering efficiency ranging from 97.0% to 99.5%.

The Energy Penalty Function main output was proposed to be the work ratio R_w , defined as the ratio of the expended energy in kJ for sulfur removal to the flue gas volume in m_N^3 . Results from the work ratio displayed a maximum relative deviation of 1.86% in respect to the detailed model, for the desulfurization efficiency interval of 97.0% to 99.5%, and an overall maximum deviation of 1.41% in respect to the detailed model.

7. ACKNOWLEDGEMENTS

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