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**Quarterly Report
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***Project Title:* Low Temperature SO₂ Removal with Solid Sorbents in
a Circulating Fluidized Bed Absorber**

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Abstract

Water injection in dry lime injection FGD processes plays a very important role for the SO₂ removal efficiency. The experimental data obtained from circulating fluidized bed absorber (CFBA) operations under water injection through the newly designed toroidal ring nozzle were represented. The results suggest the removal efficiency is a strong function of the water injection rate. In addition, those indicate that the height of water spray is more significant in wetting efficiency during fluidization in CFBA instead of the number of holes. The height should be adjusted depending on the reaction zone in order to improve the wetting efficiency of lime. The effect of gas velocity appeared not significant, resulting from that the wetting efficiency may increase at higher velocities. The loading of sorbents strongly affects on the SO₂ removal as the lime becomes hydrated and attrited, while its effect appears not significant before hydration occurs. As the lime continuously contacts with water droplets, it becomes hydrated and its attrition is significantly taking place. Since the hydration causes the high reactivity and attrition provides the fresh surface of lime, an effective wetting to allow the gradual attrition and hydration can increase the SO₂ removal efficiency and sorbent utilization. Such effective wetting may be obtained by means of spraying with smaller droplets.

I. Work Performed/Results Obtained

Experimental Procedure. The SO₂ removal tests under water injection were carried out at a batch mode in the circulating fluidized bed absorber(CFBA). Two discrete ranges of Dravo limes were used as solid sorbents for the SO₂ removal tests. The sizes of Lime 1 range between 1095 μm (16 meshes) and 2380 μm (8 meshes) in diameter and those of Lime 2 range between 595 μm (30 meshes) and 1095 μm (16 meshes). The lime samples supplied by Dravo lime company were as big as about 1.3 cm in diameter. The sorbents, therefore, were ground to two mass mean diameters (MMD) of 903 (Lime 1) and 1764 μm (Lime 2) with a Bico pulverizer (BICO co.). The fractional size distribution for the lime samples was given in the previous report. In addition, the lime sample is a high calcium quicklime formed by calcining limestone so that CO₂ is liberated. Its available lime was measured as about 90 %, and a slaking test confirmed that the lime is very reactive. The physical and chemical properties of lime samples were measured or obtained from Dravo company, and the results were also reported previously.

For the enhancement of SO₂ removals in a CFBA, a toroidal type of pressurized spray nozzle was newly designed and installed at the bottom of bed. Its design and installation was reported in the previous document. For this study, it has 2, 3, and 4 holes and 85° angles, based on the droplet evaporation rate and its trajectories in a fluidized bed. The diameter of the holes punctured with a very tiny drill is 175 μm. Water is injected by means of the high pressure pump to insure good atomization, and a filter is also placed before the pump to prevent the contaminants from clogging the small holes.

For a sulfation test, 0.5 kg - 0.75 kg of limes were injected so that the initial pressure drop in a bed reactor reaches about 15 - 20 cm H₂O. Then water was sprayed into the bed, and SO₂ concentrations at the inlet and outlet were continuously measured with a Horiba SO₂ analyzer. The fluidizing gas temperature at the inlet was about 177 °C and superficial gas velocity was 4 m/s for Lime 1 (903 μm) and 4 - 5 m/s for Lime 2 (1764 μm), respectively. The temperature and pressure drop were also recorded into a data acquisition system. After the CFBA unit was turned off, the bed materials and fines captured by the first and second cyclones were collected, and their total sulfur content and hydration were measured with a LECO sulfur analyzer and a thermogravimetric analyzer (951 TGA, Du Pont Co.), respectively.

Results and Discussion . A series of experiments was conducted in order to determine the SO₂ removal efficiency in CFBA under different wetting conditions. Figure 1 shows the

experimental results of the SO_2 removal efficiency versus the water injection rate during fluidization of Lime 1 (MMD = $903\ \mu\text{m}$). For the test, 1 kg of lime was fluidized at the superficial gas velocity of 4 m/s and fines captured by the first cyclone were recirculated into a bed. Water was initially injected at 30 ml/min of flowrate through the 2-hole nozzle, and the water injection rate was gradually increased after the SO_2 concentration at the outlet reached at steady state. The results indicate that the SO_2 removal efficiency is a strong function of the water injection rate. While the reaction of lime with SO_2 did not occur under the dry condition before injecting water, the SO_2 removal efficiency dramatically increased with water injection. As shown in Figure 1, the removal efficiency was enhanced from 10 % to 35 % as the water injection rate increased from 30 ml/min to 70 ml/min.

The effect of the number of the nozzle hole on the SO_2 removal was examined in a stationary bed without circulation of the fine solids elutriated from bed and captured by the first cyclone. The results are illustrated in Figure 2. Lime 2 (MMD = $1764\ \mu\text{m}$) of 0.75 kg in weight was injected at the fluidizing gas velocity of 5 m/s, and the water injection rate through the 2-hole and 4-hole nozzles was 60 ml/min. Figure 2 suggests that the more number of holes may not be significant in the wetting efficiency since it reduces the height of water spray due to the decreased water pressure and thus limits wetting the solids in the upper region of the bed reactor. Under no circulation of solids, the reaction may mainly take place in a dense region because the parent solids only stays in a bed reactor and the attrited fines immediately blow out at high velocity. As a result, when water is sprayed far away enough to reach the dense region, more holes of the nozzle may result in better wetting efficiency and thus higher removal efficiency.

