

Advanced Sulfur Control Concepts for Hot-Gas Desulfurization Technology

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EXECUTIVE SUMMARY

Good progress was made on both the experimental and process modelling fronts during the past quarter. All experimental tests used the fixed-bed laboratory reactor to study the sulfidation of CeO_2 with H_2S and the regeneration of $\text{Ce}_2\text{O}_3\text{S}$ using SO_2 . A number of experimental problems were solved (or at least alleviated) during the quarter including malfunctioning mass flow controllers, excessive bed pressure drop, and elimination of the H_2S plateau during early stages of sulfidation tests.

Most CeO_2 sulfidation tests were carried out at 800°C and 5 atm using a sulfidation gas containing 1% H_2S , 10% H_2 , balance N_2 . At these conditions sulfidation of CeO_2 was rapid and complete. Sulfur material balance closure was satisfactory, and, except for the unexpected H_2S plateau during the prebreakthrough period, the sulfidation results were as expected. Near the end of the quarter, the cause of the H_2S plateau was tentatively identified as being due to reaction between H_2 and elemental sulfur deposited downstream of the sorbent in the bottom of the reactor and in tubing leading to the gas chromatograph. The sulfur deposits occurred during regeneration tests, and chemically cleaning the lines between regeneration and sulfidation coupled with reducing the temperature of the transfer line during sulfidation greatly reduced the H_2S plateau. A brief examination of the effect of sulfidation temperature between 700 and 850°C showed relatively little temperature effect, although the slope of the active portion of the breakthrough curve was somewhat smaller at 700°C , which is consistent with a smaller reaction rate at this temperature.

Early regeneration studies were troubled by high pressure drop through the sorbent bed. The vapor pressure of SO_2 at ambient temperature is only about 3.4 atm, and bed pressure drop during early tests was of the same magnitude. In some regeneration tests which were accompanied by an increase in bed pressure drop with time, the regeneration run had to be terminated when SO_2 could no longer be fed to the reactor. The excessive pressure drop problem was solved by dry pressing CeO_2 into tablets, then crushing and sieving the tablets and using CeO_2 particles in the 150 to 300 μ diameter range. The bed pressure drop was reduced to about 5 psi as a result.

All regeneration tests except one were conducted at 600°C . An early test at 350°C showed the regeneration rate to be quite slow at that temperature. The regeneration gas consisted of SO_2 in N_2 with the concentration increased from 1% in early tests to 12% at the end of the quarter. In all cases (except at 350°C), regeneration was effectively complete and the regeneration rate, as measured by the slope of the breakthrough curve, was rapid. Sulfur material balance closure was generally good and elemental sulfur concentrations in the regeneration product gas as high as 12% have been produced.

Multicycle tests consisting of two and three complete sulfidation-regeneration cycles were carried out near the end of the quarter. Little, if any, sorbent deterioration was observed.

More detailed flow diagrams were prepared for three process concepts involving high temperature desulfurization with elemental sulfur production. Two of the concepts involve two-stage desulfurization using CeO_2 followed by Zn_2TiO_4 , and regeneration of Ce_2O_2S with either steam or SO_2 . The third concept involves single-stage desulfurization with Zn_2TiO_4 , regeneration of ZnS with dilute oxygen, and conversion of SO_2 to elemental sulfur using the DSRP process. Process material and energy balances using coal gas representative of the product from a Shell gasifier were completed at selected sorber and regenerator temperatures. Sensible energy provided by the coal gas coupled with exothermic energy provided by the reactions is sufficient to satisfy all process needs and to provide excess high pressure steam which may be "sold" to supplement power generation in an IGCC plant.

