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High amine emissions formation and aerosol particle size distribution in post-combustion CO_2 capture process

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ABSTRACT

Amine emissions in the CO_2 absorption system may lead to solvent loss and cause a series of environmental problems, especially in high nuclear scenario. In the 3.6 Nm^3/h test bench, when the simulated flue gas passes through the absorber, the particle concentration dropped sharply from 3.43×10^7 cm^{-3} to 4.41×10^6 cm^{-3}. The total MEA emissions reached 1519 mg/Nm^3 at the outlet of the absorber, including 476 mg/Nm^3 vapor-type amine. By overcoming the difficulty of measuring high-quality liquid particles, the results of particle size distribution (PSD) provided a revealing insight into aerosol growth. The changing trend of amine emissions was completely consistent with total particle emissions. High absorption reaction intensity can be achieved with lower lean solvent CO_2 loading, appropriate CO_2 concentration (12%), and higher solvent temperature (only for small absorbers). Then, more droplets grew up to near-micron level (> 733 nm), causing total particle emissions increased dramatically to 3500–6000 mg/Nm^3. Considering the CO_2 absorption system is equipped with a water wash column and demister, the absorber outlet particle with a size less than 1 μm (PM1) will be the main threat. The PM1 emissions and control strategies were discussed based on the PSD data. It is recommended to promote aerosol growth after the absorber rather than restrain it in the absorber.

1. Introduction

Chemical absorption is currently the most promising post-combustion CO_2 capture (PCC) technology due to its high capture efficiency, stable operation conditions, mature and flexible configurations (Rochelle, 2009). The principle is to use the aqueous solution of amine to react and separate carbon dioxide from flue gas. In recent years, many breakthroughs have been made in this field. Water-lean solvents such as biphasic solvents and ionic liquid greatly reduce the regeneration energy (Liu et al., 2019; Wang et al., 2017). Solvent additives such as nanoparticles and metal ions can enhance CO_2 absorption and desorption (Wang et al., 2016). However, when the flue gas is in direct contact with solvents in the absorber, it carries part of the solvents and its degradation products and discharges them into the air, causing solvent loss and a series of environmental problems.

There are mainly two types of solvent emissions, vapor and aerosol. Different from vapor-type emissions, aerosols in flue gas can't be simply removed by a water wash column (De Cazenove et al., 2016; Majeed and Svendsen, 2018a). Lots of works have been reported that aerosol-form emissions can be as high as g/Nm^3 level(da Silva et al., 2013; Fujita et al., 2017; Harsha et al., 2019a; Knudsen et al., 2013; Mertens et al., 2014c, 2014b; Spietz et al., 2018). While traditional mist control equipment has low efficiency in eliminating particles with diameters less than 3 μm. According to relevant research, the formation of aerosol-form amine emissions requires a large number of condensation nuclei that exist in the flue gas before entering the absorber(Fang et al., 2020). For a coal-fired power plant, the particle concentration in flue gas without any treatment can reach 10^7–10^9 per cm^3. Even the ultra-low emission technologies in China can control the particle mass concentration below 5 mg/Nm^3, its number concentration was in the range of 10^5–10^7 per cm^3 (Harsha et al., 2019b; Zhang, 2019). Therefore, it is an important subject to study aerosol emissions during the CO_2 capture process, especially in the case of the high nuclei scenario.

Aerosol formation and growth have been widely studied in atmospheric science and recently in wet-scrubbing processes. While in a CO_2 absorption system, the heterogeneous nucleation process of aerosols in

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the absorber is not a simple physical process. The reaction of CO_2 and solvent also affected the mass transfer on the surface of aerosol droplets (Khakharia et al., 2016). Khakharia proposed that the amine in the aerosol droplets reacts rapidly with CO_2 to form a nonvolatile carbamate or bicarbonate (Khakharia et al., 2015). It leads to the amine volatility reduction of droplets, and gaseous amine will condense continuously on the droplets. As the salt accumulated, water transfers from the gas to the droplets to achieve balance. This mechanism explains why the aerosols grow so fast in the absorber(Fulk, 2016).

To have a better understanding of how the aerosol growth is affected by column parameters and the aerosol growth mechanism, the particle size distribution data of aerosols is needed. Besides, many mathematical models need to be further verified and improved by experimental data (Majeed et al., 2017a, 2017b, 2017c; Majeed and Svendsen, 2018b). Khakharia studied the effect of lean solvent temperature, CO_2 concentration, and solvent reactivity. The total particle concentration and amine emissions were measured (Khakharia et al., 2015). Fujita used a solvent bubbling device to study the effect of aerosol-generating temperature and the existence of CO_2. Though the simulated gas have low nuclei concentration, the particle size change was observed (Fujita et al., 2014). Most aerosol emissions studies focus on the total amine emissions amount, the particle size distribution of aerosol after the absorber was seldom reported. When the particle mass concentration is high, it is difficult to measure the particle size distribution without a diluter. While it is still a challenge to accurately measure particles that contains a high proportion of liquid using a diluter.

In this study, CO_2 concentration, lean solvent CO_2 loading, and lean solvent temperature were investigated. In order to understand the effect of parameters on aerosol growth, both the particle number distribution and mass distribution at the outlet of the absorber were measured and modified by dilution correction factors. The particle size shifted by parameters change was observed in high nuclei scenario. Meanwhile, the amine emissions were measured by batch analytical methods. The relation between reaction intensity and aerosol growth was discussed. In addition, continuous monitoring experiments show the characteristic of particle number concentration and the Sauter mean diameter. These data also reveal that aerosol-form amine emissions are unstable and change with the absorber condition rapidly.

