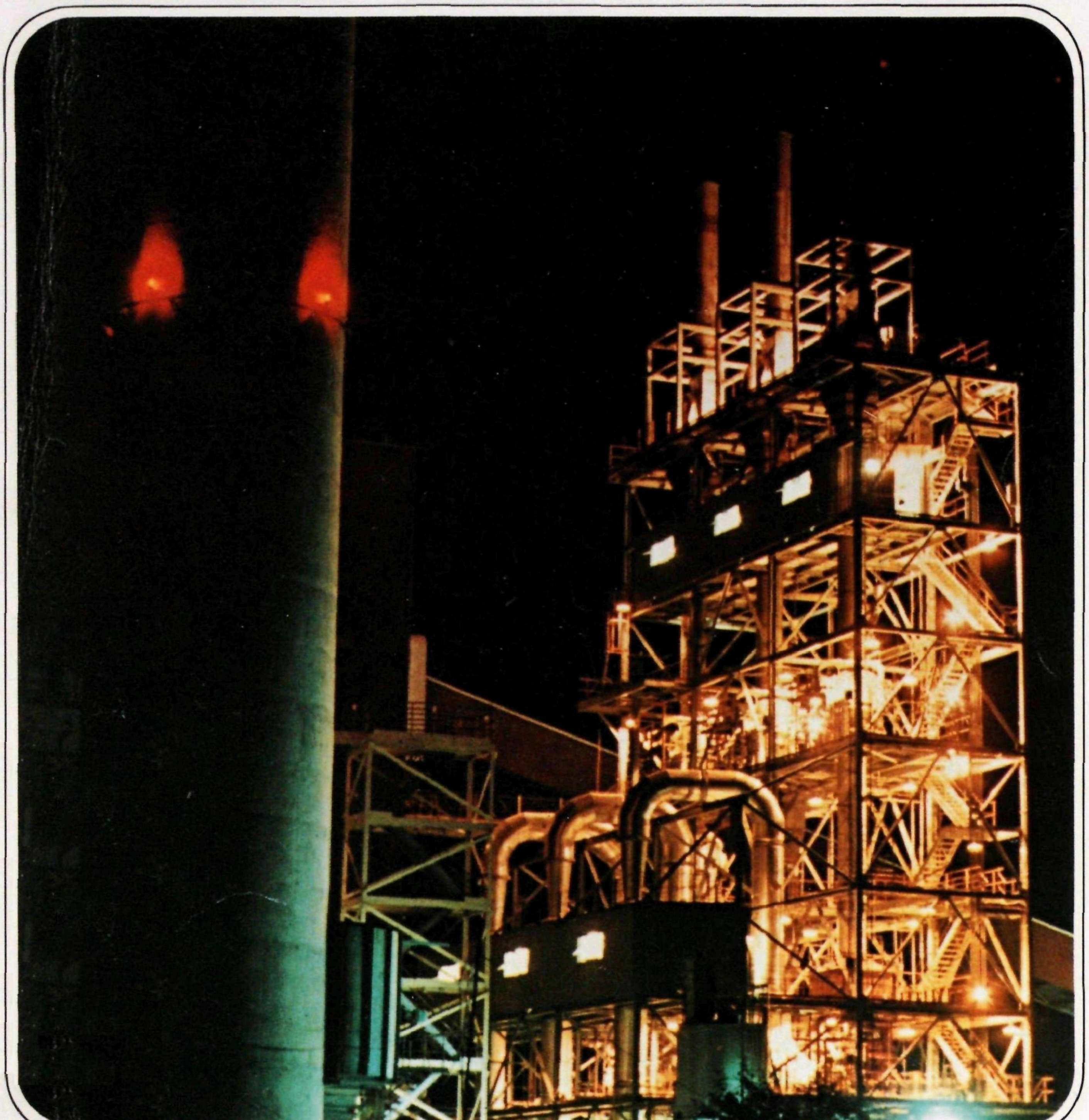
4 EPA TECHNOLOGY TRANSFER CAPSULE REPORT

FIRST PROGRESS REPORT:

LIMESTONE WET-SCRUBBING TEST RESULTS AT THE EPA ALKALI SCRUBBING TEST FACILITY

U.S. EPA OFFICE OF RESEARCH AND DEVELOPMENT PROTOTYPE DEMONSTRATION FACILITY



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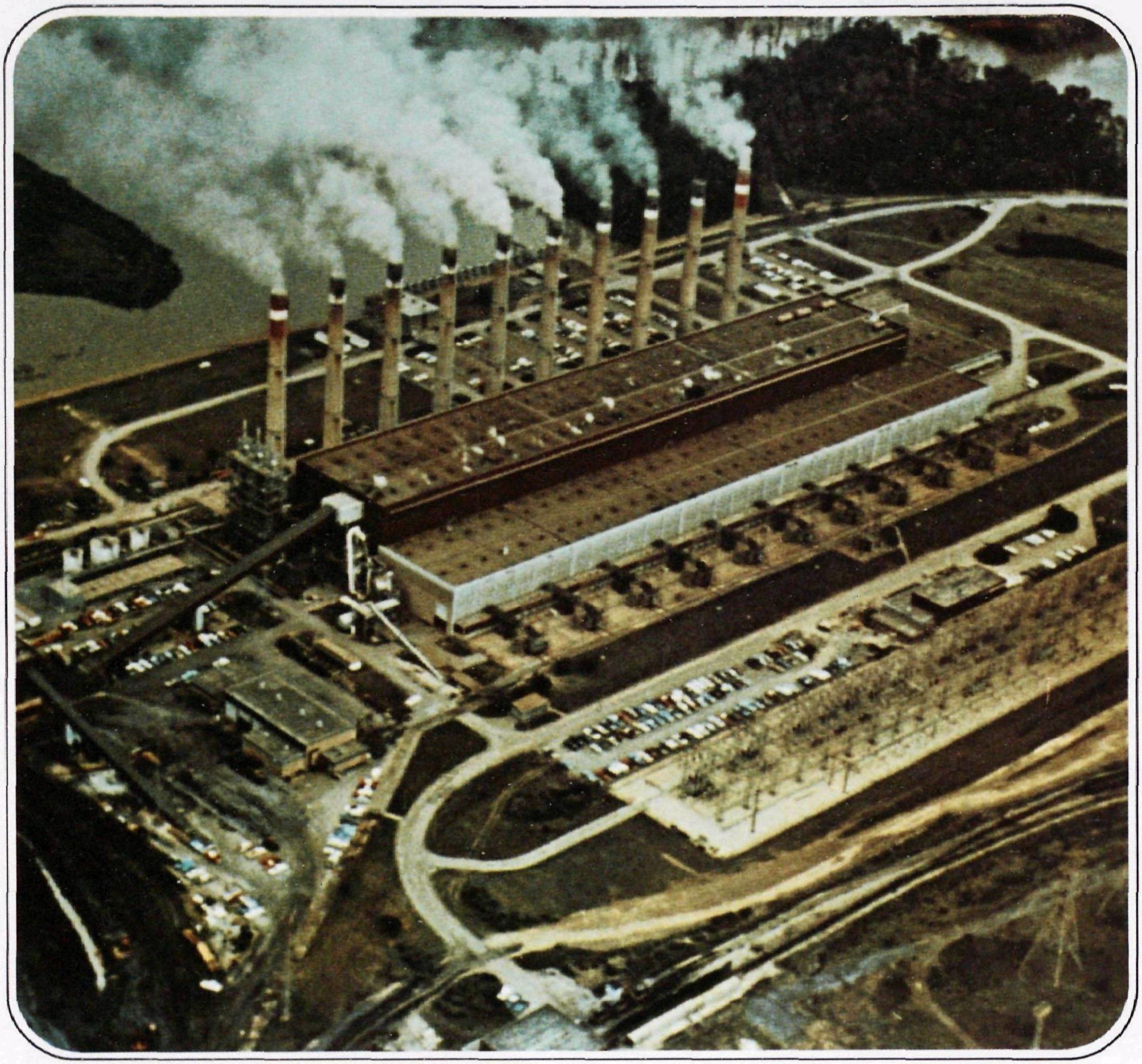

