

Pilot Scale SO₂ Control by Dry Sodium Bicarbonate Injection and an Electrostatic Precipitator

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A 500 actual cubic feet gas per minute (acfm) pilot-scale SO₂ control study was undertaken to investigate flue gas desulfurization (FGD) by dry sodium sorbents in 400°F (204.5°C) flue gases emitted from a coal fired boiler with flue gas concentrations between 350 and 2500 ppm SO₂. Powdered sodium alkaline reagents were injected into the hot flue gas downstream of the air preheater and the spent reagents were collected using an electrostatic precipitator. Three different sorbents were used: processed sodium bicarbonate of two particle sizes; solution mined sodium bicarbonate, and processed sodium sesquicarbonate. SO₂ concentrations were measured upstream of the reagent injection, 25-ft (7.62 m) downstream of the injection point, and downstream of the electrostatic precipitator. SO₂ collection efficiencies ranged from 40 to 80% using sodium bicarbonate stoichiometric ratios from 0.5 to 3.0. Much of the in-duct SO₂ removal occurred during the first second of reagent reaction time, indicating that the sulfur dioxide–sodium reaction rates may be faster than have been measured for fixed bed measurements reported in the literature. © 2007 American Institute of Chemical Engineers Environ Prog, 26: 263–270, 2007

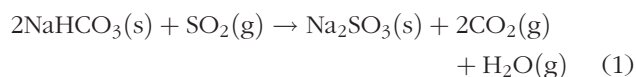
Keywords: flue gas desulfurization, coal-fired boiler, particle size measurement

INTRODUCTION

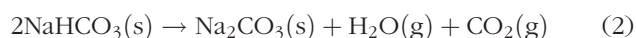
For some applications, dry sodium sorbent injection for flue gas desulfurization can be a cost effective

technology as reported by Muzio & Offen [1] and Offen *et al.* [2]. For retro-fit purposes, it would be beneficial if dry sodium injection systems could be used on boilers equipped with electrostatic precipitators (ESP) because many existing boilers use ESP for the control of particle emissions. It has been assumed that a fabric filter system is necessary to achieve high SO₂ removal efficiencies due to the enhanced contact between the boiler exhaust gases and reagent particles that occurs at the filter cake. A dry sodium powder injection system using an ESP particle collector depends mainly on the SO₂ reactions that occur while the sorbent particles are suspended in the flue gas.

Reactions between NaHCO₃ and SO₂ have previously been reported by Keener & Davis [3].



Furthermore, sodium bicarbonate is known to thermally decompose according to the following equation:



with the reaction reportedly beginning at temperatures near 100°F and occurring spontaneously at temperatures above 500°F. Sulfation of Na₂CO₃ occurs at a relatively slower and less effective rate:

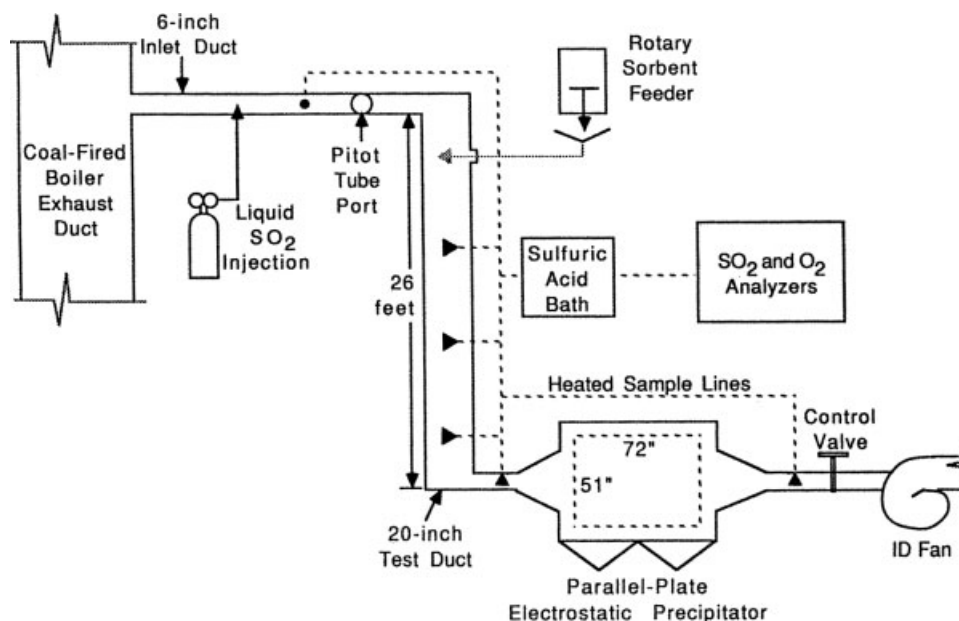


Figure 1. 500 acfm pilot-scale test facility.