2. Experiments and methods

2.1. CO_2 chemical absorption emissions test platform

The CO_2 chemical absorption emissions test platform was a bench-scale system designed to test amine emissions in different conditions. This system contained an entire absorber-stripper cyclic process and two water wash columns. The absorber was packed with Φ6 × 6 Dixon ring random packing, which had a high specific surface area of 950 m^2/m^3. The column was 1.96 m in height and the inside diameter was 60 mm. There were five sections in the absorber column and each section was packed with 0.24 m packing. The retention time of flue gas in the absorber was about 5 s. It should be noted that there was no demister in the absorber and water wash column. All the columns except the stripper were made of plexiglass. Therefore, the contact of scrubbing liquid and simulated flue gas could be directly observed. The schematic drawing of the test platform is shown in Fig. 1. More details about bench-scale system can be found in previous work (Yi et al., 2021)

2.2. Operation parameter

The mixed gas from the air compressor and the CO_2 cylinder first passed through the pretreatment column to reach the prescribed temperature and saturation. Then, the high-concentration aerosols, generated by the Laskin nozzle aerosol generator, were added into the mixed gas as the simulated flue gas. The flue gas CO_2 concentration was controlled by the gas mass flowmeter. The lean solvent flow rate and water wash flow rate were adjusted by peristaltic pumps. An infrared gas analyzer (GHX-3010E1, Beijing Huayun Company, China) is used to measure the CO_2 concentration of the absorber gas outlet. Key operation parameters were shown in Table 1.

2.3. Measurement method

The amine concentration at the gas outlet of the absorber and that of the water wash column was measured by batch analytical methods. Flue gas from the sample point was heated up to 150 °C and then passed through two bubbling absorbers, which were filled with 0.25wt% H_2SO_4 solution in the ice-cooling water bath. Most of the amine was condensed

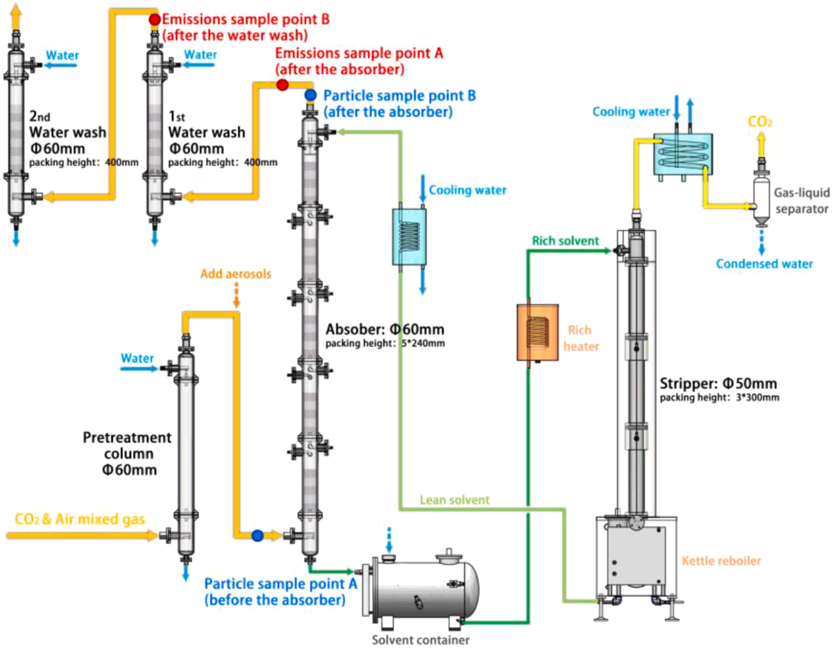


Fig. 1. Schematic drawing of the 3.6 Nm^3/h CO_2 chemical absorption emissions test platform.

Table 1
The operation parameters of the CO_2 chemical absorption system under basic condition.

Parameter		Unit	Value	Remains unchanged in other parameter studies
Solvent		–	5 mol/L MEA	Yes
Lean solvent	flow rate	L/min	0.6	Yes
	inlet temperature	°C	40	Yes
	CO_2 loading	mol/mol	0.24–0.27	Yes
Flue gas	flow rate	L/min	60	Yes
	inlet temperature	°C	40	Yes
	CO_2 concentration	%	12 (dry basis)	Yes
	nuclei density	cm^{-3}	$3.0–3.8 \times 10^7$	Yes
Water wash	flow rate	L/min	0.6	Yes
	temperature	°C	38–40	Yes
Absorber temperature		°C	40–60	No
Stripper temperature		°C	100–103	No
CO_2 capture rate		%	85–90	No

and reacted with sulphuric acid solutions to form salts. After the measurement, the solution in bubbling absorbers was weighted and analyzed by Dionex Aquinon IC with Dionex IonPac CS17 Analytical Column (4 × 250 mm). The amine emissions measurement error under repeated tests was less than 5%.