Figure 3 shows the effect of the hole number on the SO_2 removal efficiency under circulation of the elutriated solids. Tests were carried out with Lime 2 and nozzles with two and three holes, at the gas velocity of 5 m/s and the water injection rate of 60 ml/min. Circulation of fine solids started at 20 minutes after water was injected. As shown in Figure 3, the nozzle with three holes has higher SO_2 removal efficiency than the two-hole nozzle as similar as in Figure 2 before the circulation of fine solids, but it has the lower removal efficiency after the circulation begins. This resulted from that the water droplets sprayed through the 3-hole nozzle could not reach the reaction zone extended due to circulation of solids, while water from the 2-holes of nozzle reached the zone, leading to the effective wetting. These results confirm that the height of water spray instead of the number of holes is more significant factor for the optimization of water injection techniques. Figure 4 shows the effect of the water flow rate and the number of holes on the SO_2 removal efficiency under the circulation of Lime 2 at the gas velocity of 4 m/s. Water was sprayed at the flowrate of 30

ml/min and 40 ml/min through the two and three hole nozzles. The highest SO₂ removal efficiency was obtained with the water injection rate of 40 ml/min through the 2-hole nozzle. It is obvious that water sprayed higher and more can increase the wetting efficiency under the circulation of fine solids, leading to the increased SO₂ removal efficiency.

Figure 5 shows the effect of the fluidizing gas velocity on the SO₂ removal in a stationary bed. The water injection of 40 ml/min through the two hole nozzle was provided after Lime 2 was added into the bed at the fluidizing gas of 4 m/s and 5 m/s. As expected, the SO₂ removal efficiency increases slightly with the reduced fluidizing gas velocity since the residence time of SO₂ gas stream becomes greater. The effect of gas velocity on the SO₂ removal appeared not significant, however, resulting from that the wetting efficiency may increase by the relative velocity increased at higher gas velocities.

The SO₂ removal efficiency apparently increases with an increase of bed loading since the more sorbents provide a higher Ca/SO₂ ratio, which leads the removal efficiency to increase. Figure 6 indicates the SO₂ removals during fluidization with Lime 2 of 0.5 kg and 0.75 kg and with a water flowrate of 40 ml/min through the two hole nozzle. At the beginning of water injection the reaction appeared to be independent on the initial loading of bed materials, but the SO₂ removal efficiency significantly increased after the hydration and attrition of lime began. This result suggests that an effective wetting allowing the rapid hydration and gradual attrition of lime is required for the enhancement of the SO₂ removal efficiency. Such wetting may be obtained by adjusting the droplet size and water pressure.

During fluidization under wet conditions, limes are getting hydrated and obtruded easily. As a result, the removal of the product layer covering the unreacted sorbent by the attrition allows the surrounding SO₂ to contact with the fresh sorbent surface continuously, and leads to the increase of sorbent utilization. Figure 7 shows the effect of attrition on the SO₂ removal at two different water injection rates. The test was carried out with Lime 2 at the gas velocity of 5 m/s. While the parent solid stays due to high terminal velocity (10 m/s) in a bed during fluidization, the obtruded fines are immediately elutriated and captured by the first cyclone. The weight of fine solids captured by the cyclone was measured at a regular interval. As shown in Figure 7, the highest SO₂ removal efficiency and the largest extent of attrition occur at close time. In addition the peak is strongly dependent on the wetting efficiency. An effective wetting to cause the gradual attrition, therefore, is necessary for the increased SO₂ removal efficiency as well as sorbent utilization.

II. Unusual Problems/Circumstances

Nozzle clogging problems occurred during operation of CFBA. It to clean a nozzle A complete cleaning before installation of the nozzle was a way to prevent it from being clogged.

III. Tasks/Work to be Performed

A series of experiment in the bench scale CFBA unit will be continued to maximize the effectiveness of the new particle wetting system. For example, the tests will be continuously performed with other toroidal nozzles with different number of holes and angles. In addition, the wetting efficiency of the nozzle will be quantified, and its relationship with the SO₂ removal efficiency will be reported.

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Figure 1. SO₂ removal efficiency versus water injection rate.

Figure 2. Effect of the number of nozzle holes on SO_2 removal efficiency without circulation.

Figure 3. Effect of the number of nozzle holes on SO_2 removal efficiency with circulation.

Figure 4. Effect of water injection rate and number of holes on SO₂ removal efficiency.

Figure 5. Effect of gas velocity on SO_2 removal efficiency.

Figure 6. Effect of the initial weight of sorbents on SO_2 removal efficiency.

Figure 7. SO_2 removal and attrition at different water injection rate.

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