Dry Sorbent Circulating Reactor System for Removing Acid Gas Pollutants Emissions from Power and Incineration Utilities

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Abstract. Dry type acid gas pollutants removal processes offer the significant advantages of low capital and operating costs when compared to wet type acid gas removal processes. They hold great potential for the economical reduction of SO₂ and HCl emissions from power and incineration utilities that use high-sulfur coal and high halide base acids wastes. One of the project’s major goals was the development of dry, calcium-based sorption processes for removing sulfur dioxide and hydrogen chloride from the combustion gases and incineration gases produced by high-sulfur coal and high halide base acids wastes. Dry sorbent circulating reactor system for flue gas cleaning highlights a number of experimental research findings that have had a significant and lasting impact in term of scientific understanding. For example, the experimental investigation in demonstration test unit SO₂ and HCl capture by dry sorbent obtained removal efficiency more than 99%, thereby revealing the well fluid mixing with sorbent and flue gas and longer residence time in the reactor. The average HCl concentration at the inlet of the reactor maintained at 878 ppm. The HCl removal efficiency maintained at over 99.9%, and the HCl emission concentration maintained at below 5 ppm under the conditions that the flow amount of the flue gas was 40,000 Nm³/hr, the SR was below 2.0, and the pressure loss at the baghouse was 150 mmH₂O. We also identified a number of important areas for future research, including reaction mechanisms, sorbent material, transport effects, simulation of particle fluid dynamics and efficient system development.

1. Introduction

Until recently, two types of the acid gas removal units were mainly used in industrial flue gas cleaning, namely the wet scrubbing and spray-dry absorber systems. Wet scrubbers use water solutions containing scrubbing agents for removal of acid gases. Thus cleaned flue gas passes through the scrubbing system and as by-product leaves the wastewater, which then treated on-site before disposal. This system has proven over the decades to be a very efficient method of flue gas cleaning but at very high cost for operators. The system itself has a short lifespan due to the corrosion problems.

Semi dry type, known as spray dry absorber (SDA), has used to replace the wet scrubbers at the cost of higher sorbent consumption. For this scrubber there is no need for waste treatment facility since the slurry injected at the top of the reactor dried while circulating inside the reactor. While
rotating inside the reactor, sorbent particles carried by water drops until they dry completely leaving the sorbent as dry particulates exiting the system. Due to the humid nature of the particles and the rotating motion created by the baffles at the inlet of the unit, one part of the not completely dried particles will attached to inner walls of the reactor. That is why this system is due for frequent maintenance. Due to the low reaction rate of the sorbent, this system is known for high sorbent consumption.

Operators of incineration and power plants have extensively applied dry type reactors and duct injection systems in the recent time. Duct injection of dry sorbent brings many advantages but it is dependent on the proper mixing at the injection point since the residence time is generally short before the sorbent particles are collected from the flue gas stream by means of filtration (electrostatic precipitation or fabric filtration). Figure 1 illustrates the field of application for three main flue gas purified systems. Compared to SDA, the dry type reactor can achieve high efficiency rates at low Ca/S molar ratios as described in the figure 1. This in return means that the dry type reactor will require less sorbent amount for the same acid gas amount and less saturated sorbent for disposal after it has been collected by filtration units.

**Figure 1.** Comparison of various scrubbing units regarding their removal efficiency according to a) acid gas loading and b) acid gas to sorbent molar ratio.

Compared to other reactors with high particulate load, such as the fluidized bed reactor, dry type reactor has an advantage of higher reaction rates at lower reactor pressure drop. This is mainly due to its simple inner structure and optimal sorbent-flue gas mixing rates. A comparison figures are displayed in figure 2. Dry sorbent circulating reactor can achieve high acid gas removal efficiencies at lower pressure drop compared to other similar dry type reactors.

Dry type sorbent circulating reactor developed to enhance the removal efficiency of this reactor by lower amount of sorbent injection. Higher removal efficiency can be reach by optimizing the mixing zone inside the reactor to ensure that the sorbent perfectly reacted with acid gaseous. Due to its simple structure, the reactor can easily applied to replace exist spray dry reactor in an existing flue gas treatment systems or designed for completely new line. In combination with bag filter, the overall acid gas removal efficiency can be increased [1-5]. During the development stage, various operating conditions tested to ensure that the reactor is fit for application in industrial sites with highest performance. As hardware, the reactor has so far well optimized and it remains to be checked how the performance is dependent on the sorbent properties and its reactivity.