Genco & Rosenberg [4] postulated that the sulfite product Na_2SO_3 in Eqs. 1 and 3 is further oxidized as follows:



Keener & Davis [3] demonstrated in a fixed bed reactor that reaction 1 was the dominant NaHCO_3 decomposition mechanism at temperatures below 450°F. As the temperature increased beyond 450°F, reaction 2 reportedly becomes the primary decomposition pathway. This results in a reduced SO_2 removal efficiency. In addition, Davis & Keener [5] reported that porosity changes occur in the particles during thermal decomposition of NaHCO_3 and suggested that particles thermally produced by reaction 2 have larger void spaces than that of the parent NaHCO_3 .

Previous pilot scale studies of dry sodium alkaline injection have focused on the use of baghouse filters as the primary gas-particle contactor as reported by Liu & Chaffee [6] and Veazy & Kielmeyer [7]. In some of these studies the SO_2 removal was measured downstream of the baghouse providing no information with respect to SO_2 removal from the alkaline particles suspended in the flue duct. Liu & Chaffee [6] reported in-duct SO_2 removal of 12–42% upstream of a filter baghouse. Muzio & Sonnichsen [8] reported 29% in-duct SO_2 removal by nahcolite upstream of a filter baghouse and Davis & Keener [5] measured 87% by sodium bicarbonate. Garding & Svedberg [9, 10] reported data on SO_2 removal in the ductwork upstream of a pilot-scale dry sodium injection FGD system that used a filter baghouse particle collector. They concluded that much of the SO_2 removal

occurred while the injected sodium bicarbonate particles were suspended in the gas stream. Ness & Selle [11] reported in-duct SO_2 collection efficiencies up to 61% using a pilot-scale dry sodium injection system equipped with an ESP located at the Grand Forks Energy Technology Center. These studies suggested that acceptable SO_2 removal might be achieved in a dry sodium injection system without a baghouse filter. A pilot-scale study was conducted at the University of Washington power plant to measure the effectiveness of retro-fitting a dry flue gas desulfurization process to an emission control system currently employing an ESP for particulate removal. Commercially available sodium-based alkaline materials with various surface areas were used as sorbents.

EXPERIMENTAL METHODS

Pilot Plant Test Facility

Coal Fired Boiler

A flow diagram of the pilot plant test facility is shown in Figure 1. The pilot plant was designed to treat a nominal 500 acfm sidestream from a Riley Stoker pulverized coal fired boiler. The boiler had a rated steam capacity of 100,000 lbs steam/h (45,359 kg/h). The coal had an average sulfur content of 0.57%. Stack gas compositions typically contained 8% CO_2 , 4% O_2 , and 7% H_2O vapor with a flyash loading of about 0.7 grain/sdcf (1.6 g/m³). SO_2 emission concentrations ranged from 350 to 500 ppm. Flue gas for the pilot plant was extracted downstream of the air preheater at an average temperature of 405°F (207.2°C).

Reactor Duct Section

Pulverized sodium sorbents were injected into the flue gas in a thermally insulated, 20-in. (50.8 cm) diameter, 26-ft (7.92 m) long vertical duct test section. Test ports were spaced along the reactor section to allow injection of sorbents at various distances upstream of the ESP to provide variable in-duct residence times. Also the effect of in-duct sorbent particle residence time was studied by injecting the sorbents at the top of the reactor section and extracting gas and sorbent samples from various points along the vertical reactor duct section. At the nominal 500 acfm (14.2 m³/min) system gas flowrate, the linear gas velocity inside the reactor section was 3.8 ft/s (1.16 m/s) and Reynolds number was 19,700. Maximum sorbent particle residence time upstream of the ESP was 7.1 s based on the measured flue gas volumetric flowrate. The gas flowrate was controlled by: (1) a damper valve at the test duct gas inlet and (2) a control valve downstream of the ESP and upstream of the induced draft fan. The gas flow was measured during each test run using a pitot tube traverse at test duct inlet.

Electrostatic Precipitator

The ESP was a thermally insulated, single lane, parallel plate type unit with 12-in. (30.5 cm) plate to plate spacing, 51-in. (125.9 cm) plate height, and 72-in. (182.9 cm) plate length, resulting in a nominal specific collection area of 100 ft²/1000 acfm [0.328 m²/(m³ min)]. At a 500 acfm (14.2 m³/min) flowrate, the gas velocity between the collection plates was 2.0 ft/s (0.61 m/s) with a gas residence time of 3.1 s (excluding the inlet plenum). The ESP was powered by a Universal Voltronics Corp. DC power supply rated up to 45 KV, for a maximum electric field strength of 2.9 KV/cm between the charging electrode and collection plates. The collection plates were manually rapped to remove the collected dust.

Sorbent Feed System

The sorbent particle feeder used rotating brushes to force the powdered sorbent particles through 1/8-in. (0.32 cm) mesh screen from the feed hopper to a discharge hopper. The vacuum of about 4 in. (10.2 cm) of H₂O inside the test duct assisted the flow of the sorbent from the discharge hopper into the vertical reactor duct. The controlled sorbent feed rate was in the 5–100 g/min (0.66–13.23 lb/h) range. The sorbent feed rate was measured by weighing the amount injected per time before and after each test run. At the nominal 500 acfm gas flowrate, the injected sorbent concentrations ranged from 0.25 to 5.0 grains/sdcf (0.57–11.44 g/m³). The sorbent feed air flowrate was about 5 acfm (0.14 m³/min) or 1% of the total flue gas flowrate.

