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Prevention of mist formation in amine based carbon capture: field testing using a Wet ElectroStatic Precipitator (WESP) and a Gas-Gas Heater (GGH)

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Abstract

This study presents the results of two field tests that aimed at evaluating two countermeasures (WESP and GGH) to avoid acid mist formation. A WESP is shown to be very efficient for the removal of nuclei from the flue gas (100 % efficient) and thus can prevent aerosol formation inside an amine based absorber. This is however only valid in the absence of SO₂ in the flue gas entering the WESP. A decreasing WESP efficiency is noted in the presence of SO₂ with increasing voltages as a result of newly formed aerosols inside the WESP. This implies that no or very low levels of SO₂ should be present in the flue gas entering the WESP. Since most of the amine carbon capture installations have a pre-scrubber (usually using NaOH to remove residual SO₂ in the flue gas leaving the power plant's Flue Gas Desulphurisation) in front of their amine absorber, the WESP must be installed behind this pre-scrubber and not in front of it. Having a Gas-Gas Heater (or any type of flue gas cooling such as a Low Temperature Heat Exchanger) installed upstream of the wet scrubbing may prevent homogenous nucleation and thus prevent the conversion of H₂SO₄ into sulfuric acid aerosols and consequently mist formation issues in the amine based carbon capture installation. Which option to choose amongst the two countermeasures presented in this study will depend on whether a new built installation is being considered or whether a carbon capture is planned as a retrofit into an existing installation.

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1. Introduction

Sulfuric acid mist emitted from industrial processes (coal fired power plants, refinery plants, chemical plants ...) has been investigated for several decades due its effect on the plume opacity and its environmental impact (i.e. acid deposits). Whether the sulfuric acid is emitted under a gaseous form or whether it is emitted as an aerosol depends very much on the flue gas treatment processes installed on-site. Sulfuric acid mist (i.e. aerosols) is usually formed in wet flue gas scrubbers (e.g. a Wet Flue Gas Desulphurisation, WFGD) where supersaturation conditions prevail. Supersaturation conditions refer to conditions in which a high temperature gradient and a high water vapor content co-exist. This supersaturation leads to homogeneous nucleation and consequently results in sulfuric acid aerosols formation. These sulfuric acid aerosols are present in high numbers ($\cong 10^8$ cm⁻³) and have very small sizes [1]. In fact, high sulfuric acid concentrations in the flue gas do not lead to a significant increase in numbers but increases the size of the aerosols; however, they remain submicron [1]. Upon entering an amine carbon capture absorber, these sulfuric acid aerosols act as amine mist precursors [2, 3, 4 and 5] and can lead to large amine emissions [6, 7, 8, 9, 10, 11 and 12] from the top of the absorber. In the recently published book: Absorption-Based Post-Combustion Carbon capture of Carbon Dioxide, a chapter is dedicated to the aerosol issue which gives many more details and insights in this topic [13].

A summary of results which enables to predict amine (Monoethanolamine, MEA) mist formation as a function of the measured particle/aerosol number concentrations present in the flue gas entering the amine scrubber and operating conditions of the Carbon Capture (CC) plant is presented in [14]. This study reveals that if particle number concentrations in the flue gas exceed a certain threshold, amine mist formation will occur. These aerosols grow in size as they travel through the CC absorber taking up of water and amine to sizes staying well below 1 μ m (80 % < 0.2 μ m) [15]. Despite the fact that most of the aerosols (expressed in number concentrations) are well below 1 μ m, most of the water (and thus amine) is found in the aerosol sizes between 0.5 and 2 μ m. Therefore, if one aims at designing efficient countermeasures, eliminating this size fraction is crucial.

Two types of countermeasures can be thought of: (i) removal of the sulfuric acid aerosols in the flue gas upstream the absorber or (ii) removal of the MEA containing aerosols leaving the absorber. Conventional countermeasures such as a water wash and a (standard) demister are highly effective in reducing vapor based solvent emissions, but are ineffective against aerosol based emissions with sizes smaller than $1-2 \mu m$ so are not a solution neither in front or behind the carbon capture absorber. In contrast, a Brownian Demister Unit (BDU) is a very effective counter-measure against aerosol based emissions [5]. However, the BDU comes at a high cost due to its high pressure drop and large area requirements. The BDU led to a pressure drop increase of about 50 *mbar* and is therefore not considered to be a valuable countermeasure for large scale carbon capture installations. Wet Electrostatic Precipitators (WESP) for the removal of sulfuric acid aerosols have been implemented in industrial plants before [16 and 17] and are therefore a possible alternative to the BDU. Moreover, the pressure drop associated to the operation of a WESP is typically lower than 1 *mbar* and as a result, energy requirements and operating costs tend to be low [18]. Inside a WESP, particles are charged and subsequently removed from the flue gas in an electrostatic field. The aerosol charging is realized by ions produced in a corona discharge.

