

HCl removal characteristics of calcium hydroxide at the dry-type sorbent reaction accelerator using municipal waste incinerator flue gas at a real site

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Abstract—A dry-type reactor was used to reduce the HCl emitted from a municipal waste incinerator. A real-scale reactor was installed downstream of the existing semi-dry reactor (SDR) as part of D local incineration plant, whose HCl removal efficiency was investigated by applying calcium hydroxide ($\text{Ca}(\text{OH})_2$). A test dry-type sorbent reaction accelerator (SRx) was designed, and the stoichiometric ratio (SR) of the sorbent and the secondary reaction characteristics of unreacted $\text{Ca}(\text{OH})_2$ in a baghouse were analyzed. For the results, the SRx showed higher HCl removal efficiency and more economic benefits than the other existing flue gas treatment facilities of the incinerator plant. Furthermore, the highest reduction efficiency of HCl was obtained when the differential pressure in the baghouse was maintained at 150 mmH₂O. This is because as the unreacted sorbent attached to the filter surface layer became thicker and the secondary reaction of the HCl removal increased.

Keywords: Sorbent Reaction Accelerator (SRx), Stoichiometric Ratio, HCl, $\text{Ca}(\text{OH})_2$, Baghouse

INTRODUCTION

The increasing municipal and industrial wastes due to the rapid urbanization and industrialization are becoming serious social concerns. As a leading waste disposal method, landfill is applied to those wastes remaining after separating recyclable and reusable wastes, but it is difficult to select a site for a landfill. As an alternative, incineration has the advantages of reducing the volume and toxicity as well as collecting and using the energy generated during the process. The various pollutants generated due to errors in the design and operation of the incinerator, however, have been reported not only to impact the incinerator itself but also to cause air pollution and to harm the human body when they are emitted to the atmosphere [1].

Most incinerator exhaust gases contain toxic organic compounds, heavy metals, and acid gases. The amount of these gases significantly varies according to the components of the wastes being processed, the incinerator structure, and the incineration and operation methods used. Especially, HCl, an acid gas, has different concentrations depending on the type of incinerated waste and the injection amount, and its concentration sometimes exceeds the set emission limit.

HCl, one of the pollutants emitted from incinerators, is known to cause low-temperature corrosion that can easily corrode metals after being condensed when the dew point temperature drops below about 140 °C, and to cause high-temperature corrosion due to the chemical reactions with flue gases when the temperature exceeds

371 °C. It is known that Cl_2 generates iron chloride by reacting with the iron on the pipe wall, and that the generated iron chloride generates iron oxide by reacting with the oxygen in the flue gases [2]. The HCl emitted to the atmosphere stimulates the mucous membrane of the upper airway in the human body by generating hydrochloric acid mist when combined with water vapor during respiration, to cause cough and dyspnea, and to act as a factor that accelerates the generation of dioxins and furans [3].

As a method of properly removing HCl, an acid gas, the wet method has relatively high removal efficiency compared to the dry method, but it requires a broad installation area and involves disposal costs for the generated wastes due to unresponsive agents as well as maintenance costs due to corrosion and other reasons. However, the dry method has the advantage of using the energy of flue gas, reducing the generated gases, and removing other compounds, such as heavy metals, although it has relatively low removal efficiency. Using a baghouse with the dry method can increase such method's removal efficiency, and using it with the wet method can address its various disadvantages [4-8].

The existing $\text{Ca}(\text{OH})_2$ and NaOH were replaced with the highly reactive $\text{Ca}(\text{OH})_2$ to increase the low removal efficiency of the dry dust collector, and a pilot-scale dry sorbent reaction accelerator was installed to increase the reaction rate. The efficiency and performance of this system were evaluated in this study. Table 1 presents the advantages and disadvantages of both methods.

The purposes of this study were to minimize the equipment handling the air pollutants in municipal waste incinerators (i.e., combustion facilities), to address the disadvantages of the existing equipment, to increase the removal efficiency, to develop a process that can minimize the energy consumption, and to suggest the optimal operating conditions.

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Table 1. Comparison of the dry and wet methods [9,10]

Category	Advantage	Disadvantage
Dry method	① Low construction and operating costs ② Minimal installation space required ③ No wastewater ④ Simple control unit ⑤ Improves efficiency with bag filter combination	① Low removal efficiency compared to the wet method ② Periodic filter bag replacement
Wet method	① Relatively high efficiency compared to the dry method ② Removable water-soluble malodorous substances ③ Very broad way of applying treatment-range exhaust gas	① Large emplacement required ② High construction cost ③ Maintenance cost generated ④ Wastewater treatment plant required

Therefore, the HCl removal characteristics with secondary reaction of the unreacted sorbent in a baghouse according to the SR were investigated by using the operating condition change of a municipal waste incinerator and a dry-type SRx in pilot scale with the injection of highly reactive $\text{Ca}(\text{OH})_2$.

EXPERIMENT

1. Preparation of Samples

Experiments were performed using $\text{Ca}(\text{OH})_2$, a Ca-based reactant, which has excellent HCl removal efficiency and is frequently being used in the field. Performance tests were performed and the physical characteristics were investigated with a reactant provided by SN Co., Ltd.

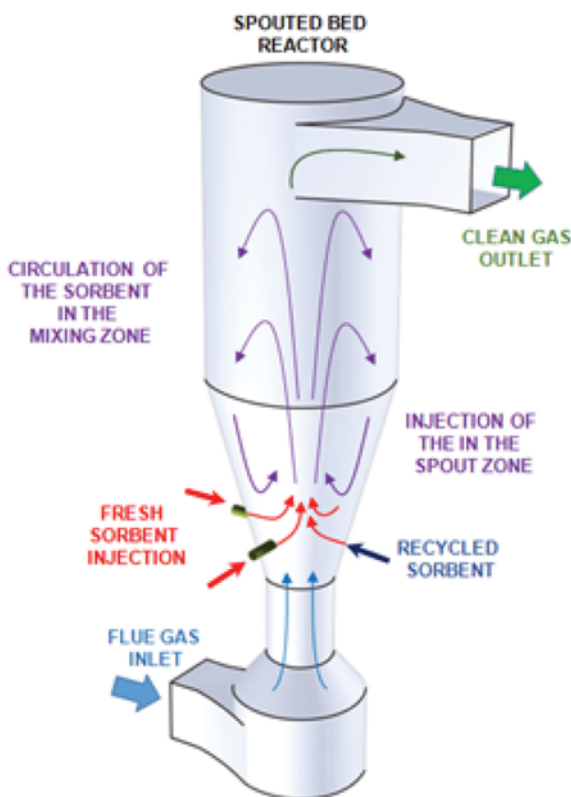


Fig. 1. Structure of dry sorbent reaction accelerator (SRx).