Flue Gas Sampling Methods

Sampling Apparatus

The flue gas sampling system measured the SO₂ concentrations at three locations: one upstream and two downstream of the sorbent feeder. The flyash and active sorbent in the flue gas were separated

from the gas stream by an in-stack University of Washington Mark 5 cascade impactor which is described in an EPRI report by Pilat *et al.* [12]. The cascade impactors separated the partially reacted sorbent particles from the SO₂ laden sampled gas and further reduced the SO₂ removal from the flue gas (compared with the use of in-stack filters which would pass the sampled gas through the sorbent filter cake). The particle-free sampled gases were cooled and dried by passing through cold, concentrated reagent grade sulfuric acid in an ice bath. The in-stack impactors were cleaned and the sulfuric acid bath replaced after each run.

Tests were conducted to confirm that after 30 min of continuous sampling, neither the sorbents collected by the cascade impactors or the sulfuric acid cooling solution caused significant artifact SO₂ removal. After 30 min the collected sorbents in the impactor and condensed water in the concentrated sulfuric acid cooling solution resulted in less than 4 and 7% removal of the gas phase SO₂, respectively. Because the test run sampling was operated less than 10 min per test run, it appeared that the SO₂ removal by the sampling apparatus was less than 5%.

Continuous analyzers were used to measure SO₂ and O₂ content in the sampled gas. The SO₂ was measured using a Thermo Electron Corp. Model 40 pulsed fluorescent analyzer. The oxygen concentrations were measured using either a Taylor Instruments paramagnetic analyzer or a Teledyne fuel cell analyzer. To account for air leakage into the pilot plant, the SO₂ concentrations measured at each gas sampling point were adjusted to a reference O₂ concentration. Air leakage accounted for less than 2% of the total gas flowrate.

Sampling Procedure

The SO₂ concentrations at the two sampling locations downstream of the sorbent feeder were measured by cycling between the inlet gas sampling location and the two downstream locations. A test series was performed by measuring the SO₂ concentrations at a set of preselected sorbent feed rates. For each test run the system was stabilized for ~5 min while the inlet SO₂ and O₂ concentrations were measured. Then, the continuous monitors were switched to either of the two downstream sampling locations and the system was operated for an additional 10 min while the concentrations at both locations were recorded onto a strip chart recorder. The sorbent feed rates were measured by collecting a sorbent feed sample over a known time period. The normalized sorbent stoichiometric ratio (NSR, expressed as mole Na₂/mole SO₂) for each test run was then calculated using the measured sorbent feed rate, inlet SO₂ concentration, and flue gas flow rate.

$$NSR = \frac{F \text{ SO}_2 V_s}{Q C_{\text{SO}_2}} \quad (5)$$

where F is the sorbent feed rate (mole Na/min), SO_2 is the SO₂ molecular weight, Q is the flue gas flow-

Table 1. Sodium sorbent properties.

	Small processed	Large processed	Small solution mined	Small processed sesquicarbonate
Supplier	Church Dwight	Church Dwight	IRI	FMC Corp
BET surface area (m ² /g)	0.41	0.22	0.41	0.43
MicroTrac MMD (μm)	11.2	28.3	10.4	10.1
MicroTrac σ _g	1.6	1.9	2.7	1.6
Impactor MMD (μm)	44.3			
Impactor σ _g	3.3			
Microscope MMD (μm)	6	15	11	
Microscope σ _g	1.9	1.7	2.0	

rate (dry standard cubic meters/minute), V_s the standard gas molar volume (m³ gas/g-mole) and C_{SO_2} is the mole fraction of SO₂ in the inlet flue gas.

Sodium-Based Sorbents

Four different sodium-based sorbents were used in this study: small-particle processed sodium bicarbonate (NaHCO₃); large-particle processed sodium bicarbonate; small-particle solution mined sodium bicarbonate; and small-particle processed sodium sesquicarbonate (NaHCO₃ · Na₂CO₃ – 2H₂O). Physical properties of the four sorbents are shown in Table 1. Each sorbent was delivered by the supplier in pulverized form. The “processed” sorbents were produced by chemically modifying mined soda ash (Na₂CO₃), forming large crystals of NaHCO₃. These large crystals were then pulverized to the desired particle size. The “solution mined” NaHCO₃ was produced by using a liquid solution to mine natural nahcolite ore (70% sodium bicarbonate, remainder inerts). Sodium bicarbonate, dissolved in the mining solution, was recrystallized into particles of desired size. The solution mined NaHCO₃ particles consisted of short rods with length/diameter ratios between 2/1 and 3/1 and the “processed” NaHCO₃ particles were more uniformly cubic in shape.

