



Project Summary

Rate Controlling Processes and Enhancement Strategies in Humidification for Duct SO₂ Capture

D. K. Moyeda, G. H. Newton, J. F. La Fond, R. Payne, and J. C. Kramlich

The fundamental rate processes that govern sulfur capture in power plant ducts during humidification of flue gases were investigated. The specific application was the reactivation of partially sulfated calcium-based sorbents from in-furnace injection. The results suggest that physical contacting between the spray water and the sorbent particles is necessary to achieve significant reaction rates. Several means of promoting such contacting were investigated and a general approach to contacting was proposed. These hypothetical predictions were tested in a subscale rig using laser-based measurements. The reactivity of slurry drops was investigated in a dilute-phase reactor. The results indicate that calcium availability (i.e., dissolution into the liquid) and droplet lifetime were the principal constraints on sulfur capture. Increased concentrations of hydrate in the slurry droplets reduced the fractional conversion of sorbent to product. This was unexpected since calcium dissolution rate control would imply that conversion is independent of slurry concentrations. Also, the internal structure of the hydrate appears to contribute to the calcium availability. This suggests that approaches which seek to develop high specific surface areas for the sorbent within the slurry droplets will enhance sulfur capture.

This Project Summary was developed by EPA's Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back.)

Introduction

The use of calcium based materials as in-furnace sorbents for sulfur dioxide (SO₂) has received considerable recent research attention as a low-cost approach to intermediate levels of SO₂ control. Two ways to enhance the attractiveness of the approach are to (1) find an inexpensive way to improve overall sorbent conversion to product, and (2) develop an option for improving electrostatic precipitator (ESP) performance in collecting the furnace injected solids. One approach to both problems is the adiabatic humidification of the duct immediately downstream of the air heater to a nearly saturated condition. Previous testing has shown that additional sulfur capture is possible within this region. Also, humidification reduces both duct temperature (increases the ESP residence time) and ash resistivity; both of these factors improve ESP performance.

To implement humidification, a number of design decisions must be made. One of these includes the amount of humidification water and its method of introduction. The ideal situation is one

that improves performance, does not cause negative operating effects, and optimizes duct sulfur capture. To design for this condition, some indication of the processes that control the sulfur capture is needed. The work reported here defines the rate limiting steps that control the capture of SO₂ in the duct humidification system.

Most of the testing to date has been at pilot-scale. Test results show that, unless the sorbent particle is coated with bulk moisture, the reactivity is too low to allow significant reaction in duct residence times. Three mechanisms by which this physical wetting can occur have been identified:

1. Preslurring and duct injection of the sorbent.
2. Spraying of water into the duct, resulting in free stream inertial impaction of the water droplet on the sorbent.
3. Condensation, in which liquid collects on the surface of a cool (i.e., ambient temperature) particle injected into a warm, prehumidified gas stream.

Mechanism 2 is the only likely candidate for duct-reativation of the furnace injected sorbent; condensation is not likely because the sorbent is never colder than the surrounding gas. Thus, the only sorbent that is likely to be reactive in dispersed phase is that portion that directly collides with water droplets. Nozzle design should include a provision, if possible, for optimizing sorbent scavenging by water droplets via inertial impaction.

Once in the slurry phase, the reaction rate can be expected to be governed by one of the following processes:

- The rate at which SO₂ is transported from the free stream to the surface of the droplet.
- The rate at which solid calcium becomes available in the liquid phase.
- The aqueous reaction rate, including ionization and direct product formation.

The experiments were designed to evaluate the contribution of each of these fundamental steps to the overall rate, and to identify candidates for enhancement. The work was divided between enhancement of (1) scavenging and (2) reactivity.

Scavenging

The scavenging experiments were conducted in a facility in which a flowing suspension of hydrate in air passed a

subscale humidification nozzle (see Figure 1). The absorption of the sorbent into the water droplets was measured by a unique laser extinction technique that was developed as part of this program. The results were correlated by a one-dimensional model of the scavenging and reactivity process developed in-house.