TVA Shawnee Steam Plant with Wet-Scrubbing Facility to Left of Boiler House

1. INTRODUCTION AND SUMMARY

The Clean Air Act of 1970 has now given increased emphasis to programs directed to decreasing SO_2 and particulate emissions from new and existing power plants and other facilities. At the present state of technology there are only three methods for substantially reducing oxide emissions: (1) switching to a low sulfur fuel; (2) desulfurizing the fuel; and (3) desulfurizing the gases produced. Among the more than 50 gas desulfurization control concepts which have been proposed and studied, the wet lime/limestone systems are considered to be the most advanced.

In a lime/limestone wet-scrubbing system, the flue gas is contacted (scrubbed) with a slurry of lime/limestone and water. The particulate matter (fly ash) is captured by liquid droplets and the sulfur dioxide (SO_2) is absorbed into the liquor where it reacts with the dissolved lime/limestone, forming the waste products of calcium sulfite and calcium sulfate (gypsum).

In June of 1968, the Environmental Protection Agency (EPA), through its Office of Research and Development (OR&D) and Control Systems Laboratory, initiated a program to test a prototype wet lime and limestone scrubbing system for removing sulfur dioxide and particulates from flue gases. Bechtel Corporation of San Francisco is the major contractor and test director, and the Tennessee Valley Authority (TVA) is the constructor and facility operator.

To minimize the time required for the technology from this project to become available to engineers and technical managers, a series of four technical reports are scheduled as work progresses. This capsule report discusses the highlights of the first detailed engineering progress report. It describes the test facility and test program and presents the results to date of the limestone wet-scrubbing testing. In addition, the reliability and operability of the test facility during long-term (2+ weeks) closed liquor loop operation is discussed.

The test facility consists of three parallel scrubber systems: a venturi followed by a spray tower; a Turbulent Contact Absorber (TCA); and a Marble-Bed Absorber. Each system is capable of treating approximately 10 Mw equivalent (30,000 acfm) of flue gas containing 2300 to 3300 ppm SO_2, and is integrated into the flue gas ductwork of a coal-fired boiler at the TVA Shawnee Power Station, Paducah, Kentucky.

The following sequential test blocks were defined for the program: (1) air/water testing; (2) sodium carbonate testing; (3) limestone wet-scrubbing testing; and (4) lime wet-scrubbing testing. The air/water and sodium carbonate tests have been completed. As of June 1973, short-term (less than 1 day) limestone wet-scrubbing factorial tests were 95 percent complete and longer-term (2+ weeks) reliability verification tests were approximately 50 percent complete. Long-term (4 to 10 months) limestone testing and lime testing are scheduled to begin in September, 1973.

The short-term factorial limestone tests were conducted at high scrubber inlet liquor pH (6.0-6.2). Series operation of the venturi and spray tower produced SO_2 removals of up to 80 percent at a total liquid-to-gas ratio of 80 gal/mcf and pressure drop of 10 in. H_2O. The three stage TCA scrubber obtained up to 96 percent SO_2 removal at a liquid-to-gas ratio of 64 gal/mcf and pressure drop of 7 in. H_2O. Removals of 80 percent were achieved with a single stage Marble-Bed Absorber at a liquid-to-gas ratio of 40 gal/mcf and a pressure drop of 11 in. H_2O.

The three initial long-term reliability verification tests with closed liquor loop operation have been run at reduced scrubber inlet liquor pH (5.7-5.9) and, consequently, at reduced stoichiometric ratio, in order to increase system reliability and limestone utilization (moles SO_2 absorbed/moles $CaCO_3$ added). For the TCA, limestone utilization was 83 percent with an SO_2 removal of 80 to 85 percent and a pressure drop of 7 in. H_2O.

The operability and reliability of the scrubber systems for the initial reliability verification tests have been good. There has been little evidence of sulfate or sulfite scale after approximately 500 hours of operation on all three systems, with effluent residence times greater than 20 minutes and percent solids recirculated greater than 10 percent (40 percent of solids is fly ash). Presently, more severe operating conditions (e.g., lower effluent residence times) are being tested to determine the regions of reliable operation for the three systems.

2. THE TEST FACILITY

The test facility consists of three parallel scrubber systems, each with its own slurry handling system. Scrubbers are of prototype size, each capable of treating approximately 30,000 acfm of flue gas from the TVA Shawnee coal boiler No. 10. Therefore, each circuit is handling the equivalent of approximately 10 Mw of power plant generating capacity. The equipment selected was sized for minimum cost, consistent with the ability to extrapolate results to commercial scale. The 30,000 acfm scrubber train was judged to meet these requirements. Boiler No. 10 burns a high-sulfur bituminous coal leading to SO_2 concentrations of 2300-3300 ppm and particulate inlet loadings of about 2 to 5 grains/scf in the flue gas.

The major criterion for scrubber selection was the potentiality for removing both sulfur dioxide and particulates at high efficiencies (sulfur dioxide removal greater than 80 percent and particulate removal greater than 99 percent). Other factors considered in the selection of the scrubbers were: (1) ability to handle slurries without plugging or excessive scaling; (2) reasonable cost and maintenance; (3) ease of control; and (4) reasonable pressure drop.