The particle size distributions of the sorbents were measured using three techniques. First, samples of each of the four sorbents were analyzed by the Church and Dwight Company, using a Leeds/Northrup Microtrac size analyzer which counted the size distribution of particles suspended in a liquid solvent. Second, in-stack particle size distributions of the small Church and Dwight sodium bicarbonate were measured using a University of Washington Mark 5 cascade impactor. Third, samples of three of the injected sorbents were collected from inside the duct on greased microscope slides and inspected under an optical microscope to observe the particles and manually count and measure the particle size distribution.

As shown in Table 1 the “small-particle” sorbents (processed and solution mined bicarbonate and processed sesquicarbonate) were of comparable surface area. The MicroTrac size analyzer measured particle mass median diameters (MMD) ranged from 10.1 to 11.2 μm with a geometric standard deviations σ_g

from 1.6 to 2.7. The “large-particle sorbent” (processed sodium bicarbonate) had an MMD of 28.3 μm and a geometric standard deviation of 1.9.

The small-particle “processed sodium bicarbonate” sorbent MMD was measured at 44.3 μm with the in-stack cascade impactor and this MMD was considerably larger than the particle diameters measured with either the MicroTrac analyzer or with optical microscope. This MMD difference was probably due to the agglomeration of the smaller particles into clumps (the impactor measured the aerodynamic in-stack diameters of the particles). The cascade impactor may have measured some of these clumps as single particles, increasing the MMD and geometric standard deviation. The Microtrac analyzer, however, counted each sorbent particle (suspended in a liquid) separately. It is uncertain if, or how, the presence of sorbent clumps affected the SO₂ removal. The particle clumps appeared loosely agglomerated, suggesting that gas phase mass transfer to individual particles may not have been significantly hindered.

EXPERIMENTAL RESULTS AND DISCUSSION

A comparison between the SO₂ removal efficiency of processed sodium bicarbonate and sodium sesquicarbonate in Figure 2 shows that the small processed NaHCO₃ (MMD = 11.2 μm) had the highest collection efficiency. Percent SO₂ removal by each sorbent is shown as a function of normalized stoichiometric ratio (NSR), with units of mole Na₂ per mole inlet SO₂. The inlet gas temperature was 400–410°F (204.5–210°C) and particle residence time upstream of the ESP was ~2.5 s. The curves through the measured data points in Figures 2–5 are best fits to the equation

$$SO_2 \text{ Removal (\%)} = m \log (\text{Stoichiometric Ratio}) + b \quad (6)$$

To provide 50% removal, the NaHCO₃ required a sorbent NSR of 1.0 gmole Na₂/gmole SO₂, while sodium sesquicarbonate required nearly 2.5 times that stoichiometric ratio. These results agree with those of previous researchers Muzio & Sonnichsen [8] and Carson [13] who reported NaHCO₃ achieved better SO₂ removal efficiencies than either sodium carbonate or

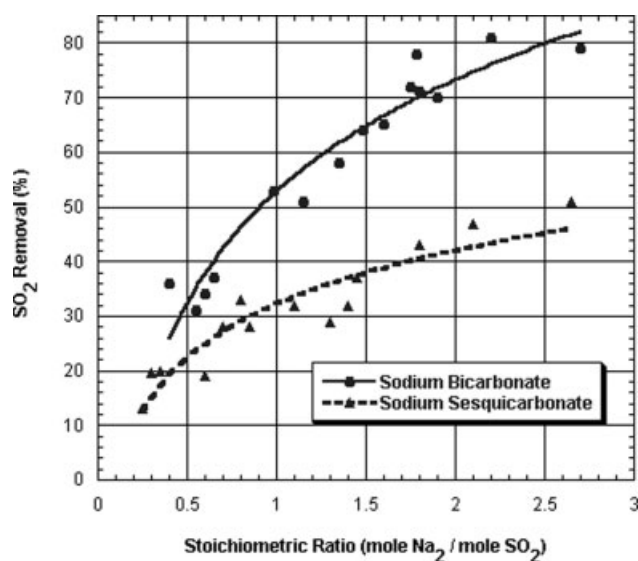


Figure 2. Comparison of SO₂ removal rates between 11- μ m diameter sodium bicarbonate and 10- μ m sodium sesquicarbonate at flue gas temperatures 400–410°F; SO₂ concentrations 450–500 ppm; and 2.5 s in-duct residence time.