2. Basic Principles and Flowchart of the Dry Sorbent Reaction Accelerator

The dry sorbent reaction accelerator involves a dry flue gas cleaning process to remove certain pollutants like SO_2 , HCl, Hg, heavy metals, dioxins and furans (see Fig. 1). In the dry sorbent reaction accelerator, the acid gas contained flue gas moves through a cylindrical apparatus (spout-fluid bed reactor) from the bottom to the top. The main principle of the apparatus is to bring acid gas in an intensive contact with $\text{Ca}(\text{OH})_2$, activated carbon, water and recirculated material from the baghouse. The spout-bed material is made up of solids, consisting of calcium hydroxide, the reaction products of the flue gas cleaning process and ash from waste incinerators. The fresh $\text{Ca}(\text{OH})_2$ is injected into the conical zone at high velocity; it causes a stream of particles to rise rapidly into cylindrical reaction chamber, and the stream velocity of particles is rapidly decreased. The particles and the acid gas are well mixed and reacted in the cylindrical reaction chamber. The overall flow pattern of particle and flue gas thereby becomes a circulating fluidized bed with upward-moving particles entrained by a cocurrent flow of fluid. A systematic cycle pattern of particle movement is giving rise to a unique hydrodynamics system that is more suitable for certain applications. The acid gas introduced into the dry sorbent reaction accelerator is first removed through the gas-solid reaction with the $\text{Ca}(\text{OH})_2$ injected into the spouted bed forming area. The secondary removal is performed through the reaction of the microdroplets, which are the mixture of low-pressure compressed air and water injected from the low-pressure secondary fluid injection area, and of the acid gas that is not removed in the spouted bed area. Thus, high HCl removal efficiency can be achieved in this design.

Fig. 2 shows the flow diagram of flue gas treatment system for municipal waste incinerator and HCl sampling ports with dry sorbent reaction accelerator (SRx). The HCl concentrations were measured at the front and rear of the SRx, and in the chimney, for comparison. Consecutive real-time measurements were performed first through the mercuric thiocyanate method, a process testing method, and then through the GASMET Dx-4000 analyzer [14], a Fourier transform infrared (FTIR) measurement method.

3. Experiment Set-up and Conditions

The body length of the SRx is 27,100 mm, and its diameter is 3,000 mm. $\text{Ca}(\text{OH})_2$ is injected into the spouted bed forming area, and low-pressure compressed air and water are injected from the low-pressure secondary fluid injection area to remove HCl. The

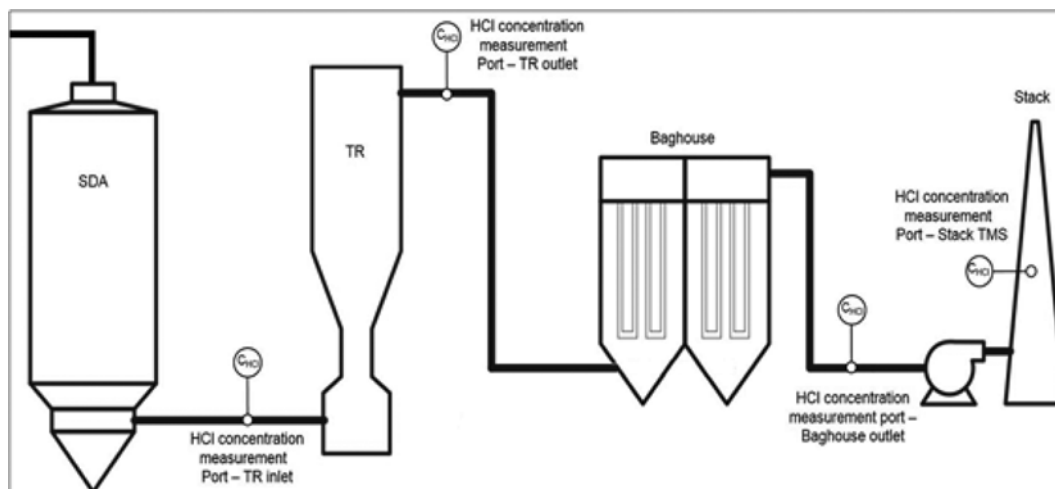


Fig. 2. Flow diagram of flue gas treatment system for municipal waste incinerator and HCl sampling ports with dry sorbent reaction accelerator (SRx) installed in front of the baghouse: (SDA) semi-dry reactor.

Ca(OH)₂ injected into the spouted bed forming area is instantly dried by the introduced gas, and the subsequent gas-solid reaction increases the HCl removal efficiency with strong fluidization. The unresponsive absorbent that is dispersed and discharged due to the fast gas flow is collected and recirculated into the reactor. Thus, the concentration of the highly reactive absorbent in the reactor and the high HCl removal efficiency of over 99% can be main-

tained. The specifications of the SRx are summarized in Table 2.

The sorbent reaction accelerator and the conventional fluidized bed reactor are compared in Fig. 3. The fluidized bed that was used in this study removed the bed substance so that the absorbent and gas had active contacts in the spouted bed, and it was improved into the tapered Y-type grid pattern structure.

Fig. 4 shows the photographs of each unit's installation for flue gas treatment of the municipal waste incinerator. Experiments were performed with the SR as a variable according to the driving condition of the municipal waste incinerator. Table 3 shows the experiment conditions of the SRx.

