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Full-scale evaluation of SO_2 capture increase for semi-dry FGD technology

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A R T I C L E I N F O

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A B S T R A C T

This paper reports on a full-scale investigation of a possible increase of the SO_2 capture ratio in current semi-dry flue gas desulphurization (FGD) technology. The FGD unit is used for two 130 t/h steam PC boilers burning a blend of lignite and hard coal with dry ash-free sulphur content at 1.83%. The FGD unit has been designed to reach an SO_2 emission limit of 1350 mg/Nm^3. The aim of the experimental work presented in this paper was to investigate the possibility of reaching SO_2 emission targets of 500 and 200 mg/Nm^3. The investigation sought to determine the real correlation of SO_2 capture ratio with Ca/S and with the difference of dry-bulb temperature and dew point in the absorber Δt_{AD}. Generally, the SO_2 capture correlation with Ca/S has flat characteristics at a capture ratio >90%, which is required to reach the 200 mg/Nm^3 target. In this case, lowering the Δt_{AD} in the absorber has only a weak effect. The 500 mg/Nm^3 target requires an SO_2 capture ratio of about 80%; in this case lowering the Δt_{AD} by about 7 °C increases the SO_2 capture by 10% points at the same Ca/S ratio.

1. Introduction

Recent years have seen emission limits strengthened for various pollutants, including sulphur dioxide. On the national level, the first stage of reducing the emission limits is determined by current national legislation [1] that set new emission limits from 1/1/2016 for combustion sources >50 MWt and from 1/1/2018 for <50 MWt. However, the strengthening of emission limits goes beyond national legislation. At European level, a new document, BREF LCP within the Industrial Emission Directive (IED) [2], was adopted in 2017, and the new emission targets following the Best Available Techniques (BAT) is to be implemented by 2021. This of course brings significant demands on energy providers, who have launched various approaches to improving the performance of current technologies rather than decide on new investments. This is particularly true in the case of flue gas desulphurization (FGD) units that were originally designed for a certain SO_2 capture ratio and are still able to operate beyond their original design values. This paper presents an investigation comprising full-scale experimental research on SO_2 capture increase potential for semi-dry FGD technology.

2. Semi-dry FGD

Semi-dry FGD technology is commonly used for SO_2 capture from flue gas. Among the direct additive technology for fluidized bed boilers and the wet scrubbing FGD for large-scale boilers and high sulphur fuels, the semi-dry FGD is typically used for medium size boilers combusting low to medium sulphur containing coal [3]. The core of this technology is typically a spray absorber, where a water suspension of calcium hydroxide (so-called “lime milk”, produced by the reaction of a quicklime with water) acts as an SO_2 capture sorbent. The reaction mechanism is as follows:

$$SO_2 + Ca(OH)_2 \rightarrow CaSO_3 + H_2O \tag{1}$$

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$$SO_2 + Ca(OH)_2 + 0.5O_2 \rightarrow CaSO_4 + H_2O \tag{2}$$

$$SO_3 + Ca(OH)_2 \rightarrow CaSO_4 + H_2O \tag{3}$$

The resulting product is a mixture of $CaSO_3$ and $CaSO_4$ hydrates, so it cannot be further reused and must either be treated as a waste product [4] or can be further treated or upgraded [11].

The SO_2 capture ratio is affected by two particular parameters – the Ca/S ratio, defined as mol of Ca in the sorbent per mol of S in the fuel, and the temperature difference between the dry-bulb temperature and the flue gas dew point in the spray absorber, further marked as Δt_{AD}. The key point is that a higher relative humidity, i.e. lower Δt_{AD}, significantly affects the rate of the reaction (1), approximately as per the following equation [5]:

$$k_{(1)} = 0.4 + 0.0064RH \tag{4}$$

Consequently, a smaller Δt_{AD} improves the SO_2 capture ratio. Nevertheless, an important limitation is that a condensation must not take place, i.e. complete flow of water into the spray absorber must be evaporated [5–7,12]. The SO_2 capture ratio is defined as follows:

$$\eta = \frac{c(SO_{2,in}) - c(SO_{2,out})}{c(SO_{2,in})} \tag{5}$$

The correlation of the sorbent stoichiometry and the SO_2 capture ratio can be described by following equation [8]:

$$\eta = 1 - \exp(-K.Ca/S) \tag{6}$$

Equation (6) allows calculation of the Ca/S ratio for a required SO_2 capture. The typical disadvantage is necessity for knowledge of K, which depends on various parameters – mostly on sorbent properties (e.g. particle size, reactivity) and flue gas composition. No explicit correlation of K with these parameters is available, however. Thus, the value of K must be determined experimentally for the specific conditions.