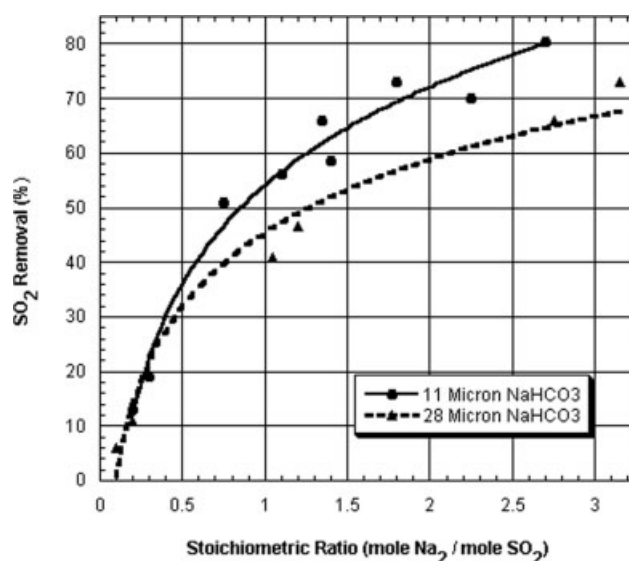


Figure 4. Comparison of removal rates between 11 and 28 μ m sodium bicarbonate at flue gas temperatures 400–410°F; inlet SO₂ concentrations 310–330 ppm; and 5 s in-duct residence time.

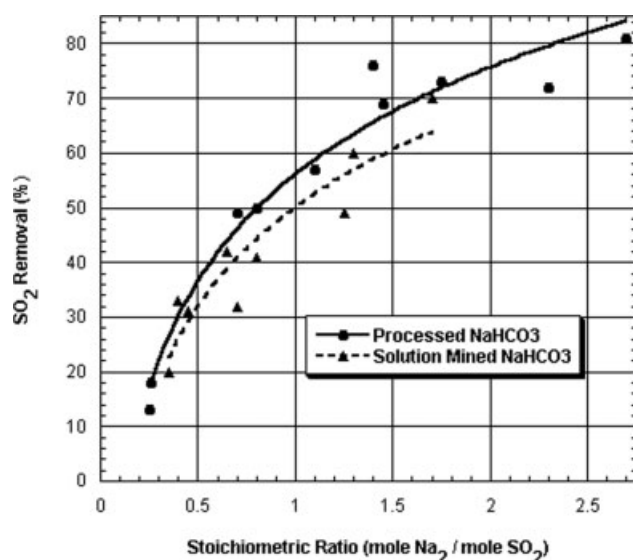


Figure 3. Comparison of SO₂ removal rates between 11 μ m “processed” sodium bicarbonate and 10 μ m “solution mined” sodium bicarbonate at flue gas temperatures 400–410°F; inlet SO₂ concentrations 320–350 ppm; and 5 s in-duct residence time.

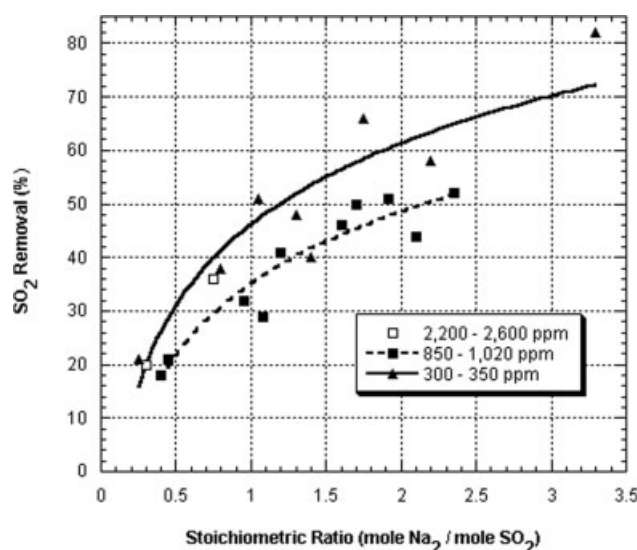


Figure 5. Effect of inlet SO₂ concentration on removal efficiency at flue gas temperatures 400–410°F and 2.5 s in-duct residence time.

sodium sesquicarbonate for dry sodium sorbent injection in pilot-scale and full-scale filter baghouse systems. Furthermore, Keener & Davis [3] and Stern [14] reported similar results using bench scale thin-bed reactors. This trend was also observed by Shah & Teixeira [15] who conducted similar studies on the removal of SO₂ and NO_x. Sodium bicarbonate, nahcolite, and trona, a sodium sesquicarbonate containing ore, were used as the dry absorbent in a 600 scfm (17 m³/min) baghouse removal system.

Figure 3 shows that the processed NaHCO₃ and solution-mined NaHCO₃ achieved similar SO₂ removal efficiencies when tested using a 5.0–5.5 s in-duct sorbent aerosol particle residence time. The two sodium bicarbonate types have similar particle sizes and surface areas, as shown in Table 1, and appear to differ only in their particle geometry's. With both bicarbonate types having similar SO₂ removal efficiencies it appears that the particle shape (rod shaped versus cubic) had little effect.