Apart from installing a BDU or WESP in front or behind the carbon capture absorber to remove these aerosols, a possible countermeasure could also be related to the prevention of the formation of sulfuric acid aerosols (i.e. prevention of supersaturation conditions) in the flue gas treatment system of the industrial installation. In some industrial plants, a Gas-Gas Heater (GGH) is installed up-/downstream of the WFGD to cool down the flue gas before entering the WFGD and heat up the gas leaving the WFGD again for discharging the flue gas through a dry stack. A dry stack prevents corrosion issues inside the stack as a result of the condensation of acid gases but also results in the flue gas plume being invisible (as opposed to a wet flue gas stack). Since the temperature across a GGH is reduced from above the acid dew point of sulfuric acid to below, a GGH is known to condense a part of these acid gases. Therefore, the GGH consists of enamelled plates which avoid a quick corrosion. Along with the removal of part of

acid gases, a GGH reduces the temperature of the flue gas reaching the WFGD significantly which will influence the supersaturation profile inside the WFGD and thus affect the homogenous nucleation.

This study presents the results of two field tests that aimed at evaluating these two countermeasures to acid mist formation:

- (i) Evaluate the efficiency of a WESP to prevent amine mist formation inside an amine carbon capture plant through the removal of mist formation precursors (i.e. in this case H₂SO₄ aerosols) [19].
- (ii) Investigate to what extent a GGH affects the H₂SO₄ concentrations present in a coal fired power plant's flue gas and determine if a GGH can prevent amine mist formation issues in amine based carbon capture [20].

Apart from presenting these two countermeasures of which most of details have been published before, this study will discuss advantages and disadvantages of both countermeasures and formulate some recommendations to plant owners designing an amine based post combustion carbon capture installation.

2. WESP as countermeasure for mist formation?

2.1. Material and methods

The layout of the experiment is presented in detail in [19] and is therefore not presented in detail here. The experiments were performed at the pilot plant (Fig. 1) at the Institut für Technische Thermodynamik und Kältetechnik (ITTK) at Karlsruhe Institute of Technology (KIT) where a flue gas of ~175 Nm³/h is generated by burning natural gas [21]. A constant dosing of sulphur trioxide is accomplished via a microreactor where SO₂ is oxidized into SO₃ at 500°C. In this study, two different SO₂ flows (4 and 50 E⁻⁶ m³ min⁻¹) are sent into the micro-reactor (with catalyst). Previous analyses [1] indicate that these values correspond to respectively around 2.5 and 15 mg Nm⁻³ of H₂SO₄ inside the flue gas right behind the quencher, upon entering of the WESP. The aerosol formation takes place in a quench cooler (co-current packed column absorber). In this absorber, the hot flue gas is cooled down fast from ~200°C to the adiabatic saturation temperature of approximately 46°C by circulating water. During the quenching, high supersaturation conditions are generated in the gas phase leading to homogeneous nucleation of sulphuric acid [22]. The ITTK pilot plant has been designed to simulate a Flue Gas Desulphurization (FGD) unit and understand the formation of H₂SO₄ aerosols. As indicated in Fig. 1, at the outlet of the quench, a split stream is directed to a WESP and then into TNO's CO₂ mini-capture plant. A single stage tube type WESP was used for these experiments. TNO's mobile carbon capture mini-plant is a conventional absorption-desorption system with a capacity of 4 m³ h⁻¹ of flue gas; Monethanolamine (MEA) was used as solvent.

A T-connection at the WESP outlet enabled to measure online the aerosol number concentration and particle size distribution (PSD) by means of an Electrical Low Pressure Impactor (ELPI⁺) analyser (DEKATI). In this measurement device, the particles are charged by corona charging and subsequently separated in a low pressure cascade impactor with 14 electrically insulated collection stages. The measured current signals are proportional to the number concentration and size.

Simultaneously, a FTIR analyser (GASMET CX 4000) was used to analyse the gas phase at the absorber outlet. The gas leaving the absorber column was heated to 180°C using a trace heated transfer line. At this temperature the aerosol phase is vaporised implying that the concentration of MEA measured by FTIR is the sum of MEA present both in the vapour phase and in the aerosols [9, 10].



Fig. 1 Pilot plant layout at the Karlsruhe Institute of Technology. SO_3 is added to the flue gas behind a natural gas combustion transforming into H_2SO_4 aerosols inside a quench column. Part of this H_2SO_4 containing flue gas is send into a WESP and consequently into TNO's mini carbon capture plant using MEA as sorbent. ELPI⁺ measurements are carried out on the flue gas leaving the WESP and going into the MEA absorber; FTIR measurements on the flue gas leaving the MEA absorber.