Based on the information available in the literature, the following scrubbers were selected:

- Venturi followed by a spray tower after-absorber
- Turbulent Contact Absorber (TCA)
- Marble-Bed Absorber

The venturi, manufactured by Chemical Construction Co., contains an adjustable throat that permits control of pressure drop under a wide range of flow conditions. Although a venturi is ordinarily an effective particulate removal device, gas absorption is limited in limestone wet-scrubbing systems by low slurry residence time. For this reason the after-absorber was included for additional absorption capability. The TCA, manufactured by Universal Oil Products, uses a fluidized bed of low density plastic spheres that are free to move between retaining grids. The Marble-Bed Absorber, supplied by Combustion Engineering Co., uses a packing of ¾-inch glass spheres (marbles). A "turbulent layer" of liquid and gas above the glass spheres enhances mass transfer and particulate removal. Figures 1, 2, and 3, drawn roughly to scale, show the three scrubber types along with the demisters selected for de-entraining slurry in the gas streams.

The test facility was designed so that a number of different scrubber internals and piping configurations can be used with each scrubber system. For example, the TCA can be operated as a one-, two- or three-stage unit, and solids separation can be achieved with any combination of clarifier, filter, centrifuge, and pond.

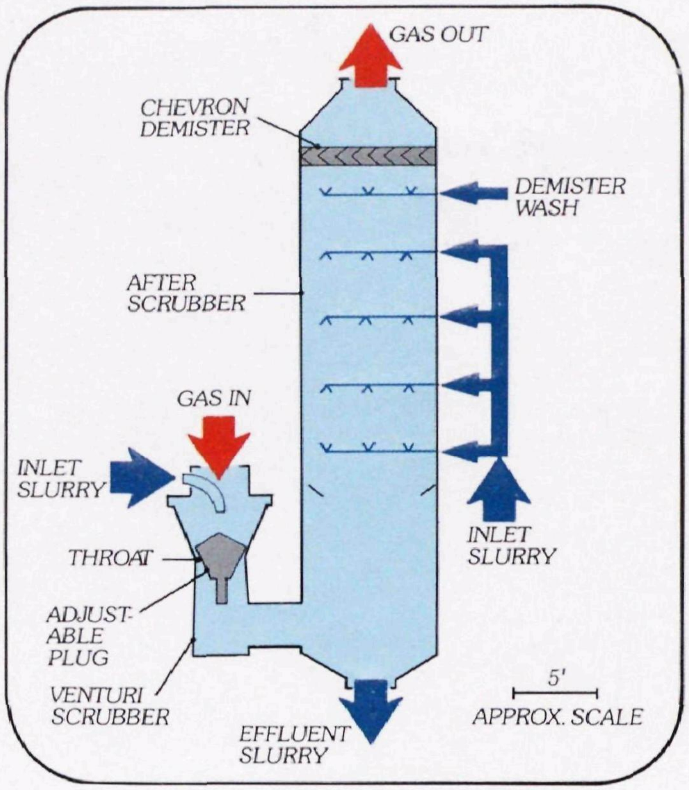

Figure 1. Schematic of Venturi Scrubber and After-Absorber

A typical TCA system configuration used during limestone testing is shown schematically in Figure 4. Process details, such as flue gas slurry saturation sprays and demister or Koch tray wash sprays, are not shown.

For all configurations, gas is withdrawn from the boiler ahead of the power plant particulate removal equipment so that entrained dust (fly ash) can be introduced into the scrubber. The gas flow rate to each scrubber is measured by venturi flow tubes and controlled by dampers on the induced-draft fans. Concentration of sulfur dioxide in the inlet and outlet gas is determined continuously by DuPont photometric analyzers.

Control of the scrubbing systems is carried out from a central graphic panelboard. An electronic data acquisition system is used to record the operating data. The system is hard-wired for data output