Figure 2 shows typical results for an air-blast atomizer. The results show that the percent of the sorbent scavenged from the duct increases almost linearly with the nozzle water flow. The one-dimensional model results, also shown on the figure, are in close quantitative agreement. Figure 3 uses the model to show that the atomizer parameters that most strongly favor scavenging are (1) large droplet diameter, and (2) high droplet velocity. Unfortunately, these considerations run opposite engineering requirements for fitting sprays into confined spaces (i.e., fine droplets and long residence times to avoid wall wetting).

An investigation of the scavenging process showed that most of the scavenging takes place in the near field spray, near the nozzle. This "high efficiency zone" can be viewed as a volume fixed in space immediately downstream of the nozzle. One obvious enhancement strategy is to focus all of the sorbent-laden duct flow through this zone. This, however, does not result in any enhancement because the additional sorbent flux is counterbalanced by the reduced residence time available for scavenging. Thus, no net improvement occurs. Also, nozzles which entrain large amounts of surrounding gas will not improve scavenging because, again, the additional flux will be balanced by reduced residence time in the high efficiency zone. Another key point is that attempts to "one-dimensionalize" the flow by, for example, using many small nozzles, will not significantly improve scavenging. Basically, this is because doubling the volume of the high efficiency zone halves the scavenging rate per unit volume. Thus, the overall integrated result is constant.

The key to enhancing scavenging is to direct the sorbent, but not the gas flow, into the near field of the nozzle. Figure 4 shows that the scavenging was significantly enhanced when the sorbent was introduced near the nozzle, rather than mixing throughout the test duct. This can be practically effected by: (1) introducing sorbent near the nozzles (this, of course, is not an option for furnace sorbent reactivation), and (2)

separating the sorbent from the gas and concentrating it about the nozzle. Because of the small size of the sorbent, aerodynamic separation is not a likely candidate; however, electrostatic concentration is a possibility.

Reactivity

The slurry droplet reactivity experiments were conducted in a duct phase plug-flow reactor. As illustrated in Figure 5, the slurry was atomized by a rotating disk, and a small fraction of droplets produced were admitted through a slot into the reacting flow. The droplets were collected for the desired amount of time by a heated cup probe, and reaction extent was determined by chemical analysis. One note of importance is that the exact conditions and design of the sampling cup must be carefully controlled to prevent significant probe capture.

The results indicated that at low SO₂ concentrations the overall slurry reactivity was limited by external diffusion of SO₂ to the droplet surface. At high SO₂ concentrations, some form of internal control over calcium availability was evident. Figure 6 shows this shift in controlling mechanism at the point where utilization levels off with increasing SO₂ concentration. One key point is that at higher slurry concentrations, indicating that incremental sorbent reactivity is reduced as the solids concentration of the slurry increases. The practical consequence of this is that enhancement of scavenging will not lead to proportional enhancement of sulfur capture because of the decreased specific sorbent reactivity. It is of interest to note that similar overall behavior was observed in spray dryers. Here, reduced reactivity is manifested as a weak increase in capture when Ca/S increased above 1.0.

The importance of internal control under realistic conditions led to a further investigation of sorbent parameters which might be expected to influence the possible rate controlling steps. The factor was the importance of the external surface area of the sorbent, which was varied by comparing an atmospheric hydrate with a pressure hydrate. For the materials used, the difference in external surface area was about a factor of 5. As shown in Figure 7, the pressure hydrate displayed essentially identical reactivity. Thus, external surface area is not indicated to be a controlling factor. Figure 7 also shows a test of the importance of internal surface area. The normal calc