2.2. Results and discussion

The results are presented separately for each of the two H_2SO_4 levels generated by the H_2SO_4 production plant. Important to note here is that apart from the H_2SO_4 concentrations being very different between both experiments, also the residual SO_2 concentration in the flue gas is very different, the efficiency of the catalyst converting SO_2 to SO_3 decreases at higher flow rates (from 80 % to below 40 %) [1]. This means that in case of high H_2SO_4 concentration, we measured around 20 mg Nm⁻³ of residual SO_2 reaching the water quencher and finally the WESP. This is in contrast to the low H_2SO_4 level where we measured no or very low levels (< 1 mg Nm⁻³) to remain in the flue gas by the time it reaches the WESP.

2.2.1 Low H₂SO₄ concentration level (2.5 mg Nm⁻³)

Fig. 2 shows a reduction of the total aerosol number concentration leaving the WESP with increasing voltage. The WESP is capable of removing sulphuric acid aerosols reaching an efficiency of almost 100 % at 20 kV over the entire size range (between 6 nm and 10 μ m). With the WESP off, a total number of 2.3E7 cm⁻³ H₂SO₄ aerosols leaves the WESP. This is reduced to a number of only 670 cm⁻³ H₂SO₄ aerosols at a voltage of 20 kV. This latter number is close to the detection limit of the ELPI⁺ which means that at 20 kV, we can state that no relevant amount of aerosols are left inside the flue gas. This corresponds to a WESP removal efficiency of nearly 100 %.

The MEA concentrations measured by the FTIR in the flue gas leaving the absorber as a function of the incoming aerosol number concentration measured by the ELPI⁺ are also presented in Fig. 2. The WESP clearly reduces the MEA emissions from the top of the absorber from close to 350 mg Nm^{-3} to values close to zero (only volatile MEA leaving the absorber) above 18 kV. This experiment indicates that the WESP is a suitable countermeasure for MEA mist formation and if operated at the correct voltage level, can fully prevent mist formation inside the absorber through the removal of the nuclei (i.e. H_2SO_4 aerosols) in the flue gas to be treated.



Fig. 2 Total numbers of aerosols (ELPI⁺ measurement) leaving the WESP and the corresponding MEA emissions from the top of the carbon capture absorber (FTIR measurement) as a function of applied WESP voltage (2.5 mg H_2SO_4 level)

2.2.2 High H₂SO₄ concentration level (15 mg Nm⁻³)

Fig. 3 shows that with an initial level of 2.2 E7 cm⁻³ aerosols (similar number concentration as presented above for the low H_2SO_4 concentration level) with WESP off, the minimal aerosol number is obtained at 18 kV corresponding to a removal efficiency of around 95 %. However, still a large number of aerosols is observed in the flue gas (> 1E6 cm⁻³) at 18 kV and these numbers even increase when going to higher voltages. This corresponds to the phenomenon presented in [23] for high levels of SO₂ in the flue gas. This study reveals that a WESP can actually generate H_2SO_4 aerosols in the presence of SO₂ [23]. It is hypothesized that the reactive species produced in the non-thermal plasma of the corona discharge of the WESP oxidize the SO₂ to SO₃ which forms sulfuric acid in the presence of water vapour. This causes supersaturation inside the WESP with subsequent homogeneous nucleation and thus aerosol formation. This is confirmed by the information presented in Fig. 3. Above 18 kV and up to 22 kV, the WESP continues to remove the larger aerosol fractions (above 300 nm) but actually starts producing very small aerosols (between 6 and 100 nm). It is this small fraction (very high numbers) that actually dominates the total number concentrations and therefore we observe an increase in total number concentration. This information is complementary to what was described in [23] since this study reveals that the aerosols created by the WESP are all very small and in the low nanometer range.

Very recently, these findings have been confirmed by the research group at the Research Center of Air Pollution Control Technology in China [24, 25]. The plasma induced effect inside the WESP in the presence of SO_2 is shown to be responsible for ultrafine sulfuric acid aerosol formation. These authors also show that at higher flue gas velocities, the plasma induced effect is weakened significantly.