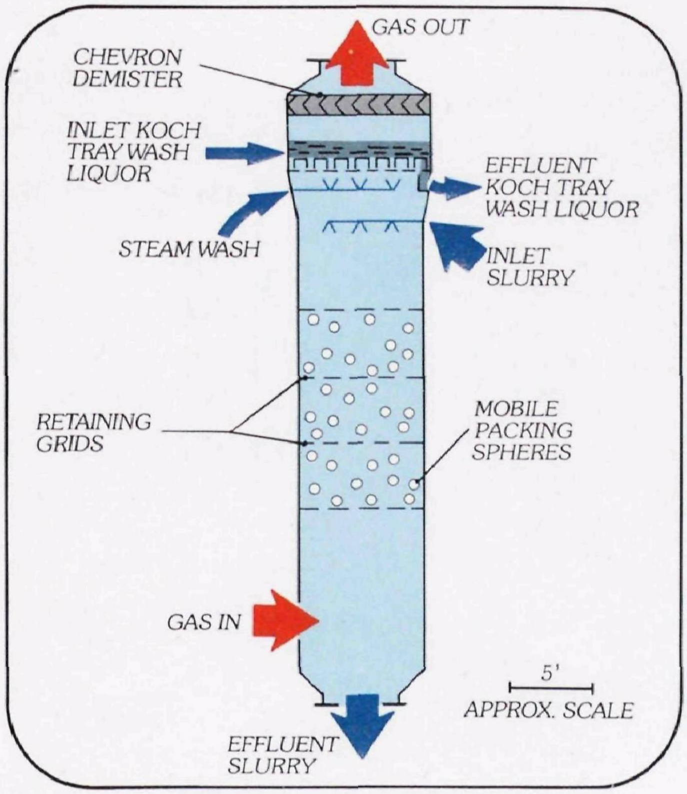

Figure 2. Schematic of Three-Stage TCA

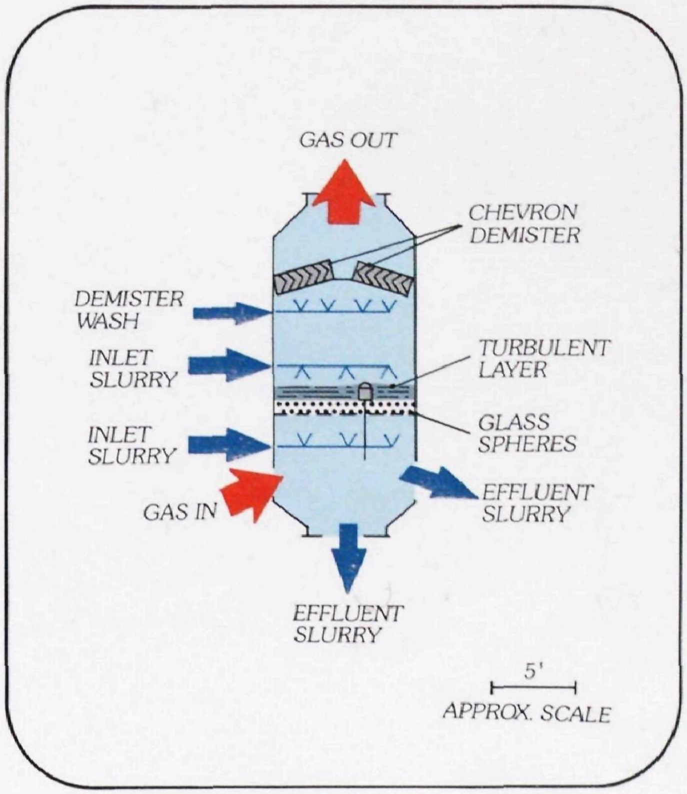

Figure 3. Schematic of Marble-Bed Absorber

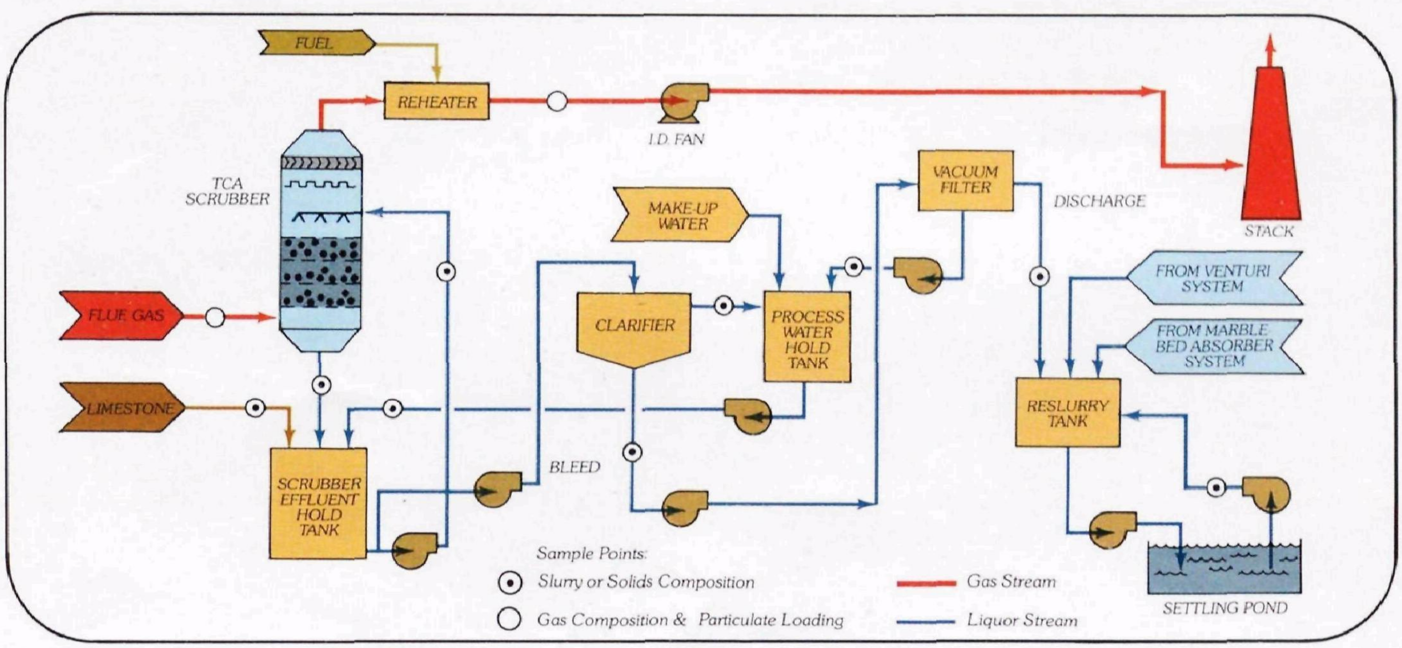

Figure 4. EPA Test Facility—Typical Process Flow Diagram for TCA System

directly on magnetic tape, and on-site display of selected information is available. Also, important process control variables are continuously recorded, and trend recorders are provided for periodic monitoring of selected data sources.

The Shawnee facility contains five major areas: (1) the scrubber area (including tanks and pumps), (2) the operations building (including laboratory area, electrical gear, centrifuge, and filter), (3) the thickener area (including tanks and pumps), (4) the utility area (including air compressors, air dryer, limestone storage silos, mix tanks, gravimetric-feeder, and pumps), and (5) the pond area.

The scrubber area (looking toward the powerhouse), scrubber control room, and the operations building and thickener area are shown in Figures 5, 6, and 7, respectively.