Table 2. Basic specifications of the dry-type sorbent reaction accelerator (SRx)

Parameter	Range
Gas treatment capacity	Max. 40,000 Nm ³ /hr
Flue gas temperature	180-200 °C
Functional role	Removal of SO _x , HCl
Geometric dimensions	Diameter: 3,000 mm; height: 27,100 mm

4. Measurement Method

Fig. 5 shows two HCl concentration measurement methods. In the first, HCl analysis of the samples was performed using the mercuric thiocyanate method, a standard method for measuring environmental pollution. As a large amount of HCl existed on the in-

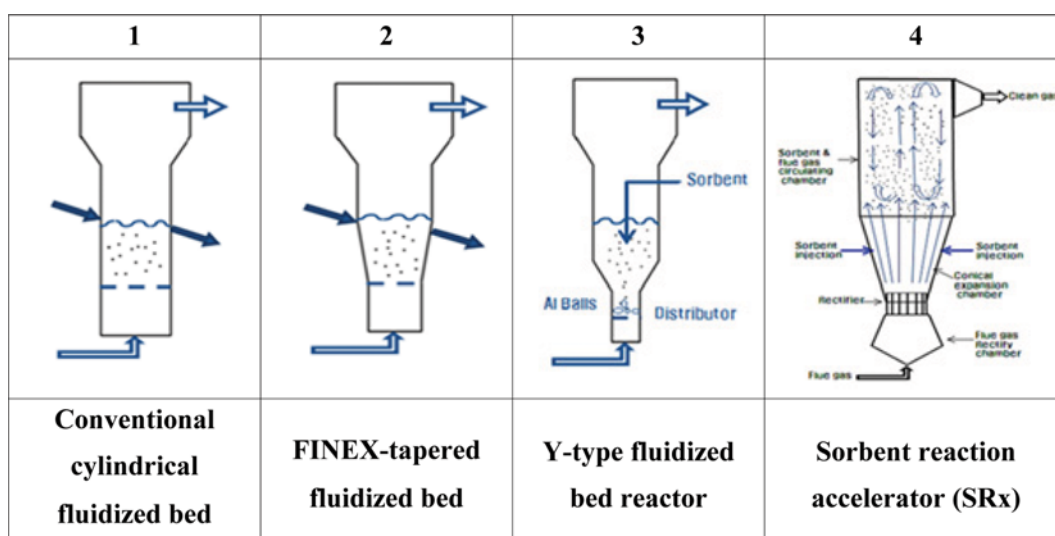


Fig. 3. Configurations of various fluidized bed reactors and a sorbent reaction accelerator.

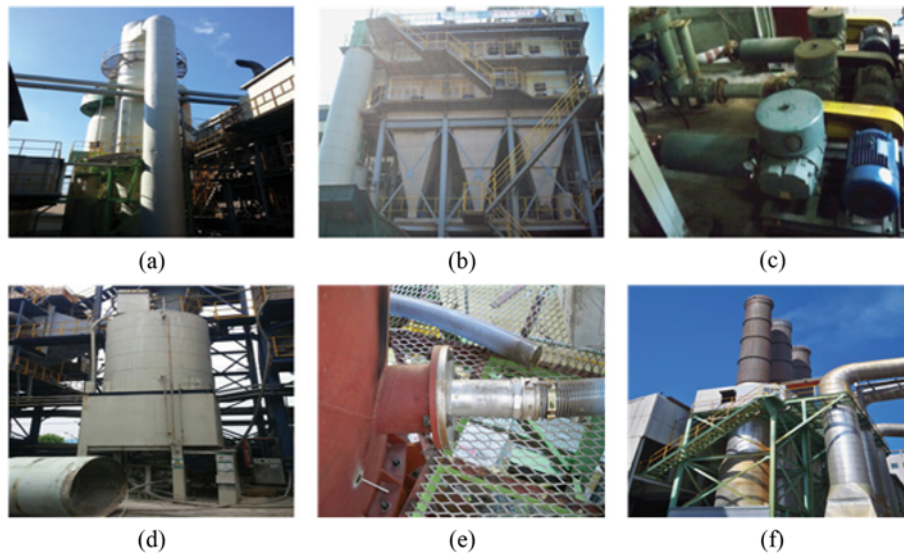


Fig. 4. Photographs of the each units installation for flue gas treatment of the municipal waste incinerator: (a) Dry sorbent reaction accelerator, (b) baghouse, (c) root blower for $\text{Ca}(\text{OH})_2$ injection, (d) $\text{Ca}(\text{OH})_2$ storage silo, (e) $\text{Ca}(\text{OH})_2$ injection nozzle, (f) stack.

Table 3. Experiment conditions

Variables [unit]	Range
Test unit [-]	SRx & bag filter
Test absorbent [-]	$\text{Ca}(\text{OH})_2$
Flue gas temperature [$^{\circ}\text{C}$]	180
Gas flow rate [Nm^3/min]	40,000
Superficial gas velocity [Nm/sec]	2
HCl inlet concentration [ppm]	728-1123
Stoichiometric ratio [-]	1.5-2.0
$\text{Ca}(\text{OH})_2$ feed rate [kg/hr]	72-110

ternal wall of the absorption bulb, it was washed with a liquid. Then 100 ml of the liquid and the washed liquid, respectively, were prepared as sample solutions. 5 ml of each of these sample solutions was placed into test tubes with glass lids, and then 2 ml ammonium ferric sulfate solution, 1 ml mercuric thiocyanate solution, and 10 ml methyl alcohol were added before sealing the test tubes and mixing well. After moving each solution to a 10 mm cell, its absorbance near the 460 nm wavelength was measured by using

an absorption spectrometer. In the second method, the HCl analysis involved the GASMET Dx-4000 analyzer, an FTIR measurement tool. Humid and highly corrosive HCl gas was extracted at a constant temperature of 180°C by a hot-wet method. Moisture existed as dry saturated steam and dusts were sequentially removed at two spots. The spectra of the molecules, except the monoatomic molecules, according to their IR absorption characteristics, were compared with the reference libraries by component and background for optimal qualitative and quantitative measurements within the shortest time period.