Fig. 3 Total numbers of aerosols (ELPI⁺ measurement) leaving the WESP and the corresponding MEA emissions from the top of the carbon capture absorber (FTIR measurement) as a function of applied WESP voltage (15 mg H_2SO_4 level)

Fig. 3 confirms the large reduction in MEA emissions from the top of the absorber from around 800 to close to 100 mg Nm⁻³ at 18 kV. A further small reduction is actually observed up to 22 kV despite the fact that the total aerosol number concentration going into the absorber is actually increasing between 18 and 22 kV. It is hypothesized that this could be due to the too small sizes of the newly created aerosols preventing their activation as mist formation precursors. Since the larger aerosols are still removed up to 22 kV, MEA emissions continue to decrease a little. In order for particles to grow due to heterogeneous nucleation, supersaturation is necessary; however, according to the Kelvin equation, smaller nuclei require larger supersaturations [26]. Because of this larger nuclei are first activated and start growing taking up water and MEA as they move through the absorber [15]. This could imply that there is a smaller chance for the smaller nuclei to be activated and take up water and MEA. To check this hypothesis, not only the measured total number concentration is plotted but also the number concentrations for aerosols larger than 200 nm at the different WESP voltages is added. This shows an increase in aerosol number concentration (> 200nm) only when going from 22 to 24 kV which agrees with the observation that only above 22 kV, also the MEA emissions rise. So the observation that the relation between number concentration for aerosols larger than 200 nm and the mission profile at the different voltage levels are similar, suggests that indeed a minimum size is needed for the aerosols to grow sufficiently and take up MEA. This implies that the hypothesis that small nuclei do not serve as mist formation precursors should not be generalized to other pilot/demo plants since it is very much a function of the supersaturation profile. This profile strongly depends on the operating conditions of the MEA scrubber such as temperatures and residence times. At 24 kV, the MEA emissions seem to rise again most likely as a result of the reduced aerosol removal efficiency (or because of the newly created aerosols) at these high voltage levels in the presence of SO₂.

3. Gas-Gas Heater as countermeasure for mist formation?

3.1. Material and methods

The measurements presented in this study were carried out at ENGIE's 600 MW (electrical output) coal fired power plant in Nijmegen, The Netherlands. The flue gas stream is split-up into two streams (half-half) in front of the Selective Catalyst Reduction (SCR) which come together again in the stack (see Fig. 4). Unique is that only one of the trains enters the WFGD right behind the ESP (Electro Static Precipitator) at a temperature of 122 °C and leaves the WFGD at around 52 °C; whilst the flue gas in the other train goes into a GGH behind the ESP where the flue gas is cooled from 122 °C to around 80 °C and then into the WFGD in which the temperature is further reduced to 52 °C. The flue gas leaving the GGH and going into the stack has a temperature of around 89 °C and is mixed with the flue gas from the other flue gas train inside the stack reheating the 'cold' flue gas to a temperature above the water dew point. The ports (A to D) from which the flue gas stream was sampled are indicated on Fig. 4.



Fig. 4 Unique layout of the flue gas treatment system at ENGIE's Nijmegen power plant made-up of two parallel flue gas trains (SCR, Selective Catalytic Reduction; APH, Air Pre-Heater; ESP, ElectroStatic Precipitator; WFGD, Wet Flue Gas Desulphurization; GGH: Gas-Gas Heater)

At each of the locations, H_2SO_4 concentrations in the flue gas were measured as well as the aerosol sizes and numbers using the ELPI⁺ device. The flue gas was also sent into a mini-absorber column (see Fig. 5) which is a scaled down mobile absorption column which can be used for CO₂ capture. This column is equipped with Sulzer laboratory packing with a packed height of 0.08 m and a diameter of 0.09 m. The column is insulated and is not equipped with a waterwash. Typical solvent flow rates in the mini-column are around 5-101 h⁻¹, with a flue gas flow of 450-15001 h⁻¹. MEA (30 weight %) was used as the CO₂ capture solvent (details in [20]).



Fig. 5 Picture of the mini-absorber used at the different sampling locations (left: presence of mist formation, right: no mist formation)

3.2. Results

Fig. 6 presents the H_2SO_4 concentrations measured simultaneously at three locations: A, B and D over the two sampling days. The sampling length of each measurement is indicated and varied between minimum one hour and maximum two hours. At location D, ten measurements were carried out spread out evenly over the two days measurement resulting in an average value of 5.2 mg Nm⁻³ H₂SO₄. These values are found to be stable throughout both measuring days and only vary between 4.6 and 6.3 mg Nm⁻³.



Fig. 6 H₂SO₄ values as measured during the two day sampling campaign at the four different measurement locations (left: first day measurement, right: second day measurement)

Fig. 7 presents the corresponding measured Particle Size Distribution (PSD) using the ELPI⁺ at the three locations. A rather low total PM/aerosol number concentration of around 6 E5 cm⁻³ is measured at location D which is rather typical for fine particulate matter concentrations behind an ESP and thus reveals the absence of H_2SO_4 in the aerosol form. Previous work has revealed that concentrations of around 1 E8 cm⁻³ are to be expected when H_2SO_4 is present in the aerosol form [15]. Despite the absence of H_2SO_4 aerosols in the flue gas, sending this flue gas into the mini-absorber revealed a clear mist formation at the top of the column; this is due to aerosol formation taking place inside the mini-absorber as in more detailed explained below.