3. Experiments

The experimental work started with identification of the current state of operation of the semi-dry FGD. It is an ABB Fläkt design, delivered in 1998. The technology was designed for two 130 t/h steam PC boilers originally built for lignite coal combustion, now burning a coal blend of 70% lignite and 30% hard coal. This FGD unit now operates at an SO_2 emission target of 1350 mg/Nm^3 at 6% reference oxygen, which corresponds to approximately 55% SO_2 capture ratio. The aim of the experiments was to investigate the absorber's performance at the target SO_2 concentration of 500 and 200 mg/Nm^3. The selection of the emission target of 500 mg/Nm^3 is based on the National Transition Plan for emission limits before 2021 and the target of 200 mg/Nm^3 is based on the proposed limits that will be valid from 2021 [2], which will fall into the range of 150–360 mg/Nm^3. For this purpose, a series of long-time duration experiments was carried out, modifying the Ca/S ratio and Δt_{AD} in the spray absorber.

3.1. Current operation conditions of the semi-dry FGD unit

At the nominal boiler load 130 t/h, typical operation parameters of the FGD unit are given in Table 1. Flue gas flow and inlet water flow values are referenced to 1 kg of burned fuel. Some of the data was measured directly – typically temperature, concentration and pressure. The rest of the data comes from the operation parameters of the boiler.

It was not possible to measure the Ca/S ratio directly in the process. The value of Ca/S was obtained based on balancing the lime milk batch preparation process. The measured values in this process are:

- mass flow of fresh quicklime CaO
- mass flow of feed water
- dry matter fraction in the lime milk

Table 1
Operation parameters of the semi-dry FGD unit.

Parameter	Value	Unit
Flue gas temperature inlet	165.3	°C
Temperature in the spray absorber	83.3	°C
Pressure in the spray absorber	99.91	kPa
Flue gas flow inlet	7.836	Nm^3/kg$_{fuel}$
Flue gas flow outlet	8.777[a]	Nm^3/kg$_{fuel}$
Water vapour partial pressure	13.3	kPa
Dew point	51.5	°C
SO_2 concentration inlet	3051	mg/Nm^3, 6% O_{2ref}
SO_2 concentration outlet	1354	mg/Nm^3, 6% O_{2ref}
O_2 concentration inlet	8.99	% vol.
O_2 concentration outlet	9.80	% vol.
$Ca(OH)_2$ inlet	478	kg/h
Water inlet	0.336	kg/kg$_{fuel}$

[a] The outlet flow, compared to the inlet flow, additionally includes evaporated water from the lime milk and false air intake. The air intake was evaluated based on measuring the oxygen concentration difference between the inlet and outlet of the absorber.

These values provide the molar flow of Ca into the spray absorber at an approximate hourly average. Better precision cannot be achieved in the real scale since the lime milk is prepared batch wise in stock for a longer time of operation. The sulphur molar flow was obtained from the boiler process data and SO_2 concentration measurement at the inlet to the absorber. The dew point in Table 1 was calculated from boiler process data and coal parameters. Further experimental validation of this numerical approach is provided in Chapter 4. The average parameters of the fuels and final blend are summarized in Table 2a, and the sorbent properties in Table 2b.