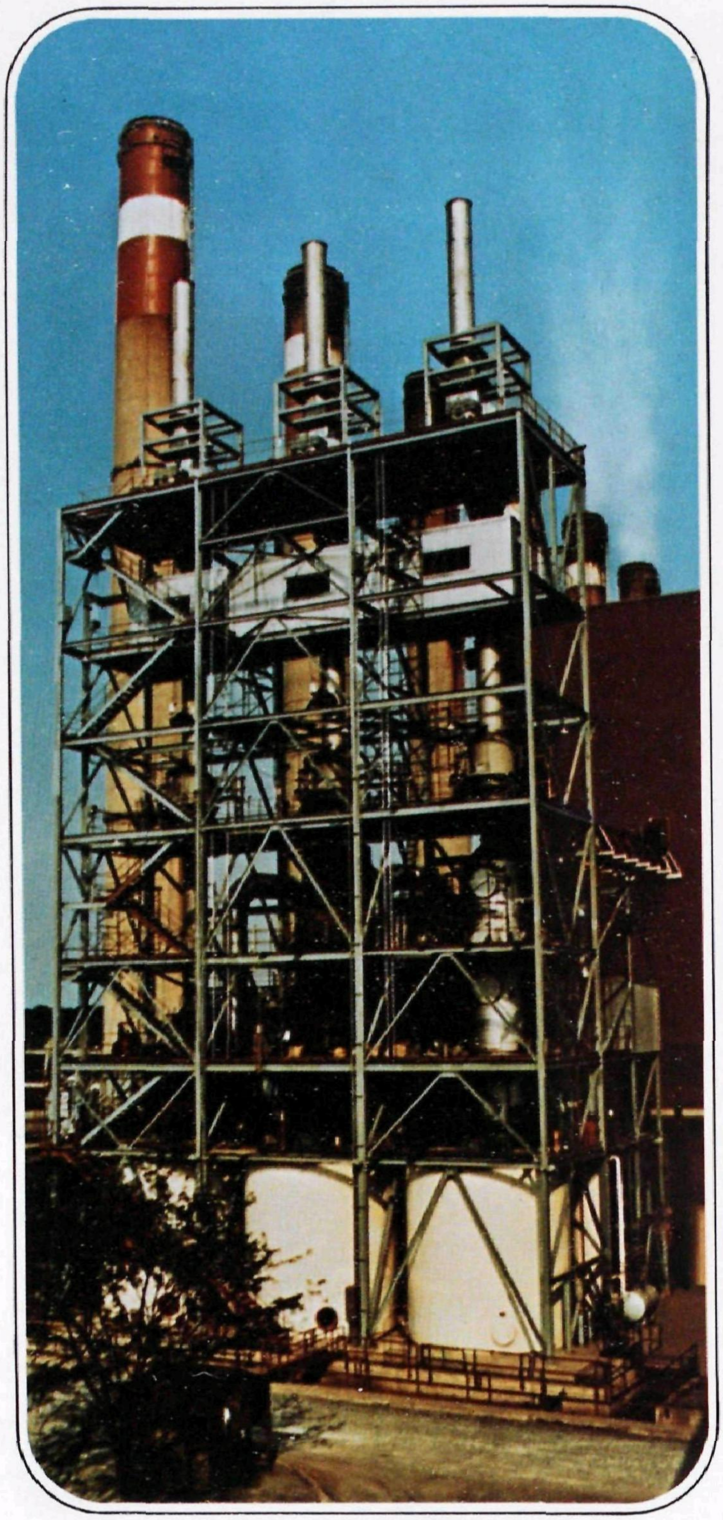

Figure 5. Scrubber Area

Figure 6. Control Room

Figure 7. Operations Building & Thickener Area

3. THE TEST PROGRAM

The following sequential test blocks have been defined for the test program:

(1) Air/water testing
(2) Sodium carbonate testing
(3) Limestone wet-scrubbing testing
(4) Lime wet-scrubbing testing

The test program schedule is shown in Figure 8. As indicated, the air/water and sodium carbonate tests have been completed. As of June 1973, limestone wet-scrubbing short-term factorial tests were approximately 95 percent complete and longer-term reliability verification tests were approximately 50 percent complete. In Table 1, a description of the reports which are presently scheduled for general distribution is presented.

AIR/WATER TESTING

These experiments, which use air to simulate flue gas and water to simulate alkali slurry, were designed to determine pressure drop model coefficients and observe fluid hydrodynamics for all three scrubbers under clean conditions.

SODIUM CARBONATE TESTING

These experiments, which utilized both high- and low-concentration sodium carbonate solutions to absorb SO_2 from flue gas, were designed to determine coefficients within the mathematical models for predicting SO_2 removal.

LIMESTONE WET-SCRUBBING TESTING

The primary objectives of this test sequence are:

(1) To characterize, as completely as practicable, the effect of important independent variables on particulate removal and SO_2 removal.
(2) To identify and resolve operating problems, such as scaling and demister plugging.
(3) To identify areas or regions for reliable operation of the three scrubber systems, consistent with reasonable SO_2 removal, and to choose economically attractive operating configurations from within these regions.
(4) To determine long-term operating reliability with attractive configurations for one or more of the scrubber systems and to develop more definitive process economics data and scale-up factors.

To accomplish the first objective a large number of short-term (4+ hours) factorial tests have been made for each scrubber system. The test sequences were full or partial factorial designs based upon the chosen independent variables, their levels, and the restraints of time as outlined in Figure 8. The choice of the independent variables and their levels was based on pilot plant test results, the restraints of the system, and results from mathematical models that relate the dependent and independent variables.

To accomplish the second and third objectives, a relatively small number of longer-term (2+ weeks) reliability verification tests are being made on each scrubber system. The variables being investigated are: (1) percent solids recirculated, (2) effluent residence time, (3) gas rate, (4) scrubber inlet liquor pH, and (5) demister types (e.g., plastic chevron vs. stainless chevron). Solids separation tests for the clarifier, filter, and centrifuge are also being made on the three systems throughout the test period.

The fourth objective will be accomplished by running reliability tests, lasting from 4 to 10 months, on attractive operating configurations for one or more of the scrubber systems. During these tests, the systems will be carefully monitored for potential long-term reliability problems such as erosion and corrosion of system components. The ability to effectively operate such systems under varying gas rate, particulate loading, and SO_2 inlet concentrations will also be studied.

LIME WET-SCRUBBING TESTING

This test series, which involves introduction of hydrated lime (calcium hydroxide) directly into the scrubber circuit, will resemble the limestone wet-scrubbing test program. Again, tests will be divided into three general categories (see Figure 8): short-term factorial tests, longer-term reliability verification tests, and long-term reliability tests.

ANALYTICAL PROGRAM

Samples of slurry, flue gas, limestone, and coal are taken periodically for chemical analyses, particulate size sampling, and limestone reactivity tests. Locations of slurry and gas sample points for the TCA are shown in Figure 4.

To meet the formidable analytical requirements of the facility at reasonable costs, equipment has been selected that minimizes manpower. For example, an x-ray fluorescence unit is used for comprehensive slurry analyses. All analytical computations and recording of results are handled by an on-site minicomputer.