Fig. 7 Comparison of the measured particle size distributions (PSD) (left) and cumulative particle size distributions (right) at all three locations (A, C and D) with the measured PSD of H_2SO_4 aerosols at a concentration of around 3 mg Nm⁻³[1]

At location C, sulphuric acid concentrations vary between 1.2 and 2 mg Nm⁻³ with an average value of around 1.6 mg Nm⁻³ (4 measurements in total). Fig. 7 presents the corresponding measured particle size distribution and very similar results are obtained as at location D: a rather low total PM/aerosol number concentration of around 6 E5 cm⁻³ is measured which implies the absence of H_2SO_4 in the aerosol form. Sending this flue gas (location C) into the mini-absorber revealed no mist formation at the top of the column at all.

Fig. 6 shows that the H_2SO_4 concentrations at location B were also relatively stable with values varying between 1.5 and 2 mg Nm⁻³ with an average value of around 1.7 mg Nm⁻³ (3 measurements in total). Due to time constraints, no ELPI⁺ measurements nor mini-absorber testing could be carried out at location B. However, in a previous test campaign consisting of only mini-absorber testing, no visual mist formation was observed at this location.

At location A, H_2SO_4 concentration measurements reveal similar values (two measurements available) than at location D i.e. around 5.8 mg Nm⁻³. A very different PSD and total number concentrations are obtained at location A as compared to the other two locations where ELPI⁺ measurements were carried out (C and D). A very high total number concentration of around 6 E7 cm⁻³ is measured at this location, which is 100 times higher than at location D and C. Moreover, the high number concentrations respond to very small aerosol/PM sizes i.e. around 80 % < 0.02 µm. Sending this flue gas into the mini-absorber revealed an immediate mist formation effect at the top of the column.

3.3. Interpretation

The flue gas leaving the ESP at the Nijmegen coal fired power plant contains between 5 and 7 mg Nm⁻³ of H₂SO₄ which is in a gaseous form because the flue gas temperature at this location is still above the acid dew point (flue gas temperature is 122 °C). The acid dew point of flue gas is a function of the H₂SO₄ concentration inside the flue gas and of the water content. Assuming a H₂SO₄ concentration of around 6 mg Nm⁻³ and a water content of around 8 vol%, the acid dew point is estimated to be around 110 °C [27]. The PSD measurements made using the ELPI⁺ confirm low particle numbers of around 6.5 E5 cm⁻³, which most likely relate to the small fly ash particles present in the flue gas.

In the absence of a GGH, the flue gas leaving the ESP (containing gaseous H_2SO_4) enters the WFGD. The WFGD does not remove any of the H_2SO_4 at the Nijmegen power plant since similar concentrations are found in front and behind the WFGD. However, high supersaturation conditions prevail inside the WFGD and homogenous nucleation takes place converting the incoming gaseous H_2SO_4 into H_2SO_4 aerosols. This is confirmed by the measured PSD at location A: (i) total particle number of 6 E7 cm⁻³ (which is factor 100 higher than in front of the WFGD) and (ii) the particle size distribution shifts completely to the smaller particle sizes which is typical for H_2SO_4 aerosols (e.g. [1]). In Fig. 7, the measured PSD at all locations are compared with the PSD of pure H_2SO_4 aerosols with a concentration of around 3 mg Nm⁻³ of H_2SO_4 . It is confirmed that the measured PSD behind the WFGD (in absence of a GGH) corresponds very well with the PSD of H_2SO_4 aerosols.

If, in contrast, a GGH is in place, the flue gas is cooled (rather gently since a GGH is a rotating device) from around 122 °C down to 80 °C inside this GGH. This means that the flue gas goes through its acid dew point (at around 110 °C) and H₂SO₄ must be condensed out of the flue gas leading to possible corrosion issues inside the GGH. However, the H₂SO₄ measurements behind the GGH (locations B and C), reveal that still between 1 and 2 mg Nm⁻³ of H₂SO₄ is left inside the flue gas. Therefore, only about 70 % (from 6 to 1-2 mg Nm⁻³) is condensed out of the flue gas onto the surface of the GGH. The remaining 1 and 2 mg Nm⁻³ of H₂SO₄ inside the flue gas can also not be present in the gaseous form since the conditions are well below the dew point. The PSD data shows that this remaining H₂SO₄ is also not found in the form of aerosols formed by homogenous nucleation since because of the low particle/aerosol numbers (similar to right behind the ESP) and a PSD similar to the fly ash PSD obtained right behind the ESP. This can be interpreted that inside the GGH, heterogeneous nucleation takes place and the gaseous H₂SO₄ condenses not only on the surface of the GGH but also partly condenses on the fly ash particles present in the flue gas. This condensation on the fly ash particles is not reflected in the measured PSD as a growth since the amount of H₂SO₄ is too limited to be seen as a significant growth of the fly ash particles. The reason why homogenous nucleation does not take place inside the GGH is the much milder supersaturation conditions since the flue gas is only cooled down