Electrical Low Pressure Impactor Plus (ELPI+, Dekati, Finland) was used to measure the particle size distribution (PSD) and concentration of the flue gas before and after the absorber. Due to the particle mass concentration being too high at the gas outlet of the absorber, the direct measurement will cause rapid accumulation of particles in the impactor, and the accuracy of the ELPI+ is significantly reduced during measurement. Therefore, the diluter DI-1000 with a nominal dilution factor of 1:8 was used for particle measurement after the absorber.

3. Results and discussions

3.1. Particle size distribution of the simulated flue gas

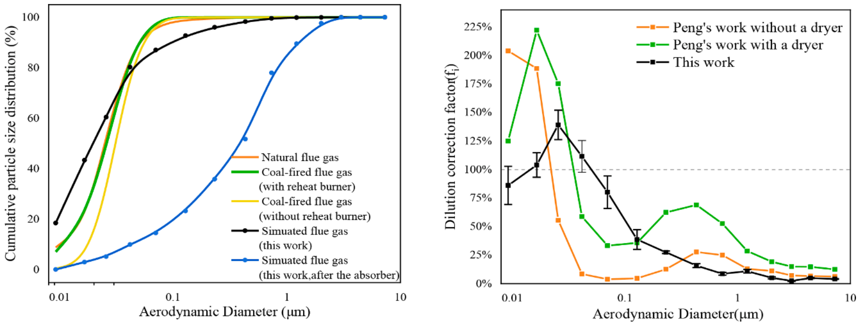
The mixed simulated flue gas was measured by an ELPI+ without a diluter. The average nuclei concentration before the absorber was as high as 3.43×10^7 cm^{-3}. The particle size distribution of flue gas was bimodal at 16 nm and 41 nm, and the particle number concentration decreased as the size increased. Li measured the particle size distribution after flue gas desulfurization(FGD) in an operating coal-fired power plant. Under the normal steady-state operating condition, the particle number distribution in the flue gas is concentrated at 40 nm (Li et al., 2019). Some previous works also show the most particles after electrostatic precipitators (ESP) were in the range of 10 to 100 nm (Li et al., 2009; Ylätalo and Hautanen, 1998). Mertens gave some particle size distribution before the absorber at different pilot plants. The particle concentration distribution also decreased as the size increased (Mertens et al., 2014b). Therefore, the simulated flue gas in this work had a similar particle size distribution to real flue gas. Fig. 2(a) shows more comparison with actual flue gas.

When the flue gas reacted with solvent and passed through the absorber, the particle mass significantly increased, and the particle size distribution has to be measured with the 1:8 diluter DI-1000. The ELPI+ diluter needs purified and pressurized(0.2Mpa) air as the dilute gas. When the pressurized gas drops to atmospheric pressure, it can't be wet saturated. However, most of particles measured in this work at the outlet of the absorber were droplets with a high concentration of water, it shrank when mixed with unsaturated gas. This phenomenon was also observed by Peng et al. (2019) with the same measure equipment, he found that humidity could influence particulate matter concentration measurements. Besides, the loss of droplets in the diluter is more complex than that of solid particles. Therefore, dilution correction factors(fi) of ELPI+ were induced to modify droplet shrink in diluter. The measurement results of the same gas with and without a diluter were used to calculate dilution correction factors. Considering dilution correction factors are also affected by particle size distribution, the gas under the basic condition was chosen to calculate correction factors in this work. This method was robust but effective. Fig. 2(b) gives results about dilution correction factors. The average error of dilution corrections of each stage was within 10%. Compared with Peng's results, it can be seen that the dilution correction factors were at a reasonable range.

The modified result shows the particle concentration after the absorber dropped sharply to 4.41×10^6 cm^{-3} in the basic condition. The particle loss was because of coagulation, solvent scrubbing, and adherence on the surface of packing. And the particle size distribution of flue gas was unimodal at 733 nm. It should be noted that particles with a diameter larger than 591 nm increased from 5.06×10^5 cm^{-3} to 2.03×10^6 cm^{-3}, which proved the particle coagulation and aerosol growth did happen in the absorber. In the pilot facility, Fujita also observed the aerosol particle peak changed from 60 to 100 nm to 1.5–3.5 μm when flue gas passed through an absorber and two water wash columns(Fujita et al., 2017).

3.2. Amine emissions and aerosol particle emissions

According to the parameters study of MEA/AMP and the preliminary experiments of MEA (Yi et al., 2021). The CO_2 concentration, lean solvent CO_2 loading, and lean solvent temperature were chosen as the high impact parameters that affect amine emissions and aerosol particle



(a) (b)

Fig. 2. (a) The simulated flue gas compared with actual flue gas(Brachert et al., 2014; Li et al., 2019) and (b) the dilution correction factors of the diluter DI-1000 (Peng et al., 2019).

growth. The former two parameters dominate the absorption reaction and amine volatility in the absorber. And the lean solvent temperature has a great impact on the column temperature profile, which also influences the absorption reaction rate, reaction equilibrium, and vapor equilibrium partial pressure of amine.

Amine emissions from the chemical absorption process consist of vapor form and aerosol form. The total amine emissions after the absorber and after the water wash column were measured by batch analytical method. The vapor emissions follow vapor liquid equilibrium and can be evaluated by the Aspen model. The aerosol-form amine emissions are the amine solution in the form of droplet, which can be calculated by total amine emissions and vapor emissions. To discuss relations between amine emissions and particle emissions, they were put in the same figure.