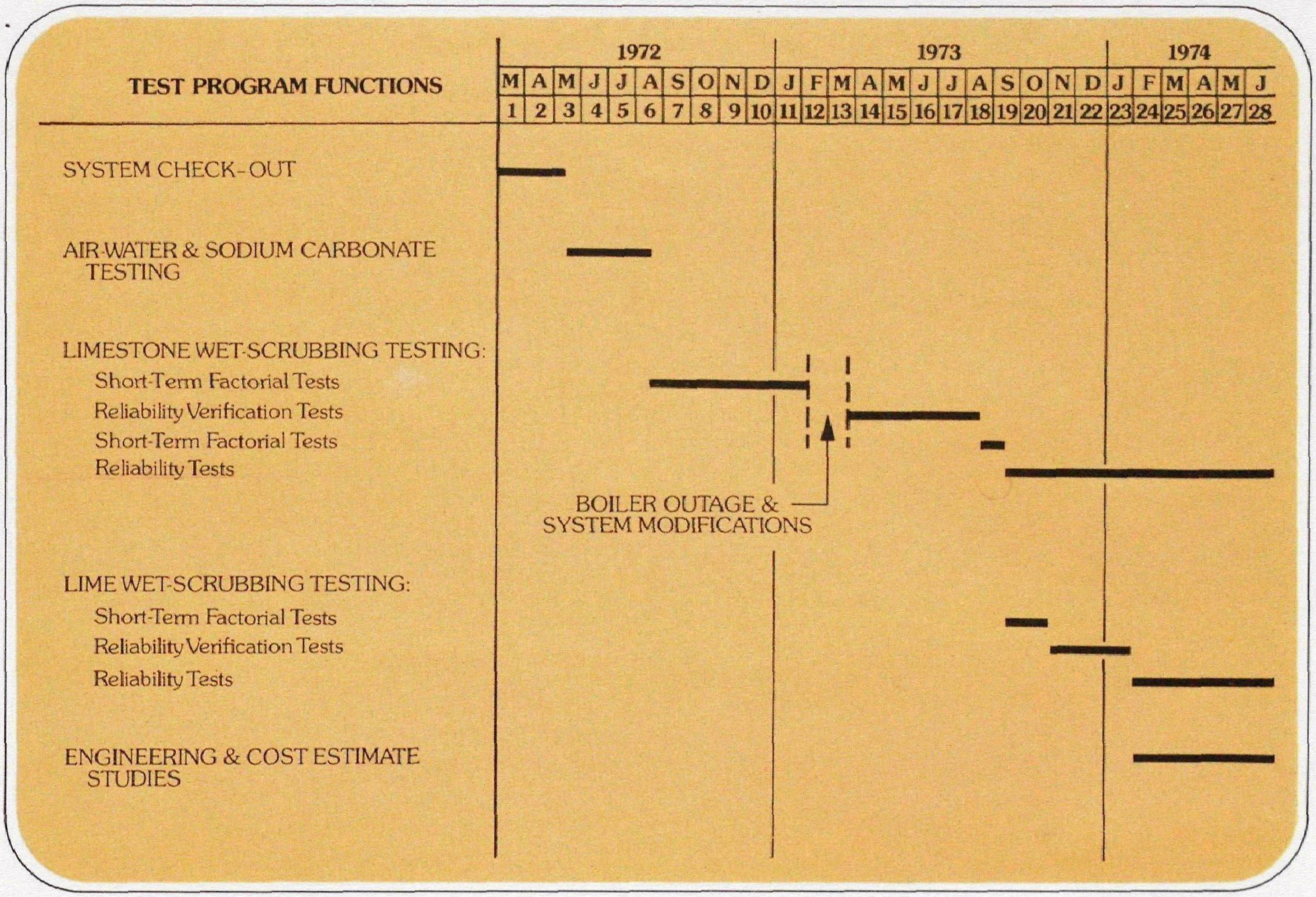

Figure 8. Shawnee Test Schedule

Table 1
*PROGRESS AND FINAL REPORT DESCRIPTION**

	Report Title	Information to be Included	Estimated General Publication Date
1.	EPA Alkali Scrubbing Facility: Sodium Carbonate and Limestone Test Results	Summary of operational problems and resolutions, planned and actual test designs, results of air-water and Na_2CO_3 testing, utilization of data for model development, results of factorial limestone testing with interpretation of data.	August 1973
2.	EPA Alkali Scrubbing Facility: Limestone Wet-Scrubbing Test Results	Summary of operating problems and resolutions associated with reliability verification testing, planned and actual test designs, interpretation of data, status of process model development, and selection of parameters for limestone long-term reliability testing.	November 1973
3.	EPA Alkali Scrubbing Test Facility: Lime Wet-Scrubbing Test Results	Summary of operational problems and resolutions associated with lime reliability verification testing, planned and actual test designs, results of factorial lime testing, status of process model development, interpretation of data, and status of limestone reliability testing.	March 1974
4.	EPA Alkali Scrubbing Test Facility: Final Report	Summary of total test program with particular emphasis on lime and limestone reliability test results, mathematical models, scale-up design, and economic studies.	July 1974

*It is planned that EPA Capsule Reports will be issued which summarize each of the progress and final reports. This Capsule Report summarizes the August 1973 progress report.

4. SHORT-TERM FACTORIAL TEST RESULTS

In this section some of the significant SO_2 removal results from the limestone short-term factorial test sequences are presented graphically. As mentioned previously, the objective of the factorial tests was to characterize, as completely as practical, the effect of independent variables (e.g., liquid-to-gas ratio) on particulate and SO_2 removal.

A majority of the short-term (4+hours) factorial tests were made at "high" scrubber inlet liquor pH's (6.0-6.2) and, consequently, high stoichiometric ratios (>1.75 moles $CaCO_3$ added per mole SO_2 absorbed). The data collected indicated that stoichiometric ratio has an insignificant effect upon SO_2 removal at values of inlet liquor pH greater than 6.0. The data also indicate that SO_2 absorption increases with decreasing inlet gas SO_2 concentration and/or with decreasing scrubber liquor temperature. Care, therefore, has been exercised in segregating these noncontrolled independent variables in the presentation of the data.

For the venturi scrubber, the SO_2 removals varied between 30 and 40 percent for gas rates from 15,000 to 30,000 acfm and for liquid-to-gas ratios from 10 to 60 gal/mcf. SO_2 removal was not significantly affected by pressure drop within the region investigated (6-12 in. H_2O).

In Figure 9, the effect of gas velocity and L/G on SO_2 removal for the four-header spray tower is shown. For these data, the SO_2 removals are outside the range of interest for commercially acceptable gas velocities (>7 ft/sec). An analysis of the data has shown that an L/G of approximately 110 gal/mcf would be required to achieve 80 percent SO_2 removal at gas velocities greater than 7 ft/sec.

In Figure 10, the effect of gas velocity and L/G on SO_2 removal for a three-stage TCA scrubber is shown. For 95 percent removal, at a gas velocity of 9.8 ft/sec and an L/G ratio of 64 gal/mcf, the pressure drop was approximately 7 in. H_2O (excluding Koch tray).

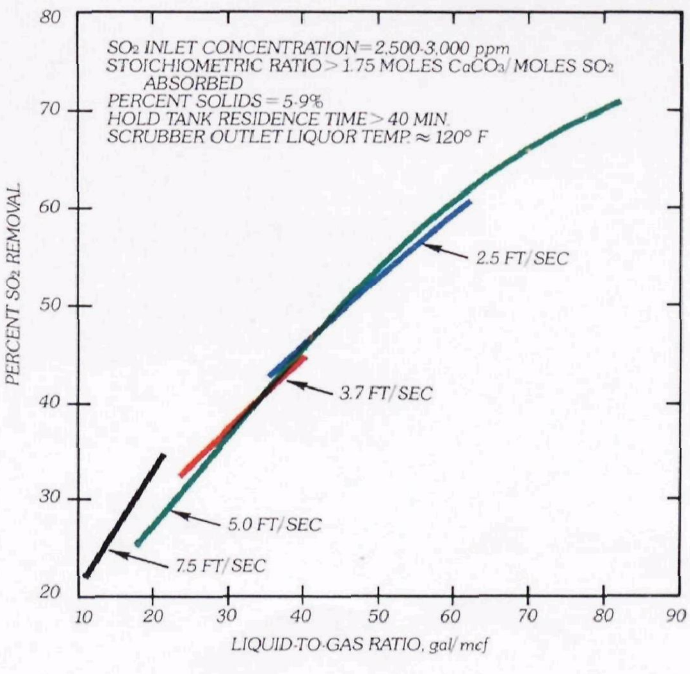

Figure 9. Effect of Liquid-To-Gas Ratio and Gas Velocity on SO_2 Removal in the Four-Header Spray Tower