The boiler was operated at power load of 130 t/h at an average thermal efficiency 90.1%. Based on these data and values from Tables 1 and 2, the FGD process parameters are as follows:

- Ca/S ratio: 1.52 mol/mol
- Δt_{AD}: 32.1 °C

3.2. Experiments at target SO_2 concentrations of 500 and 200 mg/Nm^3

The experiments were carried out during a standard operation of the boiler and the FGD unit. The control system of the FGD unit was modified to be able to keep the desired SO_2 concentrations by allowing the operator to manually set the amount of injected sorbent (i.e. change the Ca/S ratio) and to change the temperature in the spray absorber by adjusting the flow of injected water. The first phase at the target 500 mg/Nm^3 took four days and second phase at the target 200 mg/Nm^3 took eight days. The second phase included measurements at two different Δt_{AD}, each for four days. The change of Δt_{AD} was achieved by operating the spray absorber at two different achievable temperatures – 82 and 77 °C – modifying the water flow into the absorber.

In the identification of the FGD operation, the partial pressure of water vapour and the dew point were calculated based on material and energy balance. During the experiments, this numerical approach was validated by experimental determination of the flue gas dew point, using wet and dry bulb measurement at the inlet and outlet of the FGD spray absorber. For this purpose, a K-type thermocouple wrapped in a fabric cover saturated in distilled water was prepared. The final wet-bulb temperature was considered as a mean value of the stable section of the time series measured, as shown here in Fig. 1.

The dew point was determined by the following procedure: first, the saturated pressure of the water vapour using the wet-bulb temperature was obtained, based on Antoine's equation:

$$\ln\left(p_w^0\right) = A - \frac{B}{(t_{Bw} + 273.15) + C} \tag{7}$$

with constants A = 16.2886, B = 3816.44 and C = −46.13 [9]. Next, the partial pressure of the water vapour was calculated from the dry- and wet-bulb temperatures, using the following equation [10]:

$$p_w = p_w^0 - A_p \cdot (t_A - t_{Bw}) \cdot p \tag{8}$$

Finally, the dew point was obtained using p_w calculated by Equation (8) back in Equation (7) as p_w^0. The Δt_{AD} can be then obtained as follows:

$$\Delta t_{AD} = t_A - \frac{B}{A - \ln(p_w)} - C + 273.15 \tag{9}$$

Table 2a
Fuel parameters; average blend is 70% lignite and 30% hard coal by weight.

Parameter	Lignite coal	Hard coal	Blend	Unit
LHV	15.3	17.3	15.93	MJ/kg
A^d	22.5	42	28.8	%
W^r	28.5	13	23.8	%
S^{daf}	2.10	1.21	1.83	%

Table 2b
Quicklime properties.

Parameter	Value	Unit
mean particle size	30	μm
reactivity t60	1.1	min
fraction of CaO + MgO	95.1	% wt.
BET surface	2.697	m^2/g

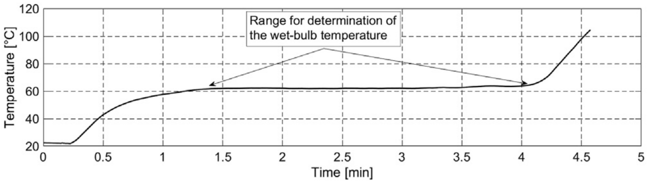

Fig. 1. Example of a time record of wet-bulb temperature measurement from the time of insertion of the thermometer into the flue gas till the wrapping dried out.

To evaluate the conditions in the absorber, the t_A and t_{Bw} were measured at the outlet from the absorber. Correction for dew point increase by SO_3 presence was not taken into account. It is assumed that in the environment of the absorber, all gases able to form mineral acids immediately react with the lime milk and therefore do not have any effect on the dew point. As for SO_3, the same principle is valid for HCl and HF if present in the flue gas. Illustration of measurement points of the t_A, t_{Bw} and flue gas composition are shown in Fig. 2.

4. Results and discussion

The inlet and outlet SO_2 concentration record over the whole 14-day period of the experiment is shown in Fig. 3; Fig. 4 shows the SO_2 concentration at the absorber output alone, giving a better resolution. On day 3 of the experiment, the change of the emission targets achieved by increasing the Ca/S ratio are easily identifiable. On days 8–9, a decrease of Δt_{AD} by increasing the water flow with parallel decrease of Ca/S was made, keeping the outlet SO_2 level constant. No significant change can be seen in the outlet SO_2 level during this change (see also Table 4). This indicates very smooth transitions between operation modes with different Δt_{AD}.