As shown in Fig. 3(a), the maximum MEA emissions were 1519 mg/Nm^3 after the absorber and 249 mg/Nm^3 after the water wash column at 12% CO_2. The maximum particle mass emissions of 3942 mg/Nm^3 were also observed in the same condition. At this condition, CO_2 in flue gas and amine in solvent had a perfect ratio to fully react. It can be seen that the reaction between CO_2 and solvent significantly promotes aerosol-form amine emissions. High reaction heat also resulted in vapor emission up to 476 mg/Nm^3. It's not a coincidence that the amine emissions and particle mass have the same trend vary with CO_2 concentration. It indicates aerosol emissions that are affected by particle mass in flue gas dominate the total amine emissions in the high nuclei scenario. Khakharia's study of AMP/PZ (Khakharia et al., 2015) and Fulk's study of PZ (Fulk, 2016) also found the amine emissions first rise and then decrease slightly with the flue gas CO_2 concentration increased. The maximum amine emissions were observed at 6% and 10%, respectively. It should be noted that both experiments were carried out with high concentration nuclei in the flue gas. The water wash column amine control efficiency in this work is approximately 82.4% to 87.6%.

Fig. 3(b) gives the results of the lean solvent temperature study. As the lean solvent temperature increased from 30 °C to 50 °C, both the vapor-form and aerosol-form amine emissions increased. The MEA emissions after the absorber increased from 625 mg/Nm^3 to 2460 mg/Nm^3, and MEA emissions after the water wash column increased from 108 mg/Nm^3 to 211 mg/Nm^3. The water wash column amine control efficiency raised from 82.7% to 91.4%. It is because the vapor-form amine accounted for a higher emission proportion in the high lean solvent temperature. Some model studies and bubbling experiments had the same conclusion that increasing lean solvent temperature led to a rise in amine emissions. However, these results were contrary to Khakharia's work[23]. His-results showed that increasing the lean solvent temperature led to a reduction of supersaturation at the top of the column.

These inconsistencies may be caused by different sizes of absorbers. As discussed above, the CO_2 absorption reaction significantly promotes the growth of aerosol. The reaction intensity in the absorber changed by lean solvent temperature is the key. When the absorber has sufficient contact area and residence time for flue gas, the absorption is mainly affected by reaction kinetics, the reaction intensity decreased with the increase of lean solvent temperature. While in the bench-scale test, the CO_2 absorption reaction did not reach equilibrium, the reaction intensity is mainly controlled by the thermodynamics. Therefore, it leads to the opposite effect.

Fig. 4 shows the relationship between lean solvent CO_2 loading and emissions. Both the MEA emissions and total particle mass significantly decreased with lean solvent CO_2 loading increased. It is an effective strategy to prevent the rapid growth of aerosol. While the CO_2 capture rate was decreased as the reactivity of solvent reduce. And the downward trend of MEA emissions after the water wash column slowed down when the lean solvent loading was higher than 0.34 mol/mol. Increasing the lean solvent CO_2 loading appropriately combine with absorption enhance strategy could be a feasible way to restrain the aerosol growth in the absorber.

It is also very interesting that aerosol emissions have same changing trend with column highest temperature. Fig. 5 gives details about the column temperature. It's obvious that the absorber column temperature profile is mainly affected by the inlet solvent temperature and chemical reaction. As the CO_2 concentration increased and lean solvent loading reduced, the absorption reaction became more intense and released more heat, and the temperature in the middle and lower part of the column increased. And the lean solvent temperature significantly affects the absorber column temperature profile. The overall column temperature is slightly higher than the previous MEA/AMP test.

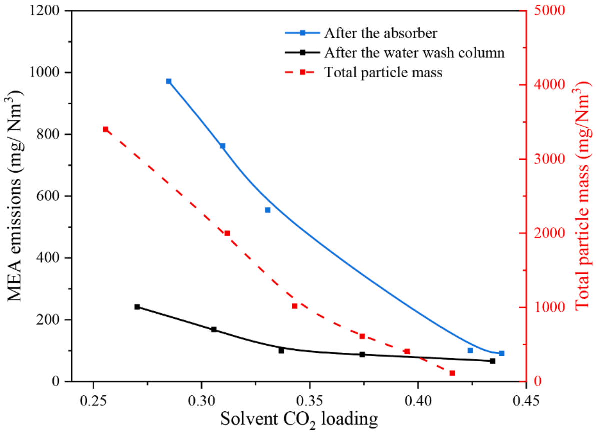
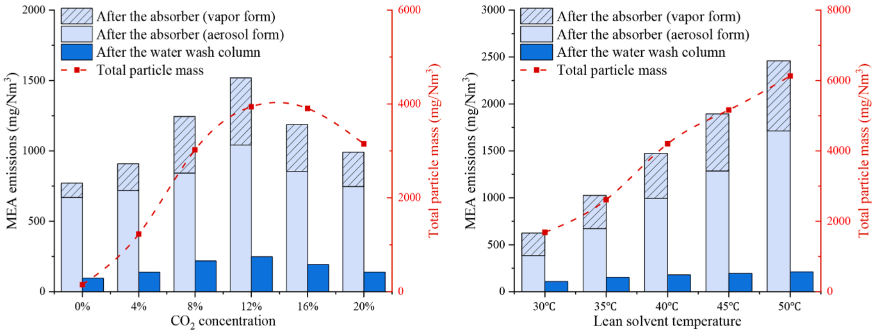