RESULTS AND DISCUSSION

1. Theoretical Review

Ca- and Na-based dry reactants for controlling HCl gas are currently widely used. Among them, $\text{Ca}(\text{OH})_2$ is deemed the most suitable absorbent for removing HCl during general waste combustion. It was reported that $\text{Ca}(\text{OH})_2$ was most excellent within the $240\text{--}350^{\circ}\text{C}$ combustion range, and that as the absorbent content increased, the HCl generation was reduced at temperatures under

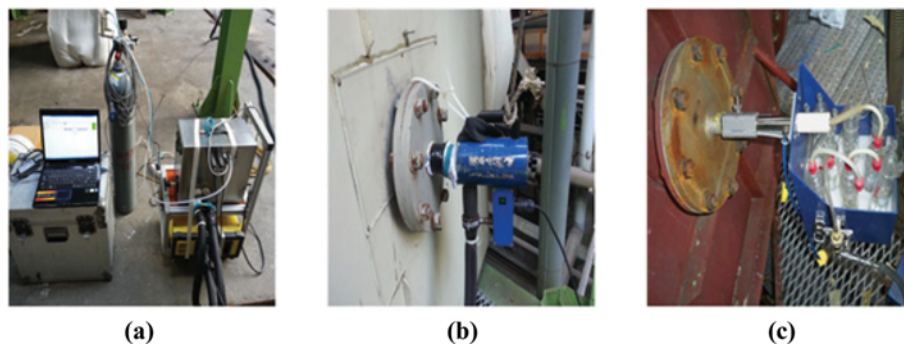


Fig. 5. Gas analyzers for HCl concentration measurement: (a) GasetmTM DX-4000 FTIR gas analyser, (b) sampling probe of DX-4000 FTIR gas analyser, (c) gas analyzer for mercuric thiocyanate method.

700 °C and was increased at temperatures between 700 and 1,100 °C. This was presumably because the CaCl_2 that was generated under 700 °C was decomposed again at over 772 °C, its melting point [11].

In a study on the reaction mechanism between HCl gas and a Ca-based absorbent, the CaO that was generated from Ca(OH)_2 , CaO, and CaCO_3 through formulas (1)-(3) below at 100-600 °C was porous and had a large surface area [12].



It was reported that the coherence increased with the H_2O content at temperatures below 150 °C, and that the equilibrium moisture content decreased with a temperature increase when the relative humidity was constant. At low temperatures of 60-150 °C, the lime

Table 4. Physical properties of the absorbent

Value	Ca(OH)_2
BET surface area (m^2/g)	37.3
Average diameter (μm)	9.82
Cumulative pore volume (cm^3/g)	0.181
Average pore diameter (nm)	10.07

change rate sharply rose according to the increase in relative humidity. The effect of the relative humidity gradually decreased for temperatures above 100 °C, and the effect of the relative humidity on the concentration disappeared at above 200 °C. The coherence increased with the H_2O content at temperatures below 150 °C. The increase in the lime change rate at low temperatures and high relative humidity severed the crystal grids of the lime reactants and formed water [13].

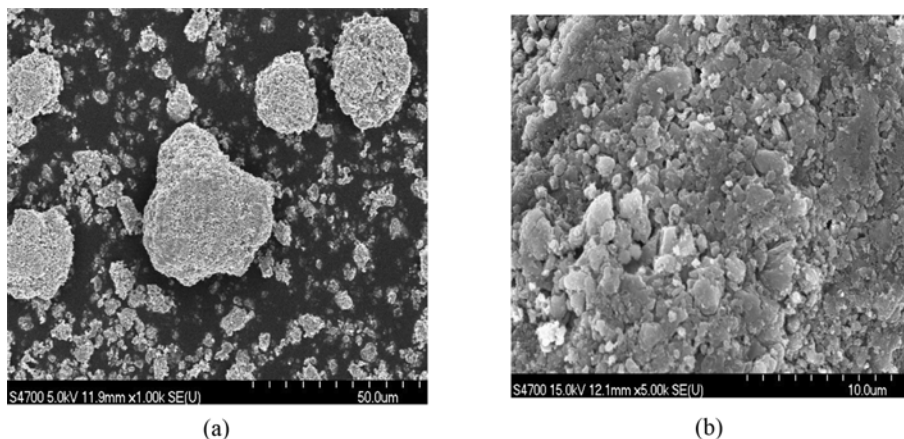


Fig. 6. Scanning electron microscopy (SEM) images of the test sorbent (Ca(OH)_2): (a) $\times 1,000$, (b) $\times 5,000$.