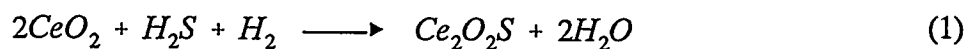
Qualitative comparisons listing advantages and disadvantages of the three process concepts are discussed on the basis of process simplicity, power requirements, and raw material and by-product requirements. At this stage, the two-stage process involving steam regeneration appears to be less attractive than the other processes. Reasons include the additional processing units--the Claus reactor and tail gas treatment--required to convert H_2S to elemental sulfur. Single-stage desulfurization with DSRP is somewhat simpler than two-stage desulfurization with SO_2 regeneration, but is more power intensive. In addition to the lower power requirement, the elemental sulfur product from SO_2 regeneration is free of water and should be more desirable. Final comparison of the two processes must be based on the economic analysis.

CERIUM OXIDE SULFIDATION/REGENERATION

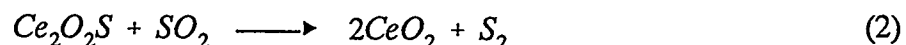
At the end of the previous quarter, initial CeO_2 sulfidation tests had been completed. Complete sulfidation with good sulfur material balance closure and approximately 90% H_2S removal during the prebreakthrough period had been achieved. These early results were quite good except for the larger than expected prebreakthrough H_2S concentration plateau. No Ce_2O_2S regeneration tests had been attempted at that time.

Significant progress was made during the current quarter on both the sulfidation and regeneration fronts. The important chemical reactions are:

Sulfidation



Regeneration



A summary of reaction conditions for all tests completed in the current quarter is presented in Table 1. The "s" in the run number is used to designate a sulfidation test while the

Table 1. Summary of Fixed Bed Reactor Test Conditions: CeO₂ Sulfidation and Regeneration, October - December 1996.

Run Date	Ce-04s1 10/7	Ce-04r1 10/7	Ce-05s1 10/11	Ce-06s1 10/14	Ce-06r1 10/14	Ce-07s1 10/28	Ce-08s1 10/30	Ce-08r1 10/30	Ce-08s2 10/31	Ce-09s1 11/4	Ce-09r1 11/4	Ce-09s2 11/6	Ce-09r2 11/6	Ce-10s1 11/13	Ce-11s1 11/13
Reactor Packing															
CeO ₂ ,g	3.00	3.00	3.00 ^a	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00
Al ₂ O ₃ ,g	3.00	3.00	3.00 ^a	6.01	6.01	6.00	5.00	5.00	5.00	3.00	3.00	3.00	3.00	3.00	3.00
Reaction Conditions															
Temp., °C	800	350 ^b	800	800	600	800	800	600	800	800	600	800	600	700	750
Press., atm	5.4 ^c	1.9 ^c	5.0 ^c	5.0 ^c	2.8 ^c	5.1 ^c	5.2 ^c	2.2~3.0 ^c	4.9 ^c	5.0	2.5	5.0	2.7	5.0	5.0
Gas Composition															
%H ₂ S	1	0	1	1	0	1	1	0	1	1	0	1	0	1	1
%SO ₂	0	1	0	0	1	0	0	1	0	0	1	0	1	0	0
%H ₂	10	0	10	10	0	10	10	0	10	10	0	10	0	10	10
%N ₂	89	99	89	89	99	89	89	99	89	89	99	89	99	89	89
Total Gas Flowrate, scfm	400	400	300	300	300	300	300	300	300	300	300	300	300	300	300
Sulfur Material Balance (% of Stoichiometric)	100.2	--	52.5	--	--	93.9	102.3	95.1 ^d	65.1	110.6	106.6	104.2	111.9	--	--

a: Special mixture of 50% CeO₂-50% Al₂O₃ obtained from W. Wilson.

b: Temperature was increased to 700°C at the end of the run.

c: Pressure is the average between the reactor inlet and outlet. In those runs designated with the superscript c, the pressure was established by the bed pressure drop. In runs without the superscript c, the pressure was established using a back pressure regulator.

d. Estimated from incomplete data.