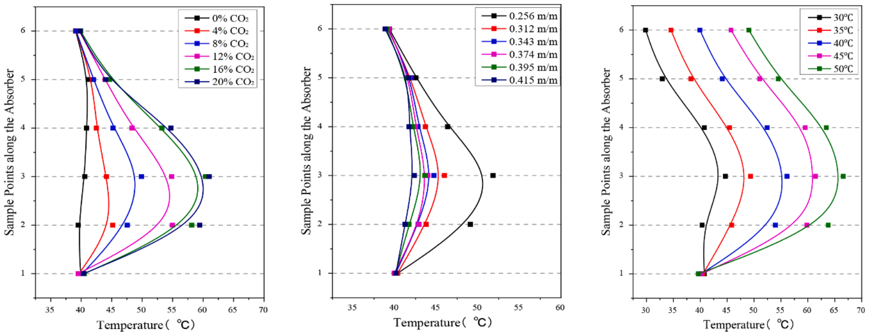
Fig. 4. Amine emissions and aerosol particle mass studies under different solvent CO_2 loading.



(a)

(b)

Fig. 3. Amine emissions and aerosol particle mass studies under different parameters (a) flue gas CO_2 concentration, (b) lean solvent temperature.



(a) (b) (c)

Fig. 5. Absorber temperature profiles during the parameter studies: (a) flue gas CO_2 concentration, (b) lean solvent CO_2 loading, and (c) lean solvent temperature (Sample points 1 to 6 are located from bottom to top of the absorber).

3.3. *Aerosol particle size distribution*

When the flue gas before the absorber contains high concentration nuclei (10^7 cm^{-3} level), the aerosol-form amine contribution over 70% of total amine emissions. To have insight into how these amine solution micron droplets are affected by CO_2 absorption system conditions, it is necessary to measure and analyze their particle size distribution. The aerosol generation conditions of simulated flue gas remain unchanged, and the distribution of nuclei before the absorber is the same as that in Section 3.1. This section investigated the same three parameters discuss above, the particle size distribution after the absorber was recorded by ELPI+ with a 1:8 diluter after 20 min of stable operation.

Fig. 6(a) gives the particle size number distribution of flue gas at the absorber outlet vary with the CO_2 concentration. The shape of the particle size distribution curve was similar when the CO_2 concentration of flue gas was between 8–20%. When CO_2 concentration was at 12%, particles with a diameter larger than 591 nm accounted for 52.2% of the total number. While in the no-CO_2 condition, only 7.6% of particles had a size larger than 591 nm. This phenomenon indicated that the aerosol growth in the absorber relies on the existence of CO_2. When the flue gas CO_2 concentration was lower than 8%, the aerosol growth was significantly retarded by the insufficient CO_2, and the distribution of particles moved to a smaller size. Fujita's solvent bubbling test also found the CO_2 in bubbling gas promoted the growth of aerosol. His-results also showed the particle number concentration increased when CO_2 exist(Fujita et al., 2014). However, the phenomenon was not obvious in this test.

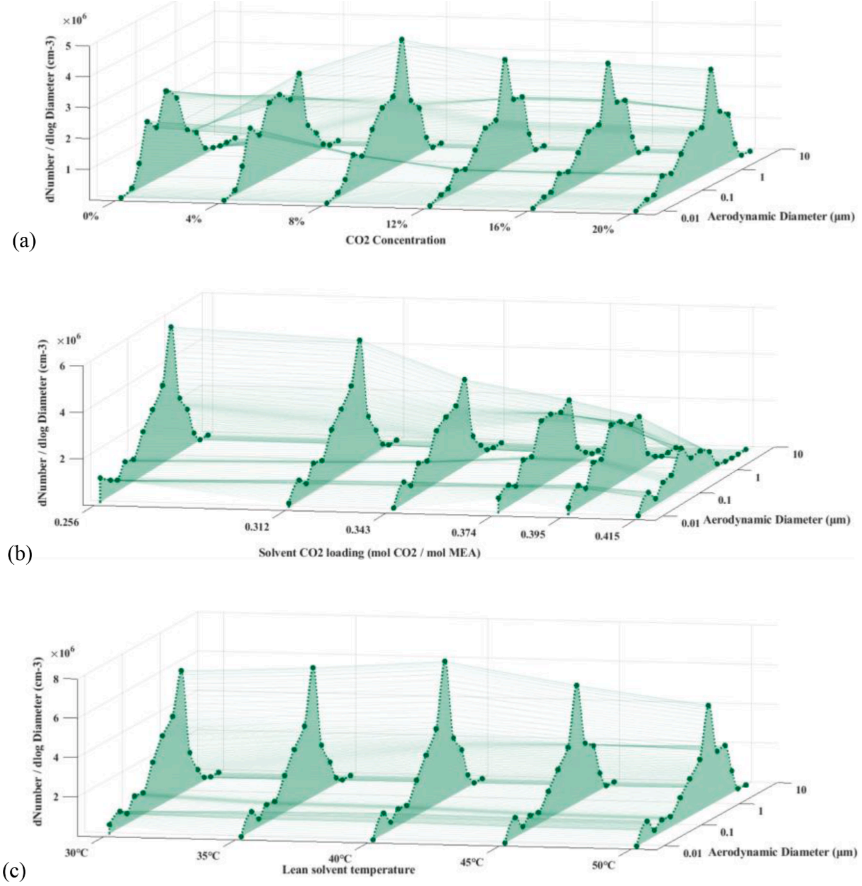


Fig. 6. Particle size number distribution after the absorber varied with different parameters (a) CO_2 concentration, (b) solvent CO_2 loading (c) lean solvent temperature.