Sorbent Utilization

As is shown in Figures 2 and 3, the sodium sorbent utilization for all the sorbents tested was less than 100%. Sorbent utilization, which is defined as the ratio of fractional SO₂ removal to NSR, can be used to describe a removal system's approach to maximum performance.

$$\text{Sorbent Utilization} = \frac{\text{Fraction SO}_2 \text{ Removal}}{\text{Normalized Stoichiometric Ratio}} \quad (7)$$

To obtain 50% SO₂ removal in 450–500 ppm flue gas, the 11- μ m sodium bicarbonate had to be injected at an NSR of 1.0, resulting in a sorbent utilization of 0.5 as shown in Figure 2. This indicates that 50% of the available sodium bicarbonate reacted with SO₂. For a similar 50% SO₂ removal, the 10- μ m sodium sesquicarbonate required an NSR of 2.5, resulting in a sorbent utilization of 0.20. Our measured relationship of the SO₂ removal efficiency to NSR is in general agreement with the model predictions reported by Wu *et al.* [16].

Reinjection of Collected Spent Sorbents

It was apparent that 100% sorbent utilization was not approached by either sorbent regardless of the NSR increase. Therefore, in an attempt to increase sorbent utilization, the spent sodium bicarbonate sorbent, collected by the ESP, was recycled. Initially the removal efficiency was ~50% with the system operating at 350-ppm SO₂ inlet concentration and 2.5-s residence time upstream of the ESP. No attempt was made to separate the spent sorbent from the flyash. At an injection rate corresponding to an unreacted sorbent NSR of ~0.6, the measured SO₂ removal was 3.8%. At a higher injection rate, corresponding to an unreacted sorbent NSR of 1.5 and a sorbent concentration of 5 grain/dscf (11.4 gm/m³), the SO₂ removal efficiency increased to 8.0%. Based on these low SO₂ removal efficiencies, it appeared that the reinjection of dry collected spent sorbent is not a feasible approach for improving sorbent utilization. Our data suggested that a significant portion of the recycled sorbent was unreacted at the time of reinjection. It has been reported by Howatson *et al.* [17] and Borgwart *et al.* [18] that a passivation layer, or “ashlayer” of reaction products form on the outer surface of the sorbent particle. This ashlayer impedes diffusion of SO₂ to the unreacted sorbent interface, thereby reducing the SO₂-removal capability of the reinjected sorbents. This also has been reported by Klingspor [19] who examined the effect of grinding the sorbent material prior to recycle. It was reported that the SO₂ sorption removal capacity of unaltered recycled sorbent was minimal. However, grinding the sorption material prior to recycle resulted in the sorption efficiency approaching initial values. Jiang *et al.* [20] used a bench scale circulating fluidized bed combustor and they reported an increased SO₂-removal efficiency with increased NaHCO₃ sorbent recycle ratio. Fresh sorbent, fed at an NSR of 1.0, was combined

with spent sorbent in ratios up to 240. SO₂ removal at this recycle ratio was greater than 90%, without any subsequent grinding of the recycled sorbent. This enhancement could be due to greater residence times and the lower temperature of 300°F (148.9°C) slowing the NaHCO₃ thermal decomposition reaction shown in Eq. 2.

Influence of Particle Size

Figure 4 shows that the smaller 11 μ m MMD NaHCO₃ particles provided a higher SO₂ removal efficiency than the larger 28 μ m MMD NaHCO₃ particles. Both sorbent sizes were tested at in-duct residence times of 5.0–5.5 s upstream of the ESP. To obtain a 50% SO₂ removal, the required NSR for the 28- μ m diameter particles was ~1.6 times that required for the smaller 11- μ m diameter particles. This general trend of increased SO₂ removal efficiency with smaller particles agrees with the higher BET particle surface areas for the smaller particles, reported in Table 1. Similar results have been previously reported for measurements at filter baghouses and fixed thin-bed reactors and the modeling results reported by Wu *et al.* (2004).

Effect of Inlet SO₂ Concentration

As is shown in Figure 5, the inlet SO₂ concentration had little effect on the SO₂ removal efficiency of the small 11 μ m MMD NaHCO₃ particles. Five test series were run using 11 μ m MMD NaHCO₃ particles and flue gas spiked with pure SO₂ up to concentrations of 2600 ppm. The flue gas temperature was 400–410°F (204.5–210.0°C). The apparent insensitivity to the inlet SO₂ concentration suggests that the same reaction rate limiting steps were dominant throughout this 300–2600 ppm concentration range and that no gas phase diffusion limitations were present. Our results agree with the pilot-scale filter baghouse SO₂ removal by injection of pulverized nahcolite ore reported by Davis *et al.* [21] where it was concluded that the inlet SO₂ concentration had no effect on its removal efficiency. However, a study examining the removal efficiency of a bench-scale thin-bed of nahcolite performed by Stern [14] suggested a dependence on SO₂ concentration. Stern's bench scale studies concluded that SO₂ removal efficiencies increased with increasing inlet SO₂ concentrations. Fellows & Pilat [22] reported that gaseous HCl concentrations in the 400–700 ppm had little effect on the HCl removal efficiencies at 455°F (235°C) measured with a lab-scale sodium bicarbonate fixed-bed reactor. Wu *et al.* [16] reported theoretical model predictions of higher SO₂ removal efficiencies with higher inlet SO₂ in the 200–3000 ppm SO₂ concentration range at 260°F (127°C).