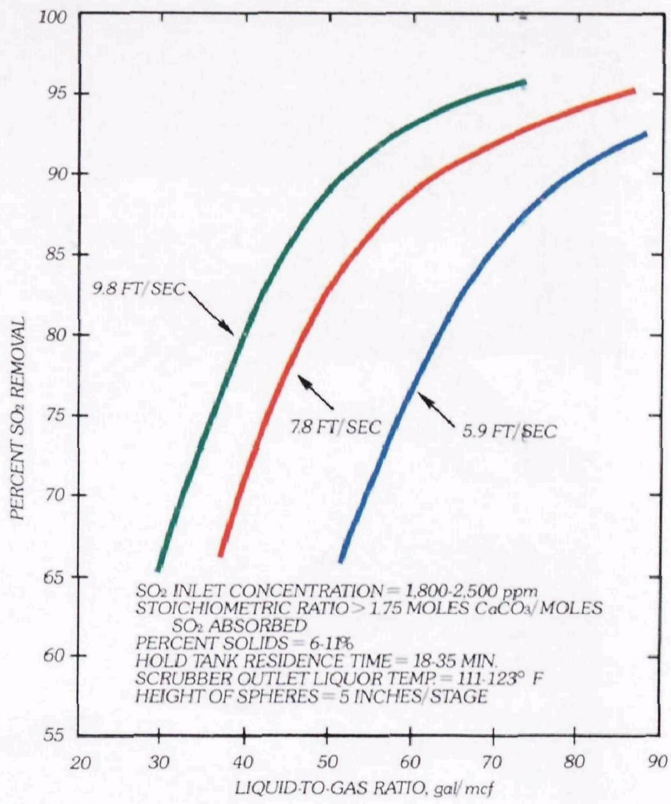

Figure 10. Effect of Liquid-To-Gas Ratio and Gas Velocity on SO_2 Removal in the Four-Grid Three-Stage TCA

In Figure 11, the effect of gas velocity and L/G on SO_2 removal for the Marble-Bed Absorber with five inches of marbles is shown. For 80 percent removal, at a gas velocity of 7.7 ft/sec and an L/G ratio of 40 gal/mcf, the pressure drop was approximately 11 in. H_2O.

The data presented in Figures 9, 10 and 11 have been replotted in Figure 12, where the effect of slurry flow rate and gas velocity on SO_2 removal for the spray tower, TCA and Marble-Bed Absorber is shown. The data indicate that SO_2 removal is a strong function of liquor rate and is only slightly affected by gas velocity within the region investigated.

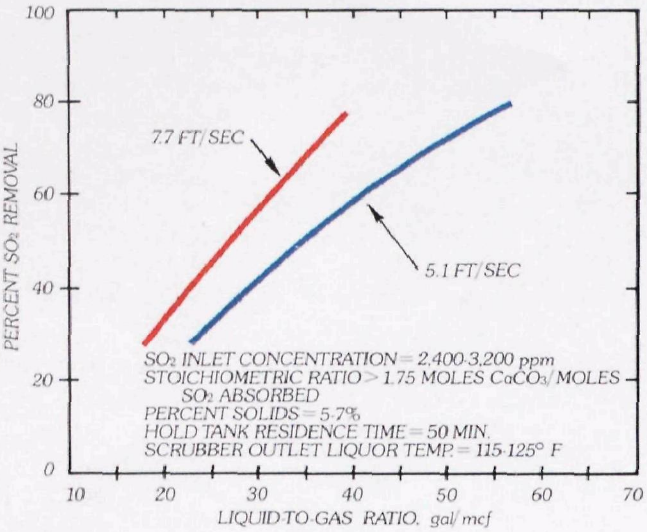

Figure 11. Effect of Liquid-To-Gas Ratio and Gas Velocity on SO_2 Removal in the Marble-Bed Absorber with Five Inches of Marbles

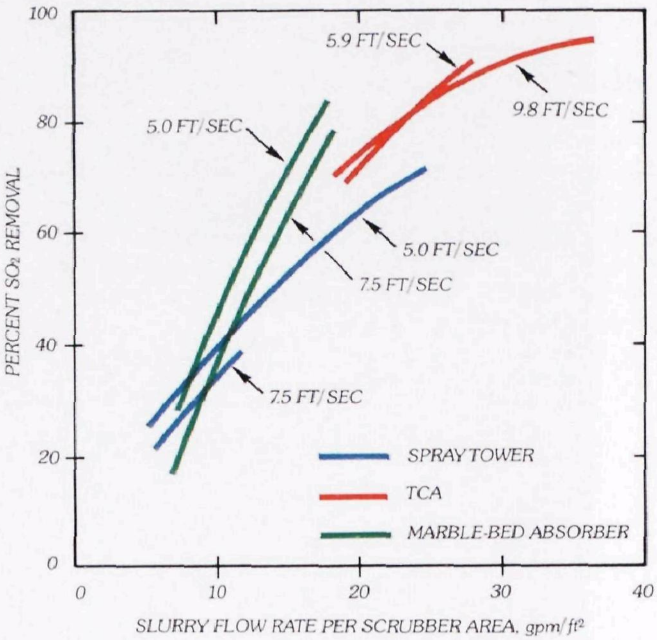

Figure 12. Effect of Slurry Flow Rate and Gas Velocity on SO_2 Removal for the Spray Tower, TCA and Marble-Bed Absorber

5. RELIABILITY VERIFICATION TEST RESULTS

As mentioned previously, the objectives of the limestone reliability verification tests are to: (1) identify and resolve operating problems (e.g., demister plugging and scaling), and (2) identify areas or regions for reliable operation, consistent with reasonable SO_2 removal and choose economically attractive operating configurations from within these regions.

A majority of these tests will be made at reduced scrubber inlet liquor pH's (5.7-5.9) to increase system reliability and limestone utilization. System reliability can be improved at reduced pH's because of higher oxidation rates, resulting in a larger percentage of "seed" $CaSO_4$ crystals within the process slurry. An increase in limestone utilization (decrease in stoichiometric ratio) results, of course, in a reduction in waste mass solids and in limestone requirements. The penalty for operating at reduced inlet liquor pH is a modest reduction in SO_2 removal from high pH performance.

A summary of the data from the initial limestone reliability verification tests of the venturi, TCA, and Marble-Bed Absorber systems is given in Table 2. The highest limestone utilization (83 percent) and SO_2 removal (80-85 percent) was obtained with the TCA system. The operability and reliability of all systems were good during the duration of the testing (e.g., no scale buildup), and the Koch Flexitray wash tray (see Figure 2) was particularly effective in deentraining slurry from the exiting gas stream. Overall material balances for sulfur and calcium were in agreement to better than 12 percent for the three runs.

Reliability verification tests will be conducted next for all systems at increased gas rates, decreased effluent tank residence times, and decreased percent solids recirculated in the process liquor. Also, the venturi system has been modified to allow for increased liquor flow to the spray tower. This will result in higher SO_2 removal efficiencies for that system.