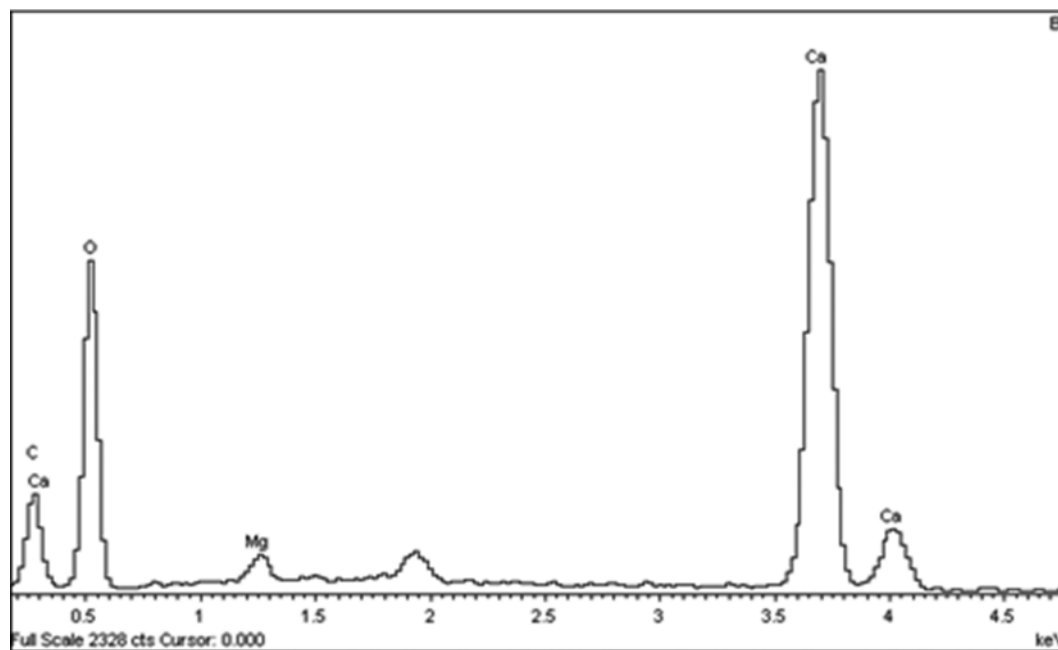


Fig. 7. Energy dispersive spectroscopy (EDS) analysis of the test sorbent (Ca(OH)_2).

2. Physical Characteristics of the Experimental Reactants

2-1. Shape, Characteristics, and Component Analysis of $\text{Ca}(\text{OH})_2$

Fig. 6 shows the SEM(HITACHI S-4700) images of the test sorbent $\text{Ca}(\text{OH})_2$ that was used in the experiments. The magnifications were 1,000 and 5,000. Table 4 shows the BET (Micromeritics Instrument Corporation ASAP 2010) measurement results for $\text{Ca}(\text{OH})_2$. The results of the measurement of the amount of gas that was absorbed by the solid surface with the gas pressure varied under the same temperature showed physical properties such as the BET surface area, average pore diameter, and cumulative pore volume. The CaO content was 74.1 wt%, which was higher than those of the conventional absorbents.

2-2. Energy-dispersive Spectrometry (EDS) Characteristics of $\text{Ca}(\text{OH})_2$

Fig. 7 shows the energy-dispersive spectrometry (EDS, HORIBA 7200-H) analysis results for $\text{Ca}(\text{OH})_2$. EDS analysis was performed to find the elements that were present in the samples. The results showed that the C-type absorbent consisted of such elements as Ca, O, and C, and that Ca and O had the highest proportions.

3. CFD Simulation Analysis Results of the SRx

Computational fluid dynamic (CFD) simulation analysis was performed to check the flue gas flow pattern inside the reactor. The GAMBIT program was used to perform modeling and meshing using the actual dimensions of a reactor. The $k-\varepsilon$ turbulence model was used for CFD simulation in Fluent. Table 5 shows the operating and boundary conditions.

The two ports for absorbent injection were handled with the velocity inflow boundary conditions. The boundary condition for the inlet of the reactor was set with the velocity, and that for the

Table 5. Composition table of the fly ash collected in the bag filter

Element	Weight %	Atomic %
C K	6.25	12.89
O K	25.55	39.58
Na K	3.82	4.12
Al K	0.46	0.42
Si K	1.38	1.22
S K	1.67	1.29
Cl K	34.79	24.32
K K	1.74	1.10
Ca K	24.34	15.05
Total	100.00	

outlet of the reactor was set with the pressure. The inflow velocity of flue gas was calculated with the flow amount, pressure, and temperature. The velocity condition of flue gas at the inlet was 11.97 m/sec, and the static pressure at the reactor outlet was $-1,000$ Pa. The sorbent injection rate at one port was 0.015 kg/sec, and the temperature at the reactor inlet was set at 180°C . Fig. 8(a) shows the SRx, (b) shows the flue gas streamlines inside the SRx, and (c) shows the sorbent particle residence time inside the reactor. The CFD simulation analysis results of the SRx showed that the flue gas and the sorbent particles were mixed well due to the formation of the flue gas turbulent flow in the reactor, and that the maximum residence time of the highly reactive absorbent in the reactor was 9.24 sec. The CFD analysis results were useful for studying the sorbent particle behavior, and they are deemed to be useful for the

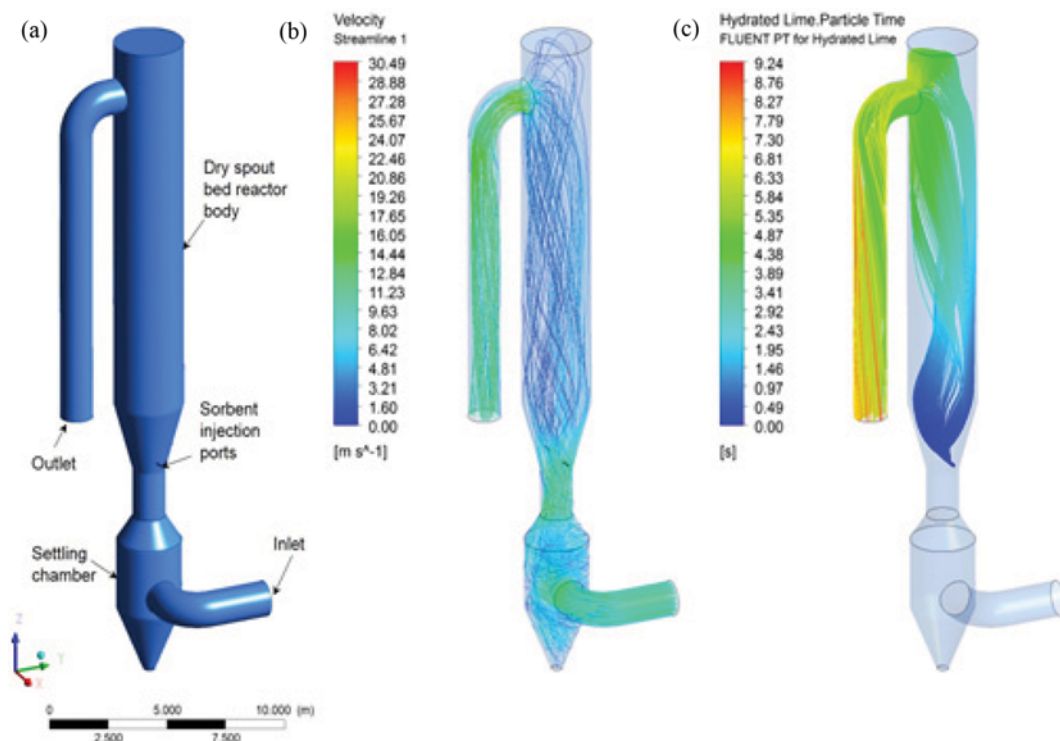


Fig. 8. Analysis of CFD for dry sorbent reaction accelerator: (a) Arrangement of inlet and outlet of flue gas, sorbent injection ports, (b) velocity streamlines of flue gas, (c) particles residence time of sorbents in the dry sorbent reaction accelerator.