from 122 °C down to 80 °C. Moreover, the cooling process is much slower inside a GGH as compared to a WFGD in which it is immediate due to the spraying of water. The GGH investigated in this study is a turning device and it therefore takes several seconds to go from the 122 °C down to 80 °C. The subsequent further cooling in the WFGD from 80 °C to 50 °C does not alter the H_2SO_4 concentration nor PSD any more since it is already condensed onto the fly ash particles.

Leading the flue gas behind a WFGD and in the absence of a GGH into an amine based carbon capture installation may lead to mist formation issues due to thigh amount of nuclei (i.e. H_2SO_4 aerosols) present in the flue gas. Based on these low particle numbers measured directly behind the ESP, no amine mist formation would be expected when sending this flue gas into a CC absorber (see [14]). However, we do observe mist formation inside the mini-absorber column and this is very likely due to the fact that homogenous nucleation also takes place inside the mini-absorber. The combination of the high inlet temperature of 122 °C and the sudden drop to ambient temperature conditions inside the mini-column creates a high supersaturation. This supersaturation then results in homogeneous nucleation and, thereby, converts the incoming gaseous H_2SO_4 into H_2SO_4 aerosols inside the mini-column which then lead to mist formation.

Since a GGH prevents the formation of H_2SO_4 aerosols both inside the GGH and inside the WFGD, no mist formation in observed the mini-column. It is important to stress that this is not due to the lower H_2SO_4 (of only 1-2 mg Nm⁻³) concentration as compared to the absence of a GGH since earlier studies [2,3 and 4] revealed that as low as 1-2 mg Nm⁻³ H_2SO_4 can still lead to amine mist formation issues. The reason is that the H_2SO_4 behind the GGH is not in an aerosol form but in a condensed form onto the fly ash particles and thus only a low amount of nuclei are present inside the flue gas. These low particle number concentrations are not sufficient to cause mist formation issues in amine based CC.

4. Conclusions

This study shows that a WESP is very efficient for the removal of nuclei from the flue gas (100 % efficient) and thus can prevent aerosol formation inside an amine based absorber. A WESP is shown to be very efficient for the removal of nuclei from the flue gas (100 % efficient) and thus can prevent aerosol formation inside an amine based absorber. This is however only valid in the absence of SO₂ in the flue gas entering the WESP. A decreasing WESP efficiency is noted in the presence of SO₂ with increasing voltages as a result of newly formed aerosols inside the WESP. Depending on the voltage level and the size of the newly formed aerosols, this may decrease the positive effect a WESP has on the MEA emissions from the absorber. This highly reduced efficiency of the WESP implies that mist formation may in some cases (depending on the operating conditions and thus supersaturation profile) no longer be prevented inside the absorber and high MEA emissions may occur. This information is very important for future pilot and demo amine carbon capture installations thinking of implementing a WESP as countermeasure to aerosol formation issues. It implies that no or very low levels of SO₂ should be present in the flue gas entering the WESP. Since most of the amine carbon capture installations have a pre-scrubber (usually using NaOH to remove residual SO₂ in the flue gas leaving the power plant's FGD) in front of their amine absorber, the WESP must be installed behind this pre-scrubber and not in front of it.

In case of industrial processes whereby the flue gases contain sulfuric acid and the flue gas enters a wet scrubbing process where temperatures drop drastically and instantaneously from above to below the sulfuric acid dew point, supersaturation conditions may exist. This leads to homogenous nucleation converting the gaseous H_2SO_4 into aerosol H_2SO_4 . Then high aerosol number concentrations are likely to be present behind the scrubber and the observed PSD is similar to reported H_2SO_4 PSD in literature (i.e. 80 % smaller than 0.02 µm). Therefore an amine based carbon capture installation treating this flue gas will suffer from amine mist formation emissions. If however a Gas-Gas heater is installed upstream of the wet scrubbing, the GGH may prevent homogenous nucleation taking place and thus preventing the conversion of H_2SO_4 into aerosol H_2SO_4 . It is important to understand that it is not the reduction in H_2SO_4 concentration by 70 % inside the GGH that prevents mist formation but the absence of homogenous nucleation and thus the presence of H_2SO_4 in its aerosol form (i.e. large aerosol number concentrations).