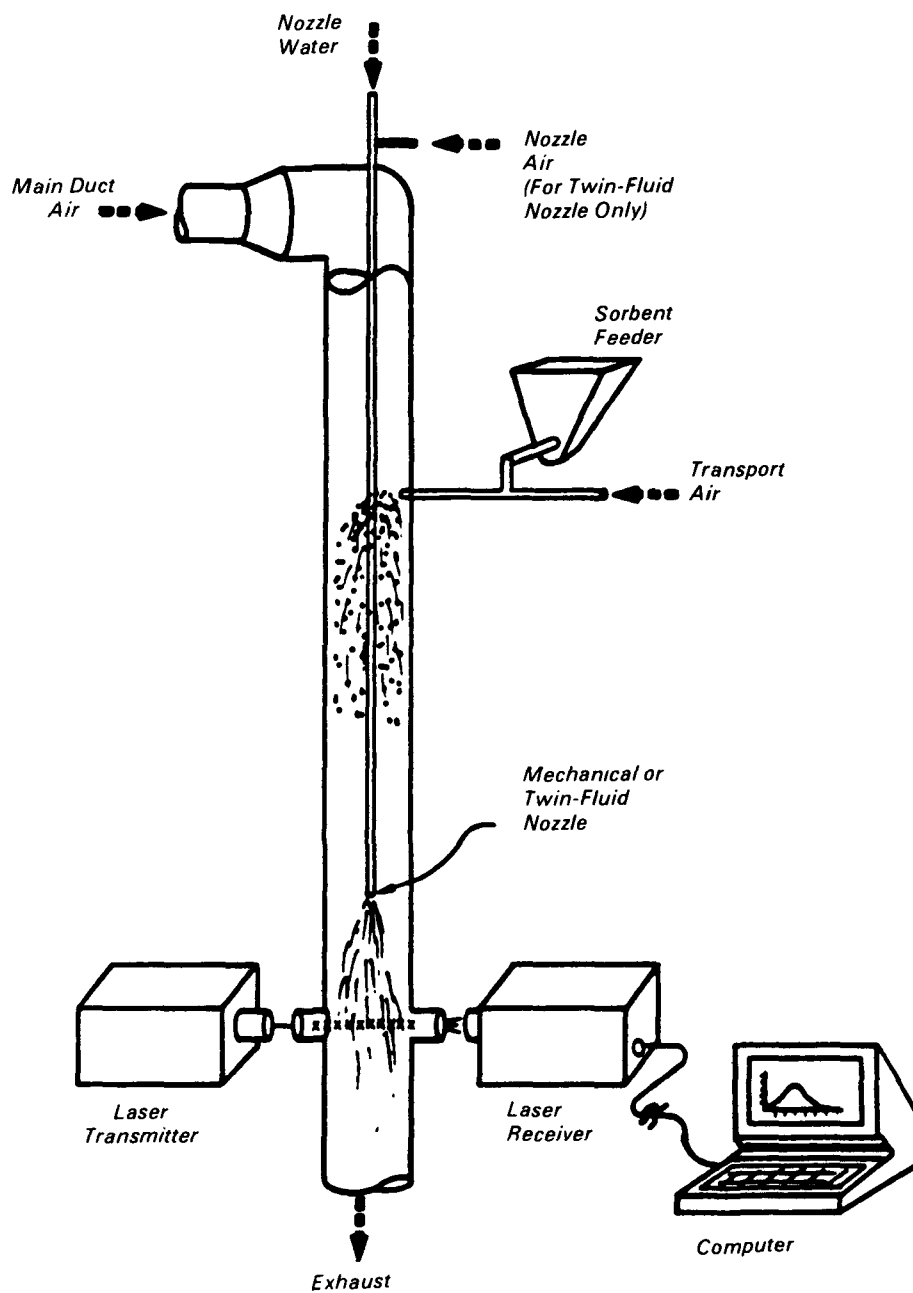


Figure 1. The scavenging rig.

hydrate was compared with an alcohol hydrate (i.e., hydrates prepared in an alcohol/water solution, which develop high internal surface areas). The figure shows that the conversion is similar in the external control regime (less than 500 ppm SO₂), but the high surface area sorbents excel at higher SO₂ values. The conclusion is that the higher surface area provides additional calcium availability,

which essentially allows the reaction to remain under SO₂ diffusion control to higher SO₂ concentrations. In other words, the "knee" in the curve is moved to higher SO₂ concentrations, and higher utilization values. For practical duct humidification, this suggests that, if a way could be found to form high surface area materials during the *in situ* hydration, higher utilizations would result.