The SO_2 concentration records show significant fluctuations at the input to the FGD absorber in Fig. 3. These fluctuations are evaluated in Table 5 in more detail as standard deviations from a mean value. They are primarily caused by natural fluctuations of the sulphur content in both the sorts of coal in the blend, which can make up to ±30% of the values in Table 2a. The second contribution is the preparation of the coal blend itself, which was conducted manually (and therefore not always precisely from the point of weight ratio), blended by a digger on site directly before feeding into the boiler's coal silo.

4.1. Dew point validation

Successful validation of the dew point calculation using the wet and dry-bulb measurement is the key factor in assessing the reliability of overall desulphurization balance in the spray absorber. An overview of the calculation and measurement of the dew point is summarized in Table 3. The validation was carried out during normal operation mode of the absorber before the experiments commenced.

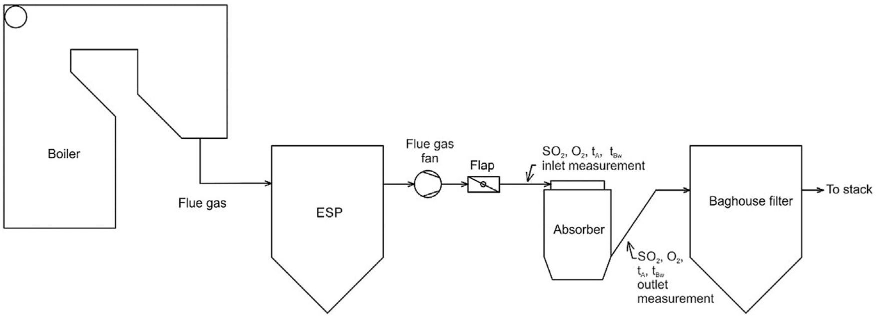

Fig. 2. Scheme of the flue gas flow and measurement points.

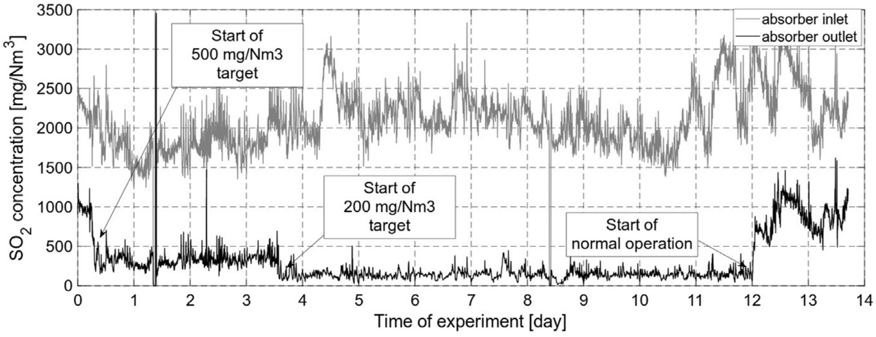

Fig. 3. Time series of the SO_2 concentration.

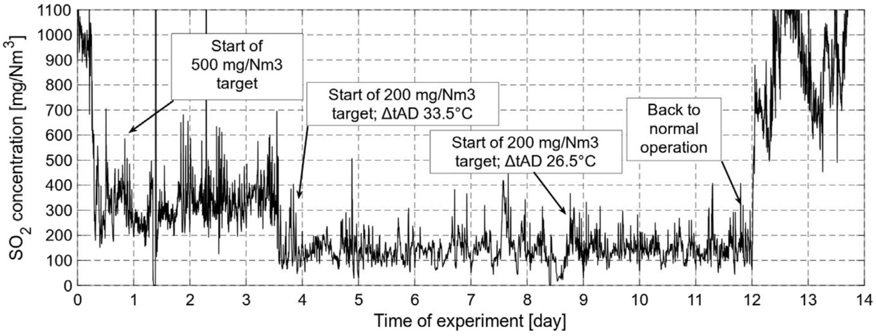

Fig. 4. Time series of the SO_2 concentration – details of absorber output.