Table 1. Summary of Fixed Bed Reactor Test Conditions: CeO₂ Sulfidation and Regeneration, October - December 1996. (Continued)

Run Date	Ce-12s1 11/22	Ce-13s1 11/24	Ce-14s1 11/25	Ce-15s1 11/27	Ce-15r1 11/27	Ce-15s2 12/2	Ce-15r2 12/2	Ce-15s3 12/4	Ce-15r3 12/4	Ce-16s1 12/19	Ce-16r1 12/19	Ce-16s2 12/20	Ce-16r2 12/20	Ce-16s3 12/23	Ce-16r3 12/23
Reactor Packing															
CeO ₂ ,g	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	6.00	6.00	6.00	6.00	6.00	6.00
Al ₂ O ₃ ,g	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00	3.00
Reaction Conditions															
Temp., °C	750	850	850	800	600	800	600	800	600	800	600	800	600	800	600
Press., atm ^a	5.0	5.0	5.0	5.0	2.0	5.0	1.0	5.0	1.0	5.0	1.0	5.0	1.0	5.0	1.0
Gas Composition															
%H ₂ S	1	1	1	1	0	1	0	1	0	1	0	1	0	1	0
%SO ₂	0	0	0	0	2	0	4	0	8	0	12	0	12	0	12
%H ₂	10	10	10	10	0	10	0	10	0	10	0	10	0	10	0
%N ₂	89	89	89	89	98	89	96	89	92	89	88	89	88	89	88
Total Gas Flowrate, scfm	300	300	300	300	300	300	300	300	300	200	200	400	300	400	300
Sulfur Material Balance (% of Stoichiometric)	--	--	--	84.6	--	91.9	--	99.2	--	--	90.8	75.0	90.4	86.6	86.0

"r" identifies a regeneration test. The last digit is used to identify the cycle number. Most sulfidation tests were conducted at 800°C and 5 atm using a feed gas containing 1% H₂S, 10% H₂, and balance N₂. The exceptions were runs Ce-10s1 through Ce-14s1 where sulfidation temperature was varied between 700 and 850°C.

The first regeneration test, Ce-O6r1, used 350°C regeneration temperature. Regeneration was quite slow and all subsequent regeneration tests have used 600°C where the regeneration rate is rapid. Regeneration pressure has varied between 1 and 3 atm. The maximum regeneration pressure is limited by the vapor pressure of SO₂ which is 3.4 atm at ambient temperature. Regeneration pressure in early tests was established by bed pressure drop. After the excessive pressure drop problem was solved, most regeneration have been conducted at 1 atm. An SO₂-N₂ gas mixture has been used in all regeneration tests. Early tests used 1% SO₂ in N₂, and the SO₂ content was gradually increased to 12% in the last regeneration tests.

Feed gas flow rates of either 300 or 400 sccm were used in all sulfidation tests while the regeneration feed gas rate varied between 200 and 400 sccm. The flow rate variations were generally made to control the total duration of the test, rather than to specifically study the effect of volumetric flow rate on kinetics.

Cerium oxide obtained from Rhone-Poulenc was used in all tests except Ce-05s1, where a special mixture of 50% CeO₂-50% Al₂O₃ obtained from W. Wilson was used. The CeO₂ was physically mixed with the Al₂O₃ in the amounts shown in Table 1. Because of the extremely small particle size of the Rhone-Poulenc CeO₂, pressure drop across the packed bed was approximately 3 atm, thus placing severe restraints of the regeneration studies. While the bed pressure drop in Ce-05s1 was reduced using the special CeO₂-Al₂O₃ mixture, this sorbent was less reactive. The pressure drop problem was solved beginning in run Ce-15s1 using this sorbent by dry pressing the Rhone-Poulenc CeO₂ powder at 25,000 psi in a hydraulic press to form tablets. The tablets were crushed and sieved, and particles in the size range of 150 to 300 μ were mixed with Al₂O₃ having a particle size range of 80 to 200 μ to form the packed bed. As a result, bed pressure drop was reduced to about 5 psi without appreciable adverse effect on the sulfidation and regeneration breakthrough curves.