Conclusions

The conclusions of this study include:

- Some way to physically wet the sorbent appears to be necessary to achieve sulfur capture rates of practical interest for duct applications. The principal way to do this in duct humidification

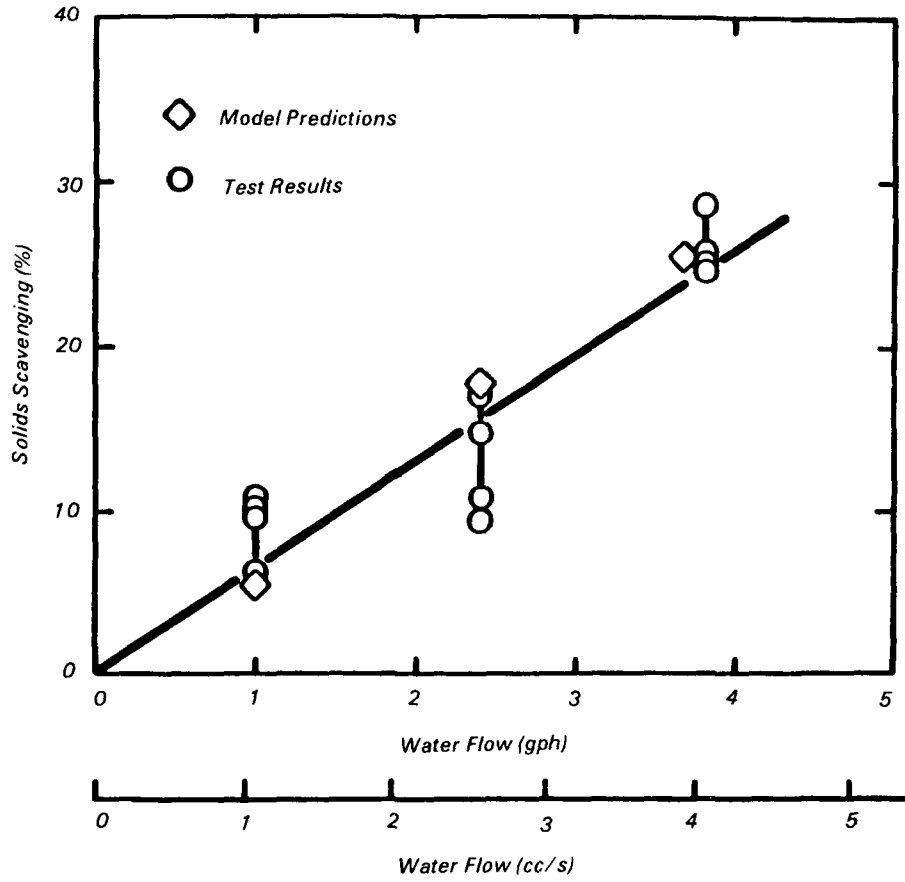


Figure 2. Scavenging results for the air-blast nozzle compared to one-dimensional model predictions.