Table 3 shows that the difference in dew point calculated from the material and energy balance, and the measured value is very good. Thus the calculated value can be considered as validated. The larger relative difference between the measurement and calculation at the inlet to the absorber may have been caused by the wet-bulb measurement, where a possible dissolving of the flue gas components (particularly SO_2) in the distilled water in the thermocouple wrapping can occur, which can in turn affect the rate and temperature of evaporation. Nevertheless, the dew point at the outlet is more important for the absorber balance, where only 1.6% relative difference between measurement and calculation was reached.

Table 3
Validation of the dew point.

Parameter	Absorber inlet	Absorber outlet	Unit
wet-bulb temperature t_{Bw}	52.8	53.5	°C
dry-bulb temperature t_A	164.0	82.1	°C
p_w^0	14.14	14.50	kPa
p_w	6.9	12.60	kPa
dew point – measured	38.7	50.4	°C
dew point – calculated	42.5	49.6	°C
relative dew point difference	+9.8%	−1.6%	

4.2. Desulphurization performance

Table 4 contains the overall results as daily averages, showing the average Ca/S ratio, SO_2 capture ratio and Δt_{AD}. Table 5 shows statistics for the desulphurization performance and contains input and output (i.e. before and after the FGD) SO_2 concentrations as mean values and standard deviations, as well as evaluation of reliability of achievement of the given emission target. The table contains hourly averages. The results in Table 5 should be considered with approximately ±60 mg/Nm^3 error, which is the measurement error of the analytic technique used for SO_2 concentration measurement. The parameter "number of averages" refers to the number of measured hourly averages within the given period; the "number of satisfactory" is a count of the hourly averages that fulfil the given emission limit with 10% confidence. The reliability is calculated as percentage of satisfactory values out of the total number of the hourly averaged concentrations.

Besides the effect of Ca/S change, Table 4 shows the significant effect of the Δt_{AD} on the SO_2 capture ratio. To reach the SO_2 target of 200 mg/Nm^3 at $\Delta t_{AD} = 33.5$ °C (normal operation), a Ca/S = 2.72 is needed. Whereas for the $\Delta t_{AD} = 26.5$ °C, the needed Ca/S ratio is 2.50. Based on this data, the correlation of Ca/S and SO_2 capture ratio can be constructed using Equation (6). This is shown in Fig. 4. The values of K from Equation (6) for mean values of Δt_{AD} are shown in Table 6.

Table 4
Summary of results, daily averages.

	Day	Ca/S [mol/mol]	SO_2 capture [%]	Δt_{AD} [°C]
SO_2 target 500 mg/Nm^3	1	2.00	80.8	34.1
Δt_{AD} as in normal operation	2	2.09	78.9	34.3
	3	1.87	83.0	33.9
	4	2.28	83.2	32.9
SO_2 target 200 mg/Nm^3	5	2.82	93.4	33.5
Δt_{AD} as in normal operation	6	2.50	93.4	33.3
	7	2.82	93.1	34.0
	8	2.73	91.9	33.3
SO_2 target 200 mg/Nm^3	9	2.34	90.4	26.2
Δt_{AD} lowered by ~7 °C	10	2.40	91.9	26.3
	11	2.69	91.9	26.6
	12	2.65	93.5	26.7
Operation at 1350 mg/Nm^3	13	0.76	61.4	25.9
Δt_{AD} back as in normal operation	14	0.69	58.7	27.1
	15	0.84	46.9	31.4
	16	0.87	47.2	34.1
	17	0.94	46.7	34.8
	18	0.98	48.3	34.6

Table 5
Hourly average statistics of the desulphurization performance; all concentrations in mg/Nm^3, 6% O_{2ref}.

Parameter	SO_2 target 500 Δt_{AD} normal	SO_2 target 200 Δt_{AD} normal	SO_2 target 200 Δt_{AD} lowered by ~7 °C
SO_2 input mean	2410	3083	2704
SO_2 input standard dev.	93	106	109
SO_2 output mean	470	215	198
SO_2 output standard dev.	72	46	43
number of averages	73	98	89
number of satisfactory	68	57	66
reliability [%]	93.2	57.1	74.2

Table 6
K values of Equation (6).