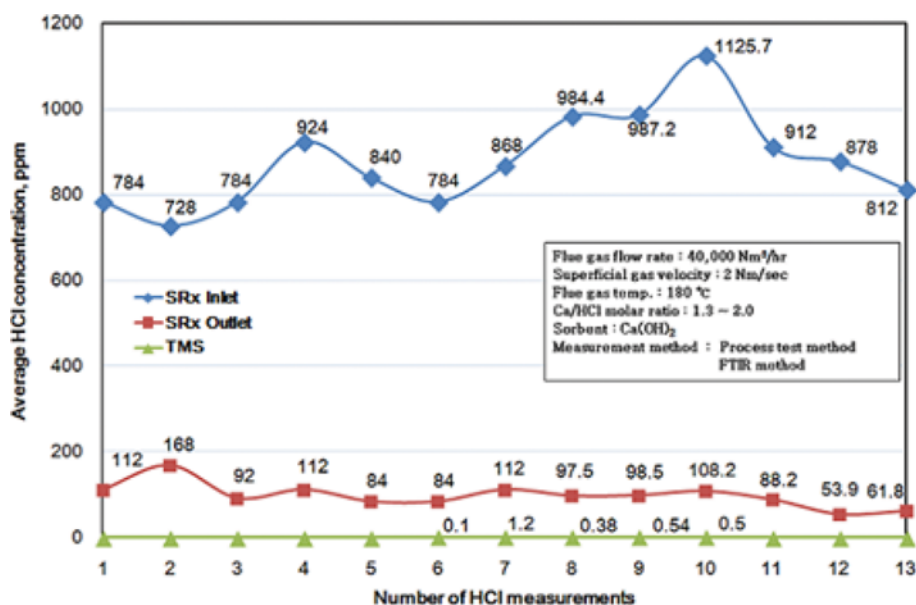


Fig. 9. Variation trends of average HCl concentration at the measuring points according to number of HCl measurements.

future study subject of absorbent dispersion optimization in a reactor according to the flue gas flow and the nozzle location.

4. HCl Removal Performance Experiment Results of the SRx

Fig. 9 shows the variation trends of average HCl concentration at the measuring points according to number of HCl measurements. The 1st-7th and 11th-13th measurements were conducted using the mercuric thiocyanate method, a standard method, and the 8th-10th measurements were conducted using the GASMET Dx-4000 analyzer using FTIR measurement method. The HCl concentrations at each position were compared while the SR was changed from 1.3 to 2.0 under the condition that the rising speed of the inflow flue gas was 2.0 Nm/sec. To get the certain overall removal

efficiency of HCl, repeated measurements of the concentration of HCl at the inlet and outlet of SRx many times.

The average HCl concentration at the front end of the reactor was maintained at 878 ppm, and that at the rear end was maintained at 98 ppm. The HCl concentration at TMS was 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 ($\text{Ca}(\text{OH})_2$) was increased. The increase in the gas-solid contact time is deemed to have contributed to the increase in the HCl removal efficiency.

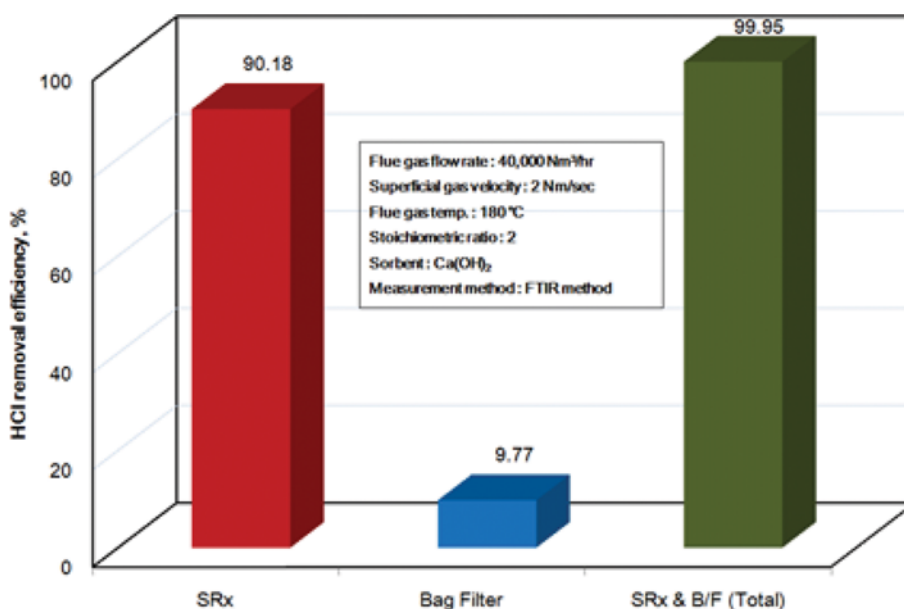


Fig. 10. HCl removal efficiency at dry sorbent reaction accelerator (SRx) and baghouse, total HCl removal efficiency.