The lean solvent CO_2 loading affects the CO_2 capture rate and the intensity of the absorption reaction. As shown in Fig. 6(b), when the CO_2 loading increased from 0.256 mol/mol (basic condition) to 0.415 mol/mol, the particle size number distribution peak shifted from 733 nm to 128 nm gradually, and the particle size larger than 591 nm decreased from 39.5% to 11.4%. It should be noted that the total particle number was also decreased with CO_2 loading raised. This phenomenon was also observed in Fujita's test (Fujita et al., 2014). Interestingly, the total particle number didn't change in his bubbling test when the solvent CO_2 loading was between 0 mol/mol and 0.28 mol/mol.

In the lean solvent temperature study, although the peaks of size number distribution were stable at 733 nm, the particle distribution shift still can be observed by comparing number concentration at the same diameter. As shown in Fig. 6(c), the particle number concentration in the range of 19 nm – 591 nm was ranked in 30 °C > 35 °C > 40 °C > 45 °C > 50 °C, while the number concentration in the range of 1630 nm – 5380 nm was the exact opposite. It indicates increasing the lean solvent temperature can promote aerosol growth and lead to the formation of more micron-sized particles

Compare with particle size number distribution, the mass distribution directly reflects emissions amount varies with droplet diameter. It can more clearly show the impact of particle growth. As shown in Fig. 7(a), the bold dashed line shows the particle size mass distribution. And the area of the blue surface under each bold dashed line represents the total particle mass under certain CO_2 conditions. Particles with a diameter larger than 591 nm contributed over 95% total mass in the presence of CO_2. While in the no-CO_2 condition, this ratio decreased to 88.4%. As mentioned above, increasing CO_2 could promote aerosol growth when CO_2 concentration was in the range of 0% - 8%, which is why the total particle mass after the absorber increased so dramatically. Interestingly, when the CO_2 concentration increased above 12%, total particle mass slightly decreased. This was probably due to the high average CO_2 loading of the solvent in the absorber. Fig. 7(b) shows that the mass concentration after the absorber decreased rapidly with the increase of lean solvent CO_2 loading. Meanwhile, the CO_2 capture rate dropped from 88.4% to 30.6%. It can be seen that large particles still contribute most of the mass emissions. The particle size between 1 and 5 μm accounted for 82.1–94.2% mass when the particle emissions were high (above 1000 mg/Nm^3). Fig. 7(c) shows the particle size mass distribution is also unimodal at 2–3 μm. The total particle mass change increased significantly as the number distribution shifted to a larger size. When the lean solvent temperature increased from 30 °C to 50 °C, the mass concentration peak raised from 3113 mg/Nm^3 per logD (log Diameter) to 16,125 mg/Nm^3 per logD and shifted from 2011 nm to 3013 nm. The particle mass had three power relations with its diameter, that was why the particle mass distribution of most conditions was concentrated in micron level (1μm-5 μm). The particles with a size larger than 911 nm only account for 22.6% of the total number, but it contributed more than 92.3% of the total particle mass. Therefore, the rapid growth of aerosol in the absorber resulted in the formation of high-quality aerosol particles in the flue gas.

From the perspective of amine emissions control, it is good news that large particles contribute to most aerosol emissions. Traditional mist eliminators like the mesh-type coalescer can remove particulate matter by inertia and impact. These demisters have nearly 100% efficiency in eliminating particles with a diameter larger than 3 μm, but their efficiency of removing small particles is very low (Fang et al., 2020).