5. Recommendations

Which option to choose amongst the two countermeasures presented in this study will depend on whether a new built installation is being considered or whether a carbon capture is planned as a retrofit into an existing installation:

- If both the industrial plant and the CC installations are new build; a flue gas cooler (e.g. Low Temperature Heat Exchanger (LTHE)) similar to the GGH presented here in the flue gas treatment layout of the power plant is recommended in order to prevent amine mist formation issues in the CC plant. However, in contrast to what is presented in this study, we propose to use the hot air stream as energy in the plant (e.g. for combustion air preheating) and not to heat up the flue gas before entering the stack (to prevent a wet stack). Apart from the gain in energy, also water savings should be expected in the wet scrubbing process downstream since less cooling power is required. Only drawback of the system is the small pressure loss over the installation leading to a higher booster fan energy consumption. However, an overall gain in energy is to be expected which could overcome the costs involved in the installation of the infrastructure.
- If a carbon capture retrofit to an existing system is considered, the installation of a LTHE system in front of the scrubbing process may not always prove to be possible. In that case, we can conclude that a WESP is a suitable option to prevent amine mist formation inside the absorber since it proves to be very efficient for the removal of the precursors inside the flue gas. Although a significant capital cost may be involved, energy requirements (low pressure drop), maintenance and thus operational costs are expected to be very low. It makes it therefore a plausible option at the industrial scale in contrast to a BDU installation where pressure drops up to 50 mbar are to be expected (e.g. [5]). However, the WESP must be installed at the right location i.e. in SO₂ free or poor flue gas.

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References

- [1] Brachert, L., J. Mertens, P. Khakharia and K. Schaber, 2014. The Challenge of Measuring Sulfuric Acid Aerosols: Number Concentration and Size Evaluation using a Condensation Particle Counter (CPC) and an Electrical Low Pressure Impactor (ELPI⁺). Journal of Aerosol Science, 61, 21-27
- [2] Kamijo, T., 2011. Amine Emission Control Technology of KM CDR ProcessTM, Presentation at the EPRI meeting: 'Amines for post combustion Capture', 16th August 2011
- [3] Kamijo, T., Y. Kajiya, T. Endo, H. Nagayasu, H. Tanaka, T. Hirata, T. Yonekawa, and T. Tsuji-uchi, 2013. SO₃ impact on amine emission and emission reduction technology. *Energy Proceedia*, 37, 1793–1796
- [4] Khakharia, P., L. Brachert, J. Mertens, A. Huizinga, B. Schallert, K. Schaber, T. Vlugt, and E. Goetheer, 2013. Investigation of aerosol based emission of MEA due to sulphuric acid aerosol and soot in a Post Combustion CO₂ Capture process. *International Journal of Greenhouse Gas Control*, 19, 138-144
- [5] Khakharia, P., H.M Kvamsdal, E.F. da Silva, T.J.H. Vlugt and E. Goetheer, 2014. Field study of a Brownian Demister Unit to reduce aerosol based emission from a Post Combustion CO₂ Capture plant. *International Journal of Greenhouse Gas Control*, 28, 57–64
- [6] Kolderup, H., K. Hjarbo, A. Huizinga, I. Tuinman, K. Zahlsen, K. Vernstad, A. Hyldbakk, T. Holten, H. Kvamsdal, P. van Os, E. da Silva, E. Goetheer, and P. Khakharia, 2012. Emission Quantification and Reduction (No. WP 1 and 3 in the project: CCM TQP Amine 6 for Gassnova SF, Norway)
- [7] Knudsen, J. and O. Bade. 2012: Emission Reduction Technologies; CLIMIT Workshop 5-6 December 2011, Oslo, Norway. Available at www.akercleancarbon.com
- [8] Carter, 2012. National Carbon Capture Center: Post-Combustion., 2012 NETL CO₂ Capture Technology Meeting, July 10, 2012. http://www.netl.doe.gov/publications/proceedings/12/co2capture/presentations/2-Tuesday/T%20Carter-NCCC-Post-combustion.pdf
- [9] Mertens, J., M. L. Thielens, J. Knudsen and J. Andersen, 2012. On-line monitoring and controlling emissions in amine post combustion carbon capture: a field test. International Journal of Greenhouse Gas Control, 6, 2-11