Table 2
A SUMMARY OF INITIAL RELIABILITY VERIFICATION RUNS

Parameters	**Venturi and Spray Tower Run 501-1A**	**TCA Run 501-2A**	**Marble-Bed Absorber Run 501-3A**
Operating Time, hr	410	550	520
Gas Velocity, ft/sec	5†	7.8	5
L/G, gal/mcf	80*	80	53
Pressure Drop, in. H_2O	10.5**	6	9
Percent Solids Recirculated	15	15	11
Effluent Tank Residence Time, min	20	20	30
Percent SO_2 Removal	70-75	80-85	65-70
Stoichiometric Ratio, moles $CaCO_3$ added/moles SO_2 absorbed	1.5	1.20	1.25
Limestone Utilization, 100 x moles SO_2 absorbed/moles $CaCO_3$ added	67%	83%	80%
Scrubber Inlet Liquor pH	5.8-5.9	5.8	5.8
Percent Oxidation of Sulfite to Sulfate	15	20-30	30
Dissolved Solids, ppm	7000	7500	8000

† Spray tower.
*L/G's of 40 for spray tower and 40 for venturi.
**9 inches across venturi and 1.5 inches across spray tower.

6. OPERABILITY AND RELIABILITY

Generally, the operability and reliability of the three scrubber systems has been good throughout the short-term factorial testing and the initial phase of the limestone verification testing. It should be emphasized that there is little evidence of sulfate ($CaSO_4$) scale for any of the tests to date. Sulfite ($CaSO_3$) scale was encountered during one series of TCA tests and has been attributed to scrubber operation at inlet liquor pH's > 6.3. Presently, more severe operating conditions (e.g., lower effluent residence times) are being tested to determine the regions of reliable operation for the three systems.

This section will highlight the most significant results, to date, affecting the operability and reliability of the systems. An evaluation of spray nozzle and material reliability has not yet been completed and, consequently, will not be reported here.

CLOSED LIQUOR LOOP OPERATION

Early in the short-term factorial test period it became apparent that it was not feasible to operate the test facility in a totally closed liquor loop without modifications. For a closed liquor loop, the raw water input to the system is nearly equal to the water normally exiting the system in the humidified flue gas and in the waste sludge transferred to the pond. In an open liquor loop system, raw water input is significantly greater than the water outflow in the exit gas and sludge. Therefore, process liquor must be discharged from the system to maintain an overall water balance. In a commercial system, such a discharge may not be acceptable due to potential water pollution problems. Also during open-loop operation, reliability may be unintentionally enhanced since the additional raw water added dilutes liquors returning to the scrubber, thereby tending to reduce scaling and plugging. Open-loop operation was not considered to be a serious problem during the short-term factorial testing, since, at a specified scrubber inlet liquor pH, SO_2 removal is not significantly affected by liquor composition.

In order to allow for closed liquor loop operation on all three systems, the following modifications were made to the facility during the 5-week boiler outage in February and March, 1973: (1) water seals on the pumps were converted to mechanical seals supplemented with air purge, (2) quench spray systems using circulating slurry were provided for the TCA and Marble-Bed Absorber, and (3) mist eliminator and Koch tray wash systems using process liquor plus raw water makeup were provided for the scrubber systems.

DEMISTER OPERATION

During the short-term factorial test period, the demister sections on the scrubber systems were provided with top (downstream) wash sprays only and, consequently, there was a continual accumulation of soft mud-like deposits on the demister blades. In order to remedy this problem, the following modifications were made to the systems:

(1) In November, 1972, a Koch Flexitray wash tray was installed in the TCA scrubber between the inlet liquor spray header and the chevron demister (see Figure 2). At first, irrigation was obtained with raw water. A subsequent modification in February, 1973, allowed for irrigation with process liquor, diluted with the available raw water makeup.

(2) During the boiler outage in early 1973, the spray tower and Marble-Bed Absorber demister systems were modified to allow for washing from both the upstream (underside) and downstream directions with process liquor, diluted with the available raw water makeup (see Figures 1 and 3).

The Koch Flexitray wash tray has been successful, to date, in eliminating solids buildup on the TCA demister blades for gas velocities up to 8.8 ft/sec and 15 percent solids recirculated. For the spray tower, washing the demister from both the topside and underside has been successful, to date, in eliminating solids buildup for gas velocities up to 5 ft/sec and 10 percent solids recirculated. Some difficulty is still being experienced with solids buildup on the Marble-Bed Absorber demister blades. This may be attributed to "channelling" of the gas through the marble-bed and the resultant high gas velocities, due to partial pluggage of the bed.

REHEATER OPERATION

Flue gas is reheated after evolving from the scrubber to increase plume buoyancy, prevent condensation in the exhaust system, facilitate isokinetic and analytical sampling, and protect the induced draft fans from solid deposits and droplet erosion. The reheaters employed are fuel oil combustion units with a separate combustion air supply and with combustion occurring in the flue gas flow stream. During the short-term factorial testing, it had been difficult to start and keep the reheaters operating and, during operation, combustion had been incomplete leading to a visible plume containing significant quantities of soot and oil. This led to difficulties in interpreting outlet particulate data and affected gas sampling by the DuPont SO_2 photometric analyzers. The difficulty appeared to result from quenching of the flame due to the flue gas flow before complete combustion could occur.

Modifications were made to the reheater systems during the boiler outage in early 1973. Internal stainless steel sleeves were installed that provide approximately 50 ft^3 of isolated combustion zone for each reheater. Also, the turbulent mixing type nozzles supplied originally were replaced with mechanical atomizing nozzles.

To date, the above modifications appear to have been effective. Essentially no soot is visible in the stack gas and the outlet particulate samples have shown no evidence of carbon from the reheaters.

HOT GAS LIQUID INTERFACE

The hot flue gas must be cooled (humidified) before entering the neoprene rubber lined spray tower, TCA, and Marble-Bed scrubbers. During the early stages of reliability verification testing, there was a continual problem of solids buildup at the gas humidification sections on the TCA and Marble-Bed scrubbers. There has never been evidence of any solids buildup within the venturi scrubber, which is an extremely reliable gas humidifying device.

By careful selection of soot-blowing schedules and humidification spray locations and flow patterns, the solids buildup problem has been brought under control. During the current series of limestone reliability verification testing, there has been no evidence of solids buildup at the hot gas-liquid interfaces for the TCA and Marble-Bed scrubbers.

For further information:

The detailed Progress Report on this project is available from the National Technical Information Service, Springfield, Va. 22151, as EPA Report PH 22-68-67, "First Progress Report: Limestone Wet-Scrubbing Test Results at the EPA Alkali Scrubbing Test Facility."

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