After tests with laboratory scale and pilot scale test units the performance of the new dry sorbent circulating reactor was evaluated directly at the industrial site. The facility was tested using calcium hydroxide sample C.
2. Experimental setup and methods

2.1. Experimental setup
This study includes tests conducted in the laboratory scale fixed bed test unit for determination of sample adsorption capacity and operated the pilot scale test unit for determination of SO$_2$ and HCl removal efficiency according to various sorbent samples. Finally, the sample C tested in combination with new dry sorbent circulating reactor at the industrial site.

The fixed bed test unit is displayed in the figure 3 and shows the test gas injection unit including the gas cylinders, gas flow control devices and the control valves, the fixed bed reactor with electrical heating jacket, temperature sensors and the fixed bed, the gas analyze device and the data acquisition unit.

Figure 2. Comparison of the various reactor types and their characteristics.

Figure 3. Flow chart of the laboratory scale fixed bed reactor.

The flow diagram of pilot scale test unit is shown in figure 4. This test unit consist the following functional units. Flue gas generation parts are includes gas burner, force draft fan, burner and the
water injection unit for steam generation. The dry sorbent circulating reactor (SRx) includes settling chamber, sorbent injection ports, temperature and pressure drop sensors. The sorbent feeding unit includes solid particle feeder, particle ejector and high pressure purified compressed air lines. The bag filter unit consists bag filter cases, pleated filter bags, bag cleaning unit, collected dust box with rotary valve and temperature and pressure drop sensors. The flue gas flow control unit includes average pitot tube for measuring flow rate, pressure sensor, temperature sensor and pressure drop sensor, frequency inverter, induced draft fan and stack. The system also includes the gas sensing ports where the concentration of acid gases contained in the composition of flue gas online measured by the portable FTIR gas analyzer (DX4000, Gasmet Technologies Inc.).

In the figure 4, the acid gas sampling ports indicated. These ports used for online measurements alternatively to check the base inlet acid gas concentration, without injection of sorbent, and the continuous concentration, by injecting the sorbent at certain sorbent to acid gas (Ca/S) molar ratios.

**Figure 4.** Flow diagram of pilot scale test unit with indication of acid gas concentrations measurement ports.

In the figure 5, a real image of the test pilot test system is depicted.

**Figure 5.** Real image of the pilot scale test unit.

### 2.2. Test material

For lab scale unit fixed tests 5 samples were prepared. The physical properties of the samples A to E displayed in the table 1. For tests with the pilot test unit only samples A, B and C were tested.
Particularly, the active surface area for sorption, based on the considered Brunauer-Emmett-Teller (BET) theory as one of most important properties for adsorption process.

Table 1. Sorbent samples and their physical properties.

| Sample/ Parameter          | A     | B     | C     | D     | E     |
|---------------------------|-------|-------|-------|-------|-------|
| CaO, %                    | 73.0  | 72.8  | 71.5  | 72.5  | 71.9  |
| Ca(OH)$_2$, %             | 96.5  | 96.1  | 94.5  | 95.8  | 95.0  |
| 325M (Pass 45μm)          | 67.8  | 84.0  | 86.0  | 93.0  | 90.6  |
| H$_2$O (300°C), %         | 1.1   | 1.6   | 1.1   | 0.6   | 0.9   |
| Bulk Density, g/ml        | 0.484 | 0.500 | 0.430 | 0.570 | 0.500 |
| Tap. Density, g/ml        | 0.579 | 0.640 | 0.560 | 0.710 | 0.660 |
| BET Surface, Area, m$^2$/g| 41.6  | 45.1  | 38.4  | 47.0  | 33.2  |
| BJH Pore Volume, cm$^3$/g | 0.19  | 0.21  | 0.19  | 0.18  | 0.17  |
| BET Pore Size, Å          | 188.2 | 186.2 | 203.2 | 154.2 | 212.6 |

SEM images of samples A, B and C are displayed in the figure 6.