Fig. 10 shows the HCl removal efficiency at dry sorbent reaction accelerator (SRx) and baghouse, total HCl removal efficiency. The HCl removal efficiency of the SRx under the flue gas flow amount of $40,000 \text{ Nm}^3/\text{hr}$ and the SR of 1.7 was 90.18%, and that of the baghouse under the pressure loss of $150 \text{ mmH}_2\text{O}$ was 9.77%. The

total HCl removal efficiency at the baghouse rear end (TMS data) under the baghouse pressure loss of $150 \text{ mmH}_2\text{O}$ was about 99.95%. This was the pressure drop of the baghouse filter increased because of the thickness of the deposited dust cake on the filter bag surface increased, and the secondary reaction with the HCl and unre-

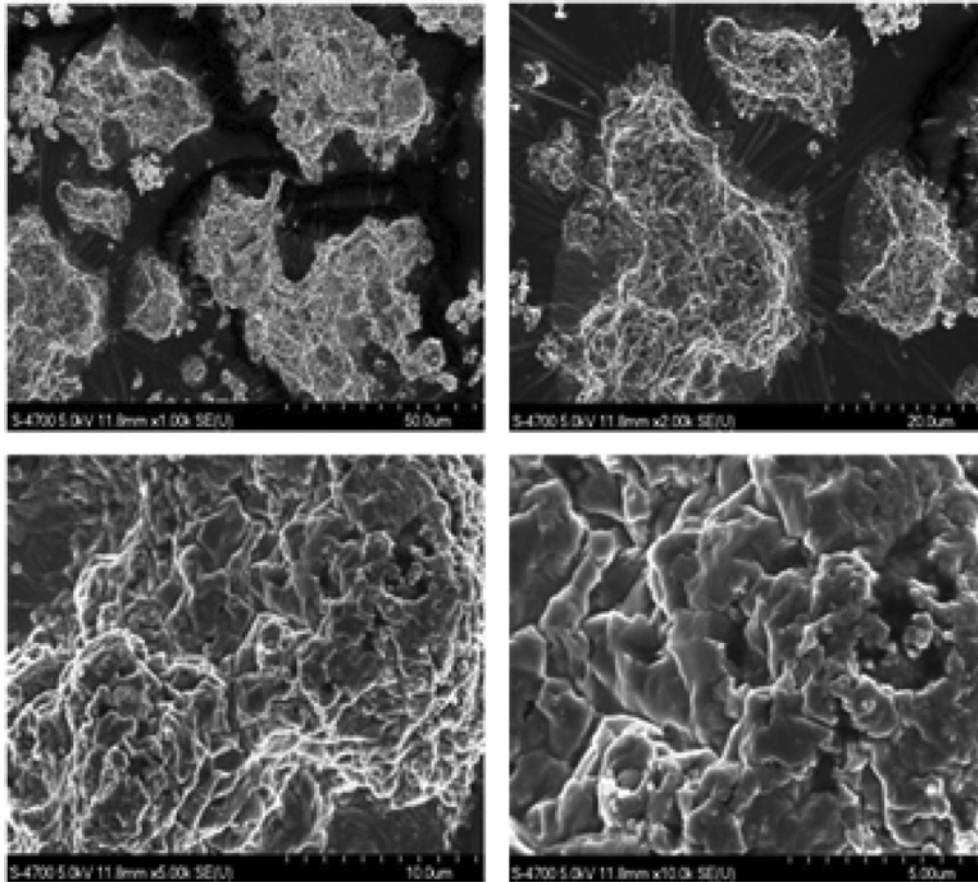


Fig. 11. SEM images of the collected particles at the bottom of baghouse: (a) $\times 1,000$, (b) $\times 2,000$, (c) $\times 5,000$, (d) $\times 10,000$.

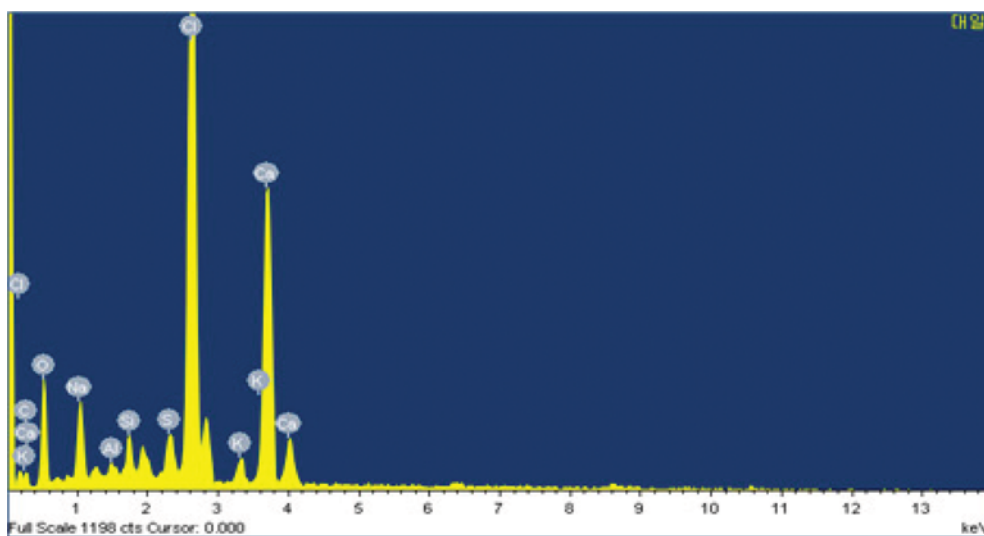


Fig. 12. EDS analysis of the collected particles at the bottom of baghouse.

acted calcium hydroxides had occurred.

The crystal structure of the surface of collected particles at bottom of the baghouse was observed using a scanning electron microscope (SEM). The collected particles contained the fly ash of waste incineration and unreacted calcium hydroxides. Micro images of the samples were obtained from the secondary electrons that occurred when the accelerated electrons were radiated to the sample surfaces. At the same time, the surface components of the fly ash were analyzed by measuring the content of the elements of the samples from the X-ray wavelength using the energy-dispersive spectrometer (EDS) that was attached to the SEM. Fig. 11 shows the SEM images of the particles that were collected in the baghouse in the $\times 1,000$, $2,000$, $5,000$, and $10,000$ magnifications. The collected particles generally had very irregular shapes, and the surface contained numerous porous structures. Fig. 12 and Table 5 show the EDS analysis results of the collected particles. The collected particles were mainly composed of C, O, Na, Al, Si, S, Cl, K, and Ca, and C, O, Cl, and Ca had the highest proportions. Most of the Ca originated from the unreacted calcium hydroxides.