Δt_{AD} [°C]	K [−]	fit goodness R^2
33.5	0.90	0.92
26.5	1.19	0.96

Table 5 shows that reaching the emission target of 500 mg/Nm^3 was achieved with more than 93% reliability. The mean output value of 470 mg/Nm^3 is associated with 72 mg/Nm^3 standard deviation. It is noticeable that the input SO_2 concentration with 93 mg/Nm^3 standard deviation reflects the above-mentioned variability of the input. The target of 200 mg/Nm^3 differs significantly in terms of the reliability in association with the Δt_{AD}. The results clearly show that an increase of the Ca/S ratio alone leads to significantly lower reliability compared to changing the Δt_{AD}.

The graph in Fig. 5 shows that the effect of lowering Δt_{AD} is more significant at lower SO_2 capture ratios. For this specific case, the effect is interesting in terms of reaching the emission target of 500 mg/Nm^3, which corresponds to about an 80% SO_2 capture. In this case, a decrease of the Δt_{AD} by 7 °C increases the SO_2 capture ratio by 10% points without changing the Ca/S ratio. To reach the target of 200 mg/Nm^3, a capture ratio above 93% is required, where the effect of lower Δt_{AD} is significantly weaker.

Table 7 compares measured and calculated Ca/S ratios for two averaged sections of the experimental campaign with a 200 mg/Nm^3 emission target. The necessity for this analysis is due to a certain inaccuracy of Equation (6) in the region of high SO_2 capture ratios, typically above 90%. At these capture ratios, a large increase of Ca/S corresponds to a relatively small increase in SO_2 capture. Table 7 shows the measured values of the Ca/S and the calculated values of the Ca/S as per Equation (6) using K from Table 6. For each Δt_{AD}, the averages of day 5–8 and day 9–12 of the experiment are used, respectively.

Table 7 shows that the analysis generally yields less than 10% difference, which can be considered satisfactory with respect to the duration and scale of the experiments. It also shows that the Ca/S ratio is underestimated for a given SO_2 capture in the case of a smaller Δt_{AD} when applying Equation (6).

A few observations from the experiments at the SO_2 emission target of 200 mg/Nm^3 need to be highlighted. It is possible to reach this target, which corresponds to the SO_2 capture ratio >93%, even though this specific semi-dry FGD was designed for a 75% capture ratio. However, it is not possible to fully and continuously maintain this emission target, particularly on account of the inability of the technology to react to changes in SO_2 concentration in the flue gas and flue gas flow sufficiently flexibility. This limitation mainly consists in the large pre-prepared volume of lime milk, thus not allowing a flexible change of dry matter content. It is therefore not possible to use it to increase the flexibility of the Ca/S ratio control. A second limitation can be seen in Fig. 5, where only a small change needed in SO_2 capture ratio requires a large change of Ca/S – in the region >90% – where the effect of Δt_{AD} change is low.

Summarizing the achieved results, the approach proposed here to enhancing the SO_2 capture ratio is most applicable for combustion sources in the power input range 50–100 MW. The new SO_2 emission limits for newer plants using coal will be in range of 150–360 mg/Nm^3 at 6% O_2. In our model example, represented by an emission target of 200 mg/Nm^3, the required SO_2 capture ratio is approximately 93%. For the semi-wet FGD, this is can be achieved at Ca/S ~2.3 with the Δt_{AD} lowered to about 26.5 °C, which does not provide much of a buffer before condensation in cases of instantaneous process variations. However, concerning reliability, it would be much more favourable to apply Δt_{AD} lowering, instead of increasing the amount of sorbent only. These conditions should therefore be considered as a boundary case, showing the real limits of the semi-dry FGD. This means that for a larger combustion plants, i.e. above 100 MW, the enhanced semi-dry FGD