If the CO_2 absorption system is equipped with a water wash column

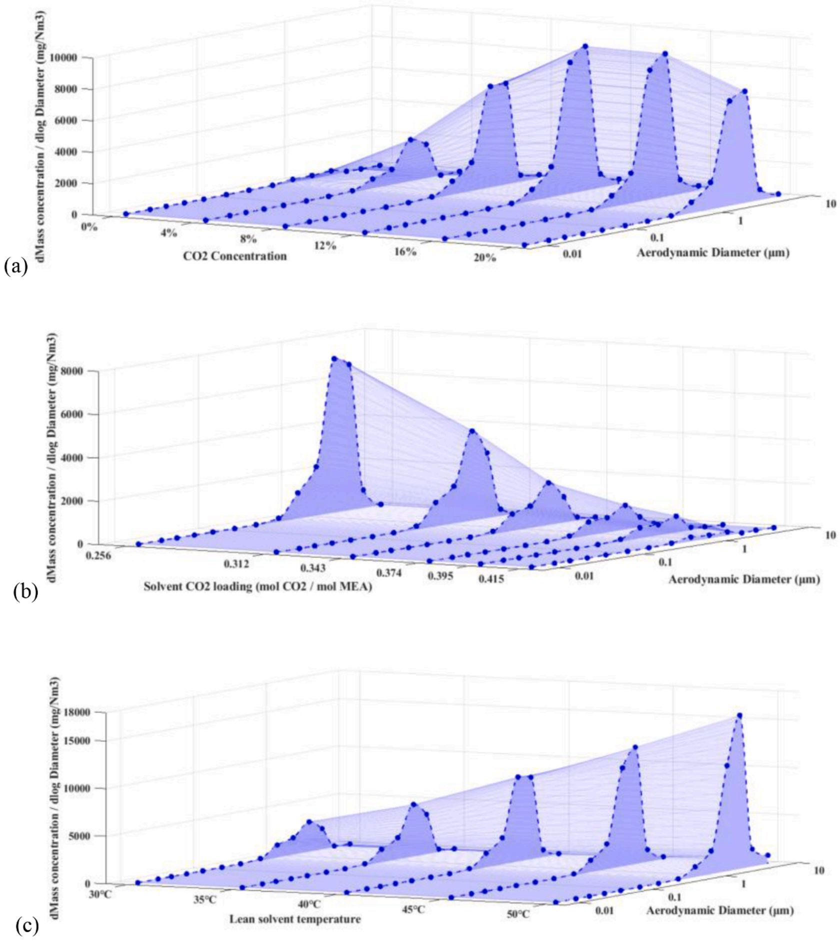


Fig. 7. Particle size mass distribution after the absorber varied with different parameters (a) CO_2 concentration, (b) solvent CO_2 loading (c) lean solvent temperature.

and a demister. Considering the particles can grow fast in the water wash section(Kang et al., 2017; Majeed and Svendsen, 2018a). The absorber outlet particles with a size less than 1 μm (PM1) will be the main threat. Therefore, the PM1 emissions under different parameters study were compared (Fig. 8). With the increase of lean solvent temperature, the PM1 emissions had no obvious change trend. This was quite different from total amine and particle emissions, which increased rapidly. While the PM1 decreased with the increase of solvent CO_2 loading, the increase of PM1/PM10 ratio indicated that the aerosol growth was slowed down and particles concentrated in a smaller size. This phenomenon was also observed in no CO_2 condition, the PM1/PM10 ratio was up to 30% and PM1 is as low as 50 mg/Nm^3.

There are mainly three strategies to control aerosol emissions in CO_2 absorption system: 1) remove the nuclei in flue gas before the absorber; 2) slow down the aerosol growth and restrain amine transfer between solvent and aerosol particles in the absorber; 3) promote the aerosol growth combine with anti-mist technology.

The first strategy is effective because the nuclei in flue gas significantly affect amine emissions. However, the cost of controlling submicron nuclei by using the Brownian demister unit (BDU) or wet electrostatic precipitator (WESP) is relatively high(da Silva et al., 2013) (Mertens et al., 2014a).

Strategy 2 and Strategy 3 are incompatible. Based on the result of particle size distribution, particle emissions became low only when the CO_2 absorption reaction intensity is low. So, to apply Strategy 2, part of the system's chemical absorption ability must be sacrificed. It is very contradictory for system condition optimization.

Strategy 3 is very promising to control aerosol emissions. However, increasing the aerosol growth speed in the absorber won't be a good choice. Increasing the lean solvent temperature, decreasing the solvent CO_2 loading, and adjusting the flue gas CO_2 concentration close to 12% was good for aerosol growth in the absorber. But Fig. 7 shows the PM1 emissions won't reduce because more ultra-small particles will grow up to near 1 μm. These submicron particles also contain a high concentration of amine. Besides, when the aerosol grows faster in the absorber, more large particles (> 1 μm) will carry amine into the water wash cycle. The same problem may be encountered in promoting aerosol growth before the absorber. Therefore, it is more recommended to accelerate aerosol growth when it passes through the absorber. For example, adopt multistage water wash, low-temperature water wash, or steam injection. Assuming Strategy 3 can improve particles' efficient control size from 1 μm to 0.6 μm at the outlet of the absorber. It will reduce 65% - 86% of particle mass emissions, which is quite efficient in controlling aerosol-form amine emissions.

3.4. *Continuous monitoring experiments*

The flue gas emission characteristics under stable working conditions were discussed. In this section, the particle number and average particle size were monitored during parameters changed. The result shows the flue gas emission characteristics change rapidly with the system conditions. The change of Sauter mean diameter dominates the total particle mass after the absorber. While there was no obvious connection between particle size and particle number concentration.

As shown in Fig. 9(a), the CO_2 concentration of flue gas changes from 0% to 20% at a 4% interval. Each condition was maintained for approximately 4 min. The CO_2 was controlled by the gas mass flowmeter. When the CO_2 changed from 0% to 4% and then to 8%, the Sauter mean diameter of particles also sharply increased. The response time of particle size to the change of CO_2 conditions is only approximately 20 s, which is equal to the time that flue gas passed through the absorption system and go into the ELPI+. The total number concentration of particles has a fixed period of fluctuation. Maybe related to the intermittent operation of air compressor. Its overall trend increased at beginning of 400 s, then decreased slightly for the rest of the time.

Regarding the solvent loading study, the electric heating of the stripper was turned off to increase lean solvent CO_2 loading gradually. As shown in Fig. 9(b), the CO_2 loading of the lean solvent increased with time and it tended to be gentle when it was close to full loading (0.5 mol/mol). The Sauter mean diameter of particles decreased from 1.8 μm to 1.0 μm, and the total number concentration decreased from 5.0×10^6 to 3.9×10^6 cm^{-3}. The mutual reduction of these two values led to a rapid decline in total particle mass. The lean solvent temperature was changed by the cooling water flow rate. Fig. 9(c) gives particles monitoring results. When the lean solvent temperature increased, the total number concentration fluctuated and decreased slightly. While the Sauter mean diameter of particles went steadily up. The total particle mass has the same trend as the Sauter mean diameter due to they have three power relation.