- [10] Mertens, J., H. Lepaumier, D. Desagher and M.L. Thielens, 2013. Understanding Ethanolamine (MEA) and ammonia emissions from amine based post combustion carbon capture: lessons learned from field tests. *International Journal of Greenhouse Gas Control*, 13, 72-77
- [11] Moser, P., S. Schmidt, K. Stahl, G. Vorberg, G. A. Lozano, T. Stoffregen, and F. Rösler, 2014. Demonstrating Emission Reduction Results from the Post-combustion Capture Pilot Plant at Niederaussem. *Energy Procedia*, 63, 902-910
- [12] Baburao, B., S. Bedell, P. Restrepo, D. Schmidt, C. Schubert, B. DeBolt, I. Haji and F. Chopin, 2014. Advanced Amine Process Technology Operations and Results from Demonstration Facility at EDF Le Havre. *Energy Procedia*, 63, 6173 – 6187
- [13] Khakharia, P., J. Mertens, M.R.M. Abu-Zahra, T.J.H. Vlugt, and E.L.V. Goetheer, 2016. Overview of aerosols in postcombustion CO₂ capture. Published in Absorption-Based Post-Combustion Capture of Carbon Dioxide ISBN 978-0-08-100514-9
- [14] Mertens; J., L. Brachert, D. Desagher, B. Schallert, P. Khakharia, E. Goetheer, 2014. Predicting amine mist formation based on aerosol number concentration and size measurements in flue gas. *Energy Proceedia*, 63, 893-901
- [15] Mertens, J., L. Brachert, D. Desagher, M.L. Thielens, P. Khakharia, E. Goetheer and K. Schaber, 2014. ELPI⁺ measurements of aerosol growth in an amine absorption column. *International Journal of Greenhouse Gas Control*, 23, 44-50
- [16] Brown, C. and P. Hohne, 2001. Eliminating a sulfuric acid mist plume from a wet caustic scrubber on a petroleum coke calciner. *Environmental Progress*, 20, 182–186
- [17] Srivastava R.K., C. Miller, C. Erickson and R. Jambhekar, 2004. Emissions of sulfur trioxide from coal fired power plants. Journal of the Air & Waste Management Association, 54, 750–62
- [18] EPA-452/F-03-029. Air Pollution Control Technology. Fact Sheet. EPA-CICA Fact Sheet. Wet Electrostatic Precipitator (ESP). Wire-Pipe Type. (http://www.epa.gov/ttncatc1/dir1/fwespwpi.pdf)
- [19] Mertens, J., C. Anderlohr, P. Rogiers, L. Brachert, P. Khakharia, E. Goetheer and K. Schaber, 2014. A Wet Electrostatic Precipitator (WESP) as countermeasure to mist formation in amine based carbon capture. *International Journal of Greenhouse Gas Control*, 31, 175-181
- [20] Mertens, J. R. Bruns, B. Schallert, N. Faniel, P. Khakharia, W. Albrecht, E. Goetheer, J. Blondeau and K. Schaber, 2015. Effect of a Gas-Gas-Heater on H₂SO₄ aerosol formation: implications for mist formation in amine based carbon capture. *International Journal of Greenhouse Gas Control*, 39, 470-477
- [21] Brachert, L., T. Kochenburger and K. Schaber, 2013. Facing the Sulfuric Acid Aerosol Problem in Flue Gas Cleaning: Pilot Plant Experiments and Simulation. Aerosol Science and Technology, 10, 1083-1091
- [22] Wix, A., L. Brachert, S. Sinanis and K. Schaber, K., 2010. A simulation tool for aerosol formation during sulphuric acid absorption in a gas cleaning process. Journal of Aerosol Science, 41, 1066-1079
- [23] Anderlohr, C., L. Brachert, J. Mertens and K. Schaber, 2014. Collection and Generation of Sulfuric Acid Aerosols in a Wet Electrostatic Precipitator. Aerosol Science & Technology, 49, 144-151
- [24] Huang, J., H. Wang, Y. Shi, F. Zhang, X. Dang, H. Zhang, Y. Shu, S. Deng and Y. Liu, 2016a. Investigation of a pilot-scale wet electrostatic precipitator for the control of sulfuric acid mist from a simulated WFGD system. *Journal of Aerosol Science*, 100, 38-52
- [25] Huang, J., H. Wang, Y. Shi, F. Zhang, X. Dang, H. Zhang, Y. Shu, S. Deng and Y. Liu, 2016b. Performance of a pilot-scale wet electrostatic precipitator for the control of sulfuric acid mist. *Environ Sci Pollut Res*, doi:10.1007/s11356-016-7151-x
- [26] Fletcher, N.H., 1958. Size effects in heterogeneous nucleation. Journal of Chemical Physics, 29, 572-576
- [27] ZareNezhad, B., 2009. New correlation predicts flue gas sulfuric acid dew points. Oil & Gas Journal, 107, 60-63