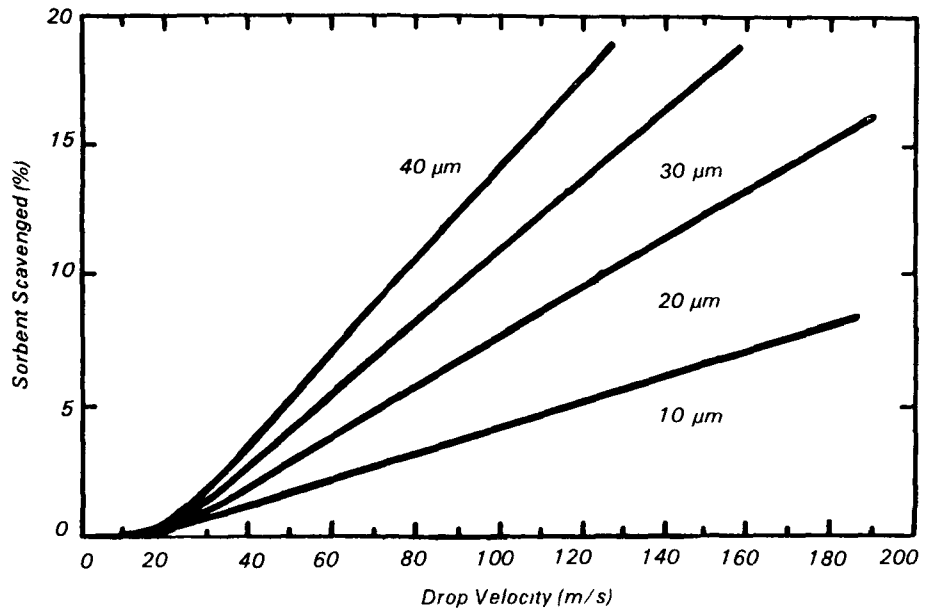


Figure 3. Predicted scavenging performance as a function of drop velocity and diameter (monosized spray)

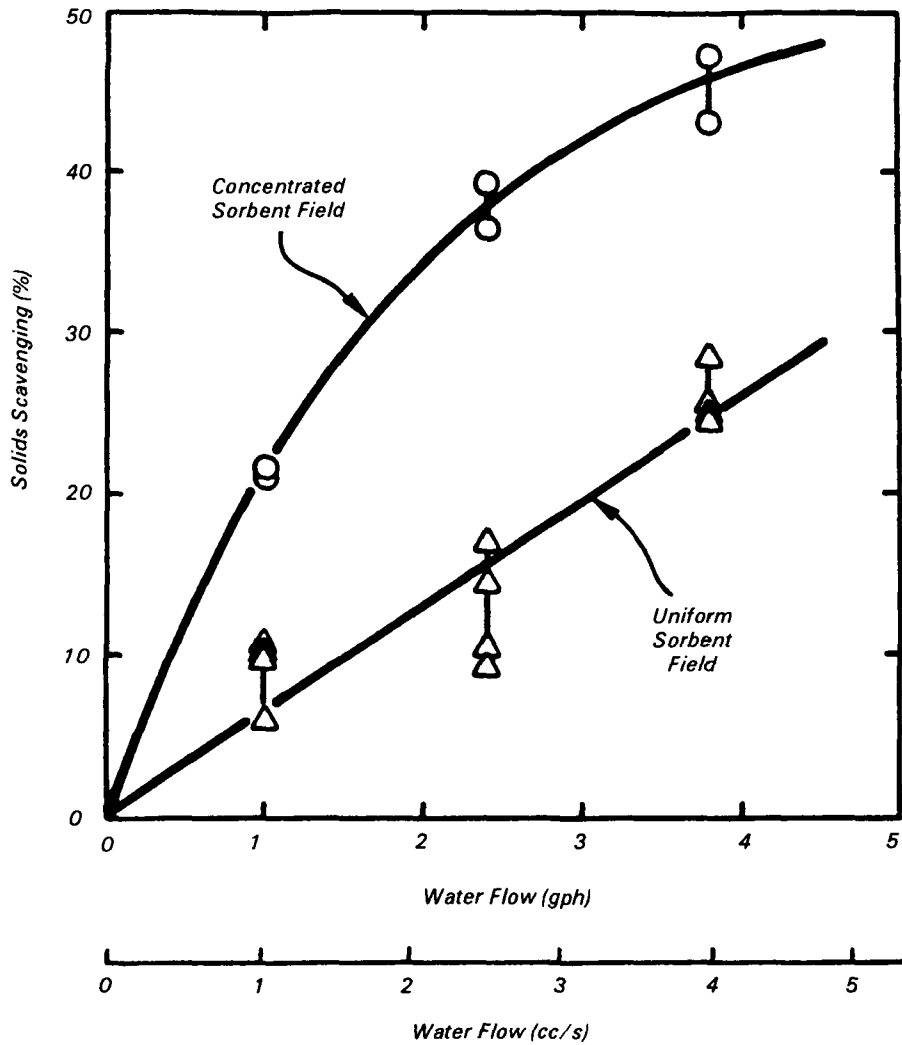


Figure 4. Increased scavenging by sorbent concentration.

appears to be sorbent scavenging by inertial impaction.