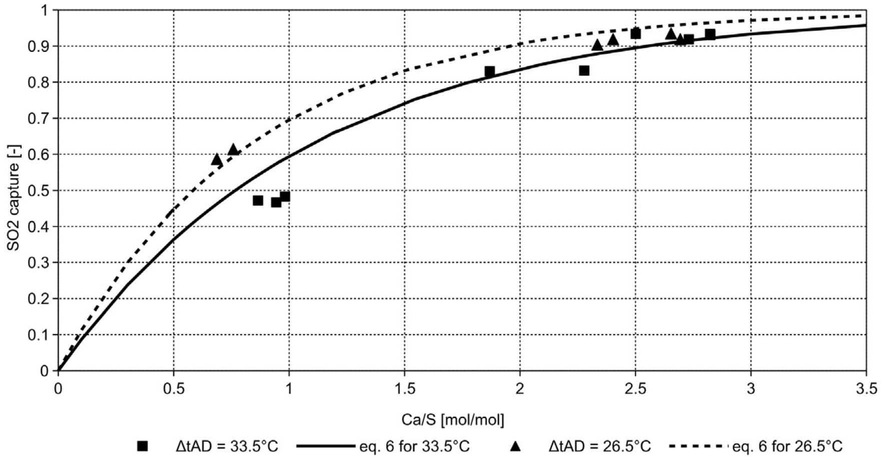

Fig. 5. Correlation of SO_2 capture ratio with Ca/S ratio.

Table 7
Measured and calculated Ca/S ratios.

Δt_{AD} [°C]	Ca/S [mol/mol]	SO_2 capture [%]	Ca/S by Equation (6) [mol/mol]	relative difference
26.5	2.34	92	2.13	−9%
33.5	2.82	93	2.95	+4.6%

is not powerful enough to fulfil the emission limits, which drop to 10–130 mg/Nm^3 for the >300 MW plants. Moreover, it is important to consider the sulphur content in the fuel. This blend of coal belongs to the low to medium-sulphur ranks of coal. High-sulphur coals would require >99% SO_2 capture ratio, which cannot be ensured using this technology.

5. Conclusions

Full scale and long-time duration experiments with a semi-dry FGD have validated theoretical considerations concerning the relationship between the stoichiometry of the sorbent and SO_2 capture ratio (Ca/S), and have shown the potential of the semi-dry FGD system to operate at significantly higher SO_2 capture ratios than those for which it was originally designed. The experiments have also proven the significant effect of setting the operating temperature of the spray absorber, which changes the Δt_{AD} – the difference between absorber temperature and the dew point. About 7 °C, Δt_{AD} is able to improve SO_2 capture by about 10% points within the SO_2 capture ratio of approximately 50–85%. The higher SO_2 capture levels should preferably be reached by lowering the Δt_{AD}, since this approach shows significantly better reliability in reaching the required emission limit in comparison with a Ca/S change only.

The experiments have also identified some weak points of this technology that need to be taken into consideration. At the required capture ratio of over 90%, the semi-dry FGD is less flexible in following changes in flue gas flow and SO_2 concentration – in principle due to the flat characteristics of the SO_2 capture-Ca/S correlation in this region. A second weak point is the impossibility of changing the concentration of the lime milk flexibly as a response to changing inlet SO_2 concentration. This is because the lime milk is prepared as batch in stock for a longer period of operation. Obvious technical limitations for this system being operated beyond the design limit are the possible overloading of lime milk preparation, as well as higher loads on transportation of FGD solid products and the larger content of unreacted sorbent in them.

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List of symbols

A^d	ash content in dry sample [%]
A_p (in Equation. (8))	psychrometric constant: 662.10^{-6} [1/K]
Ca/S	Ca to S stoichiometric ratio [mol/mol]
$c(SO_{2,in})$	SO_2 concentration in the flue gas at absorber inlet [mg/Nm^3]
$c(SO_{2,out})$	SO_2 concentration in the flue gas at absorber outlet [mg/Nm^3]
$k_{(1)}$	rate constant of the reaction (1) [mol/m^{2}s]
K	constant of Equation (8) [1]
LHV	lower heating value [MJ/kg]
p_w	partial pressure of water vapour [kPa]
p_w^0	saturated partial pressure of water vapour [kPa]
p	total pressure [kPa]
RH	relative humidity of the flue gas [1]
S^{daf}	sulphur content in dry-ash-free sample [%]
Δt_{AD}	dry-bulb temperature and dew point difference [°C]
t_A	dry-bulb temperature [°C]
t_{Bw}	wet-bulb temperature [°C]
W^r	water content in raw sample [%]
η	SO_2 capture ratio [1]

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