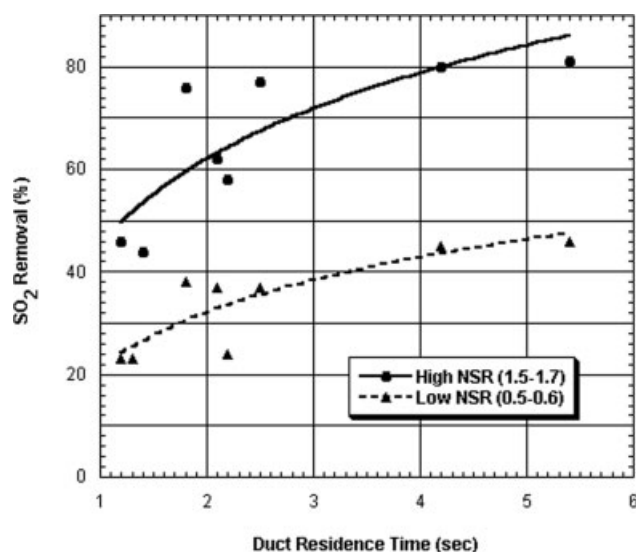
Effect of ESP Operation on SO₂ Removal

As is shown in Table 2, decreasing the ESP corona voltage caused a slight increase in the SO₂-removal efficiency. Under normal ESP operating conditions, the corona voltage was at 2.6 KV/cm. Decreasing the corona voltage probably decreased the ESP particle

Table 2. Effect of ESP corona voltage on SO₂ removal.

Duct residence time (s)	Stoichiometric ratio	Corona voltage (KV/cm)	SO ₂ removal ESP inlet	SO ₂ removal ESP outlet
2.5	1.0	2.6	25	40
2.5	1.0	1.3		45
2.5	1.0	0		45
5	1.5	2.6	75	80
5	1.0	2.6		52
5	1.0	1.3		60
5	1.0	0		63

11- μ m diameter NaHCO₃ at 400°F, 350 ppm SO₂.

**Figure 6.** Effect of in-duct residence time on SO₂ removal efficiency at 400–410°F flue gas temperature and 350–380 inlet SO₂ concentration.

collection efficiency, thereby providing a longer gas-particle contact time inside the ESP.

Table 2 shows that the SO₂ removal across the ESP for the two upstream residence times of 1.0 and 2.5 s were 25 and 40%, respectively at an NaHCO₃ stoichiometric ratio of 1.0. Using a higher stoichiometric ratio of about 1.5 and an upstream residence time of 5 s, the upstream and downstream SO₂-removal efficiencies were 75 and 80%, respectively. The relative increase in SO₂ removal across the ESP appeared to depend on the extent of SO₂ removal (SO₂ reaction with the sorbent) upstream of the ESP. As is shown in Figure 6, if the sorbent residence time upstream of the ESP is short (less than 1.5 s), then the SO₂ sorption can continue during the time the sorbent particles are suspended inside the ESP. With longer sorption particle residence times upstream of the ESP (greater than 5 s), most of the SO₂ sorption had already occurred and little additional SO₂ was collected inside the ESP.

Effect of In-Duct Particle Residence Time

The reaction rate of SO₂ with the 11- μ m MMD NaHCO₃ sorbent particles was obtained by measuring the SO₂ concentration at several locations downstream of the sorbent particles injection point corresponding to 1.4- to 5.3-s residence times. The SO₂ removal efficiencies as a function of duct residence time are shown in Figure 6 for stoichiometric ratios of low (0.5–0.60) and high (1.5–1.7) magnitudes. The curve through the data points are logarithmic. The sorbent particles appeared to react quickly at these residence times and much of the SO₂ removal occurred within the first 2 s of particle injection. For a NSR of about 1.5, the SO₂ removal after 1.5 s was greater than 55%. After 2.5 s, the removal was greater than 65%. The rate of removal leveled-off at higher residence times, i.e. 75% at 5.3 s. Our 400°F (204.5°C) measurements showed significant SO₂-removal efficiencies (35–70%) within the 3-s duct residence time. Our in-duct measured SO₂ removal efficiencies do not agree with the model predictions reported by Wu *et al.* [16]. They reported small SO₂ concentration changes (less than 5%) for a 1-s sorbent particle duct residence time at a gas velocity of 15 m/s (49.2 ft/s), particle diameter of 5–10 μ m, and temperatures in the 120–210°C range (248–410°F).

NaHCO₃–SO₂ Reaction Rate

The measured in-duct SO₂ removal rates shown in Figure 6 indicate that the initial NaHCO₃–SO₂ reaction rates between the aerosol particles and the SO₂ appeared to be faster than have been previously reported.

CONCLUSIONS

The results of our pilot plant study have provided the following conclusions:

1. With 300–400°F flue gas containing 350–500 ppm SO₂ significant SO₂ removal in the 40–80% range was achieved by injecting sodium bicarbonate particles directly into the gas stream upstream of an ESP.
2. Sodium bicarbonate provided better SO₂ collection than did sodium sesquicarbonate. At a stoichiometric ratio of 1.0 with sodium bicarbonate the

SO₂ collection efficiency was 50% compared with only 25–30% for sodium sesquicarbonate.