- Scavenging can best be enhanced by focusing the sorbent, but not the flow field, onto the region about the humidification nozzle.

- Under practical conditions, calcium utilization appears to be limited mainly by calcium availability within the slurry drop, and by the lifetime of the drop before evaporation to dryness.

- Calcium availability is enhanced by high internal surface areas for the hydrate.

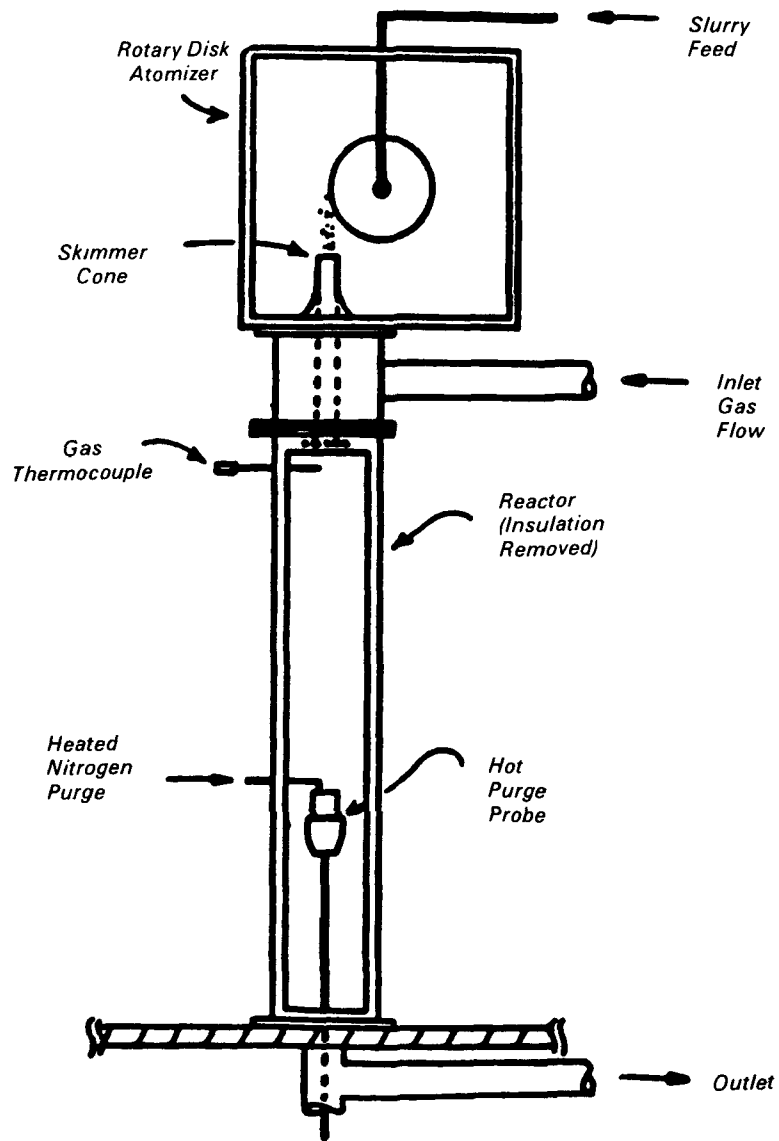


Figure 5. Single-drop reactor.