4. Conclusions

Aerosol growth in the absorber is one of the main reasons for high-quality aerosol emissions. The aerosol particle concentration dropped to its 1/10 when it passed through the absorber, and the peak of particle size distribution changed from 16–41 nm to 733 nm. It proved the particle coagulation and aerosol growth did happen in the absorber.

The intensity of the absorption reaction has a significant effect on the growth of aerosol. In the 3.6 Nm^3/h test platform, the absorber is not long enough to reach reaction equilibrium, which means the reaction is mainly controlled by thermodynamics. The particle number size distribution after the absorber shifted to a larger size as the lean solvent temperature increased from 30 °C to 50 °C, and the MEA emissions after the absorber and water wash column increased accordingly. The aerosol grew fastest when the CO_2 concentration was 12% because the solvent and CO_2 had a perfect ratio to fully react. Decreasing solvent CO_2 loading also led to the particle size number distribution peak shifting from 128 nm to 733 nm, and the MEA emissions increased from 91 mg/Nm^3 to 971 mg/Nm^3.

The aerosol particle mass and amine emissions have the same trend during parameters studies, which proved aerosol emissions dominated the amine emissions in the high nuclei scenario. Conventional CO_2 capture systems are usually equipped with demister. To eliminate the main threat of aerosol emission after absorber (PM1), it is recommended to promote aerosol growth after the absorber rather than restrain it in the absorber. It should be noted that promoting aerosol growth before or

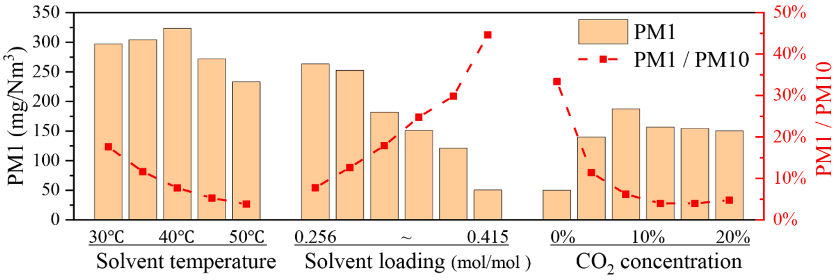


Fig. 8. PM1 and PM1/PM10 ratio after the absorber varied with different parameters.

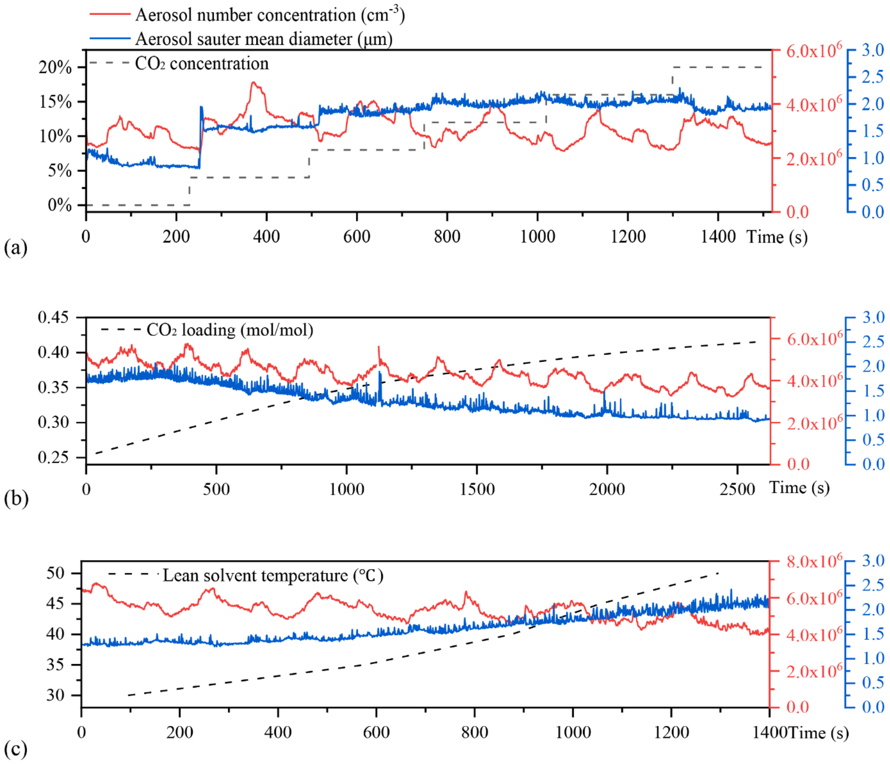


Fig. 9. Continuous monitoring during the parameter studies (a) flue gas CO_2 concentration, (b) solvent CO_2 loading, and (c) lean solvent temperature.

in the absorber may lead to more PM1 emissions.

CRediT authorship contribution statement

Ningtong Yi: Conceptualization, Methodology, Writing – original draft, Investigation. **Mengxiang Fang:** Supervision, Writing – review & editing. **Wentao Di:** Methodology, Investigation. **Tao Wang:** Writing – review & editing. **Xiang Gao:** Validation. **Wei Zhang:** Funding acquisition. **Chunliang Ge:** Funding acquisition. **Jingjuan Yuan:** Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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