![SEM images](image)

**Figure 6.** SEM Images of three samples used for pilot plant tests.

3. Results and discussions

First, the calcium hydroxide samples with different physical properties tested using the lab scale fixed bed test unit. In order to evaluate the acid gas sorption capacity, a sample was inserted in the fixed bed cup holder and the unit was heated until the setting temperature of 160°C was reached. Then SO$_2$ gas was fed until the setting concentration of 800 ppm stable value was reached. The complete saturation of the samples reached when the outlet concentration matches with the inlet SO$_2$ concentration. The shape of the SO$_2$ trend curve gives an indication of when the breakthrough point reached, from which point the concentration starts to decrease slowly until complete saturation. The physical properties of the sorbent are therefore very important for high acid gas removal efficiency. In case of sample A, the breakthrough point reached faster than other samples but it takes longer time until the complete sorbent saturation reached. The results displayed in the figure 7.
Figure 7. SO$_2$ outlet concentration for all 5 calcium hydroxide samples of various origins.

In figure 8, the HCl concentration measured at the outlet of SRx (figure 8a) and at the outlet of bag filter (figure 8b) displayed. Samples B and C have similar behavior. Better results achieved with sample A. Inside the SRx the HCl concentration continuously decreases with the increase of CA/S molar ratio for all samples. In case of the measurements at the outlet of the bag filter the complete saturation of the sorbent reached at the Ca/S of 2.5 from which point on there is no significant decrease of HCl concentration visible.

(a)                                                                       (b)

Figure 8. HCl concentration measured a) at the outlet of the SRx reactor and b) at the outlet of bag filter unit for temperature of 130 °C inside the SRx reactor.

Since the acid gases SO$_2$ and HCl fed into the pilot plant system at the same time, the SO$_2$ removal efficiency is considered as the reaction of secondary order. Due to the higher formation energy of CaCl$_2$ as results of the reaction between HCl gas and the Ca(OH)$_2$ solid particles. As the reference [6] shows, there is a competitive reaction between SO$_2$ and HC lags when the two acid gases are in the same system. Therefore, a significant SO$_2$ removal in the system is only visible when the sorbent fed at higher Ca/S molar ratios. Sample A exhibits better results showing SO$_2$ concentration decrease starting from Ca/S of 2. In case of the reaction inside the bag filter, better performance of the sample A is clearly visible. In case of samples B and C their removal efficiency remains poor even at high Ca/S molar ratios as can be seen in the figures 9a and 9b.
Figure 9. SO$_2$ concentration measured a) at the outlet of the SRx reactor and b) at the outlet of bag filter unit for temperature of 130 °C inside the SRx reactor

The results from the test with industrial scale dry type reactor are displayed in figure 10. The average HCl concentration at the front end of the reactor maintained at 878 ppm, and that at the rear end maintained at 98 ppm. The HCl concentration on the tele-monitoring system (TMS) maintained at 0 ppm and no more than 5 ppm. The reason that the SRx had high HCl removal efficiency was presumably because the flow velocity of the flue gas was reduced and its residence time was increased; thus, the contact time between the gas and the reactant, Ca(OH)$_2$ was increased. The increase in the gas-solid contact time deemed to have contributed to the increase in the HCl removal efficiency.

Figure 10. HCl removal efficiency at dry sorbent circulating reactor (SRx), baghouse and the total HCl removal efficiency.

4. Conclusion
In order to simulate the real conditions at the waste to energy plant, acid gases SO$_2$ and HCl injected into the system upstream the dry sorbent circulating reactor. The gases pass through a static mixer installed inside the duct and then enter the dry sorbent circulating reactor where the gases mixed with the sorbent particles before exiting the reactor. The measurements at the reactor outlet and the bag filter outlet used to check the overall acid gas removal efficiency as well as the efficiency of the single unit, dry sorbent circulating reactor and bag filter. After the sorbent particles escaped from the reactor, the inflow sorbent particles occurred the secondary reaction with acid gaseous on the surface of filter bag and inside the particle cake formed on the surface of the filter bags. The contribution of the bag
filter to overall acid gas removal efficiency relatively lower compared to the reaction inside the dry sorbent circulating reactor due to the short residence time the acid gas pends inside the particle cake and reacts with the sorbent. In case of the industrial scale system tests, it sums to around 10% contributed to total HCl removal efficiency. The average pressure drop of industrial scale system maintained 150 mmH₂O.

Results show that the sorption performance is dependent on the physical properties of each sample and the most important of them turn out to be the active surface area. Test with fixed bed show that best results reached with samples of small pore size and large active BET surface area.

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