Fig. 13 shows the qualitative and quantitative analyses results of the collected particles at the baghouse based on the EDS analysis. An X-ray diffraction analyzer (XRD, X'Pert APD, Philips) was used to measure the diffraction during the interaction between the X-ray and the substance. The analysis of the particles collected at the bottom of baghouse showed NaCl, KCaCl₃, and CaCl₂ as the major peaks. Based on the XRD analysis results, most of the Ca elements of the highly reactive Ca(OH)₂ combined with Cl and generated KCaCl₃ and CaCl₂, resulting in high HCl removal efficiency (99.9%).

CONCLUSION

Research on the dry-type reactor was conducted in this study as an alternative to the existing wet-type and semi-dry-type reactors, because these two latter reactors, which are currently being used as air pollution prevention facilities in waste incinerators, require higher investment and involve higher operation costs due to their large-capacity facility scale compared to the dry-type reactor, and have a secondary environmental pollution problem with water treatment. As the HCl concentration in the front end instantly varied according to the kind and amount of waste that was processed, there was a limit to conducting measurements, with several variables fixed in this pilot-scale experiment. The SR and the secondary reaction characteristics of the unresponsive absorbent in the baghouse were compared using Ca(OH)₂, which is known to have excellent reaction with HCl, as the absorbent for the experiment. The following conclusions were reached.

The rise in the SR increased the HCl removal efficiency under the condition that the internal temperature of the sorbent reaction accelerator (SRx) was 180 °C and the residence time was 5 sec. The SR was changed from 1.5 to 2.0 during the measurement, and a high efficiency of over 99.9% occurred from 1.5 based on the TMS data. Currently, incinerators are operating under the SR of 1.0-1.5. The high pressure loss of the baghouse resulted in high efficiency due to the secondary HCl removal reaction of the unresponsive absorbent. The HCl removal efficiency was maintained at over 99.9%, and the HCl emission concentration was maintained at below 5 ppm under the conditions that the flow amount of the flue

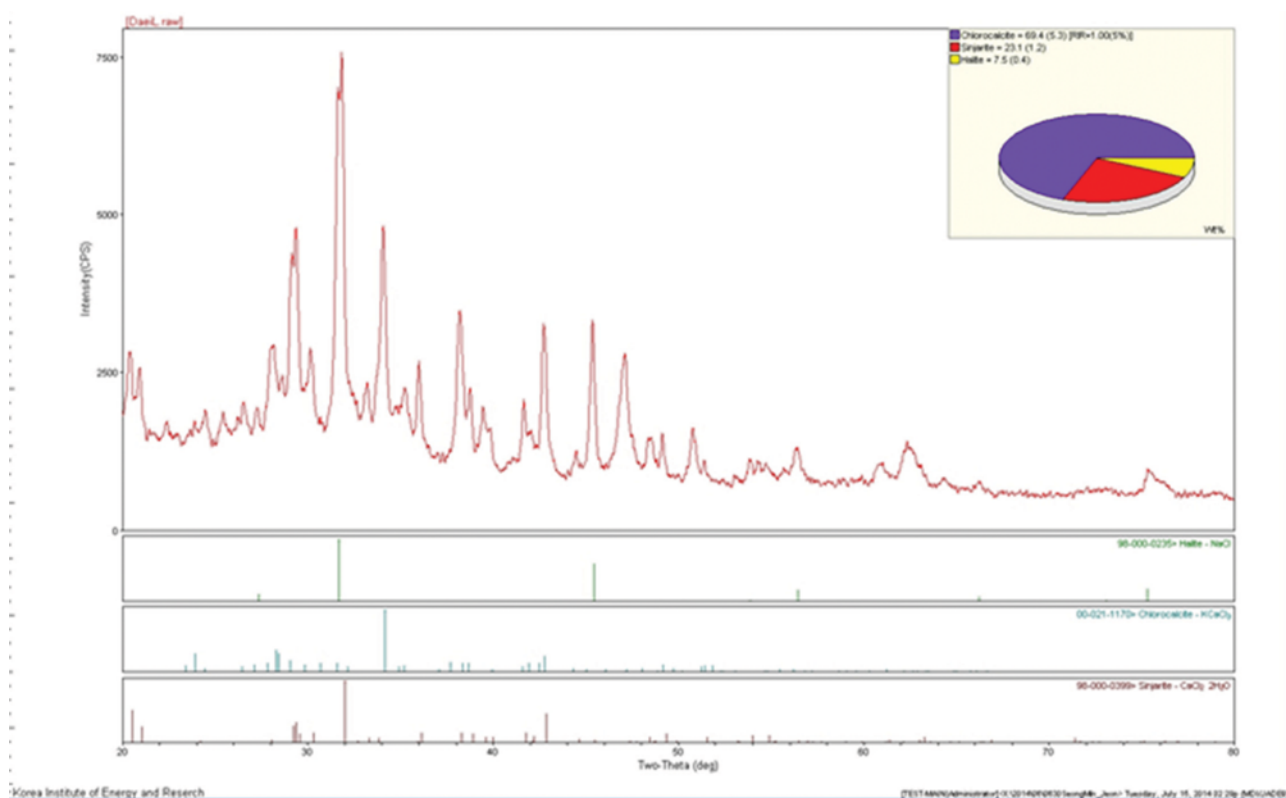


Fig. 13. Qualitative and quantitative analysis of the collected particles at the bottom of baghouse based on the EDS analysis.

gas was 40,000 Nm³/hr, the SR was below 2.0, and the pressure loss at the baghouse was 150 mmH₂O.

Therefore, the dry-type sorbent reaction accelerator (SRx) can reduce the facility and operation costs by replacing the wet-type and semi-dry-type technologies that are currently being used in most power plants, steel works, oil refineries, and incinerators. The SRx can maximize the removal efficiency of acid gas when linked with a baghouse, and can simplify the flue gas disposal process without any need for water treatment. The SRx can be an efficient and economical technology.

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