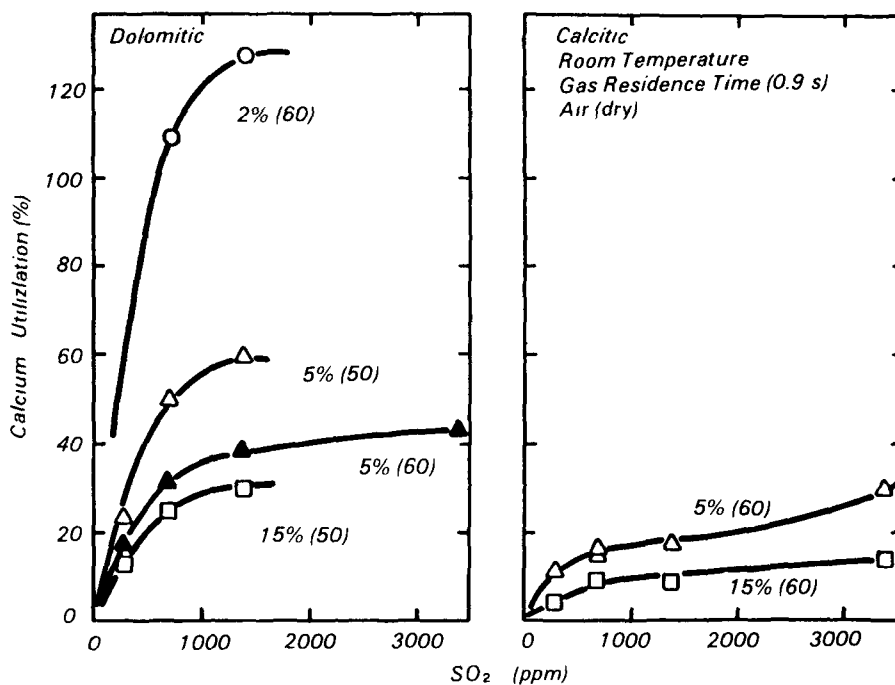


Figure 6. Calcium utilization as a function of SO₂ concentration at different slurry concentrations. The first number represents the slurry concentration, and the second number represents the atomizer voltage (a higher value represents a finer spray).

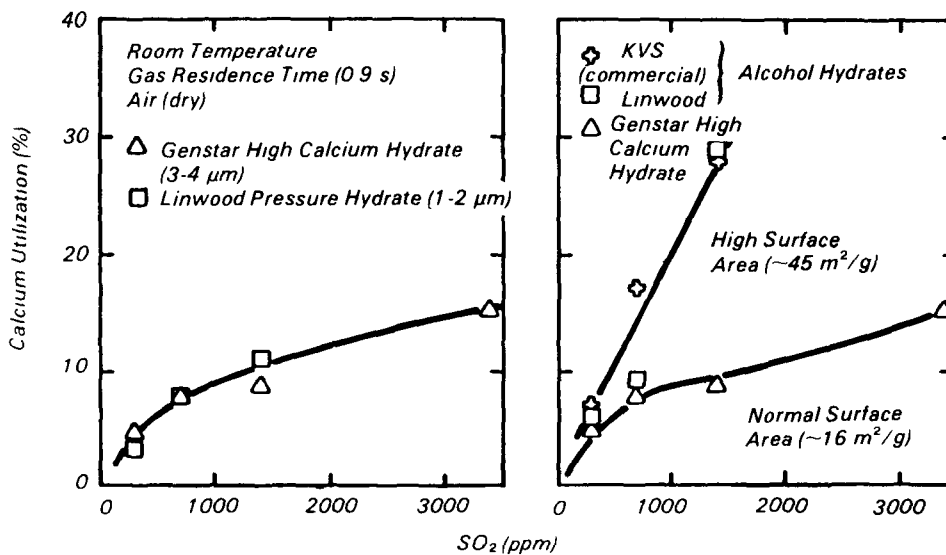


Figure 7 Influence of mass mean particle size and surface area on calcium utilization.

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Brian K. Gullett is the EPA Project Officer (see below).

The complete report, entitled "Rate Controlling Processes and Enhancement Strategies in Humidification for Duct SO₂ Capture," (Order No. PB 88-245 915/AS; Cost: \$19.95, subject to change) will be available only from:

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