3. The SO₂ collection efficiency was greater for the smaller 11- μ m diameter particles than for the larger 28- μ m diameter NaHCO₃ particles.
4. Most of the SO₂ transformation to sulfite occurred in the test duct before the particles were removed by the ESP.

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LITERATURE CITED

1. Muzio, L.J., & Offen, G.R. (1987). Assessment of dry sorbent emission control technologies: Part I. Fundamental processes, *Journal of the Air Pollution Control Association*, 37, 642–654.
2. Offen, G.R., McElroy, M.W., & Muzio, L.J. (1987). Assessment of dry sorbent emission control technologies: Part II. Applications, *Journal of the Air Pollution Control Association*, 37, 968–980.
3. Keener, T.C., & Davis, W.T. (1984). Study of the reaction of SO₂ with NaHCO₃ and Na₂CO₃, *Journal of the Air Pollution Control Association*, 34, 651–654.
4. Genco, J.M., & Rosenberg, H.S. (1976). Sorption of SO₂ on ground nahcolite ore, *Journal of the Air Pollution Control Association*, 26, 989–992.
5. Davis, W.T., & Keener, T.C. (1980). Chemical kinetic studies on dry sorbents—Literature review phase 1, DOE/FC/10184-2, August.
6. Liu, H., & Chaffee, R. (1969). Evaluation of fabric filter as chemical contactor for control of SO₂ from flue gas, NTIS PB-194–196.
7. Veazy, F.M., & Kiemeier, W.H. (1970). Feasibility of fabric filter as gas-solid contactor to control gaseous pollutants, NTIS PD-195-884.
8. Muzio, L.J., & Sonnichsen, T.W. (1984). Dry SO₂ particulate removal for coal fired boiler, EPRI Report No. EPRI CS-2894, Volume 2.
9. Garding, M., & Svedberg, G. (1985). Flue gas desulfurization by dry injection: Pilot scale tests, summary of final report 1985-0807, The Swedish Royal Institute of Technology.
10. Garding, M., & Svedberg, G. (1988). Modeling of dry injection flue gas desulfurization, *Journal of the Air Pollution Control Association*, 38, 1275–1280.
11. Ness, H.M., & Selle, S.J. (1978). Control of western power plant sulfur dioxide emissions: Development of ash-alkali FGO process and dry adsorption techniques at the grand forks energy technology center, USDOE environmental control symposium, NTIS DOE/EV-0046, 1978.
12. Pilat, M.J., Raemhild, G.A., Powell, E.B., Fioretti, G.M., & Meyer, D.F. (1978). Development of a cascade impactor system for sampling 0.02 to 20 μ m diameter particles, EPRI Report No. EPRI FP-844, Volume 1.
13. Carson, J.R. (1980). Removal of sulfur dioxide and nitric oxide from a flue gas stream by two sodium alkalis of various sizes, M.S. Thesis, University of Tennessee.
14. Stern, F.R. (1978). Bench scale study of sulfur and nitrogen oxide adsorption by nahcolite and trona, M.S. Thesis, University North Dakota.
15. Shah, N.D., & Teixeira, D.P. (1978). Bench-scale evaluation of dry alkalis for removing SO₂ from boiler flue gases, presented at the symposium on the transfer and utilization of particulate control technology, Denver, CO.
16. Wu, C., Khang, S., Keener, T.C., & Lee, S. (2004). A model for dry sodium bicarbonate duct injection flue gas desulfurization, *Advances in Environmental Research*, 8, 655–666.
17. Howatson, J., Dewald, H., Outka, D., Diller, D., Cain, M., & Smith, J. (1980). Reaction of nahcolite with sulphur dioxide, *Journal of the Air Pollution Control Association*, 30, 1229–1230.
18. Borgwardt, R., & Harvey, R. (1972). Properties of carbonate rock related to SO₂ reactivity, *Environmental Science & Technology*, 6, 350.
19. Klingspor, J.S. (1987). Improved spray dry scrubbing through grinding of FGD recycle material, *Journal of the Air Pollution Control Association*, 37, 801–806.
20. Jiang, X.L., Keener, T.C., Khang, S.J., & Hao, J.M. (1990). Experimental study and modeling of SO₂ removal in a circulating fluidized bed absorber by NaHCO₃ sorbent, presented at the 83rd annual meeting and exhibition, Pittsburgh, PA, June 26–29.
21. Davis, W.J., Keener, T.C., & Lavelly, L.L. (1978). Research on the removal of SO₂ by additive injection techniques on a stoker-fired boiler, Paper No. 78-28-4, 71 of APCA annual meeting.
22. Fellows, K.T., & Pilat, M.J. (1990). HCl sorption by dry NaHCO₃ for incinerator emissions control, *Journal of the Air & Waste Management Association*, 40, 887–893.