The last entry in Table 1 presents results of the tests in terms of sulfur material balance closure expressed as percent of stoichiometric sulfur according to reactions (1) and (2). These material balances were calculated by comparing the experimental sulfidation or regeneration breakthrough curves with a nonreacting tracer breakthrough curve at the same conditions. Poor material balance closure associated with run Ce-05s1 was due to the slow sulfidation rate of the alternate CeO₂-Al₂O₃ mixture. The test was terminated before completion.

Progress during the quarter did not occur without interruptions, due both to equipment malfunction and the need to alter gas chromatograph operating conditions. For example, the two-week gap between runs Ce-06r1 and Ce-07s1 was caused by failure of the SO₂ mass flow controller. In November, the O-ring shoulder on two quartz insert tubes broke in successive runs, and about one week was lost in getting them repaired. No breakage has occurred since

then. Finally, following the December series of regeneration tests using successively higher SO_2 concentrations, Ce-15r1 through Ce-15r3, the increasingly short duration of the tests made it necessary to alter gas chromatograph conditions to reduce the time interval between samples. Regeneration test Ce-15r3 was effectively complete in 20 minutes, and only four product samples were obtained with the five minute sampling interval in use at that time. Prior to run Ce-16s1, the chromatograph operating conditions were altered to permit sampling every 3.3 minutes; however, this change also required that the chromatograph be recalibrated.

Sulfidation Results

Results of a typical early sulfidation test, Ce-06s1, in terms of an H_2S breakthrough curve, are shown in Figure 1. Rhone-Poulenc CeO_2 was mixed with Al_2O_3 in a 1:2 mass ratio, and the total gas flow rate was to 300 sccm. The reduced CeO_2 to Al_2O_3 ratio and flow rate were both made in an effort to reduce bed pressure drop. These changes were unsuccessful, however, and the 5 atm pressure in Table 1 represents the arithmetic average of the bed inlet and outlet pressures. The back pressure regulator was not used in this test.

After a brief start-up period, the H_2S concentration in the sulfidation product gas increased to a steady plateau corresponding to approximately 0.05% H_2S , which was maintained until 75 minutes elapsed time. Thereafter, the H_2S concentration increased rapidly to form the traditional breakthrough curve. Approximately 35 minutes were required for the concentration to increase from 0.1% H_2S to 0.85% H_2S , and a steady-state concentration of 0.95% H_2S was approached after a total elapsed time of 130 minutes. No non-reacting tracer test was made at these reaction conditions but the shape and timing of the breakthrough curve suggest that the sulfur material balance closure should be comparable to other CeO_2 sulfidation runs.

The H_2S breakthrough curve for run Ce-08s1 is shown in Figure 2. Sulfidation conditions and H_2S breakthrough results were similar to those of Ce-06s1. After 20 minutes the H_2S concentration increased to a plateau range of 0.03% to 0.08% where it remained until 75 minutes elapsed time. Rapid breakthrough then began, and 25 minutes were required for the concentration to increase from 0.1% to 0.9% H_2S . A steady-state H_2S concentration of 0.99% H_2S was reached after 145 minutes. Results of a non-reacting tracer test at these conditions are also shown in Figure 2. The shaded area between the two curves is proportional to the quantity of H_2S removed by reaction with CeO_2 . Numerical integration of this area showed that the quantity of sulfur removed corresponded to 102% of the stoichiometric sulfur associated with complete conversion of CeO_2 to $\text{Ce}_2\text{O}_2\text{S}$ via reaction (1).

The H_2S breakthrough curve for run Ce-09s1 as well as a non-reacting tracer test at the same conditions are shown in Figure 3. The familiar H_2S concentration plateau of about 0.08% H_2S was followed after 75 minutes by rapid breakthrough, and a steady-state of 0.98% H_2S was approached after 120 minutes. Numerical integration of the area between the curves corresponded to sulfur removal of 107% of stoichiometric.

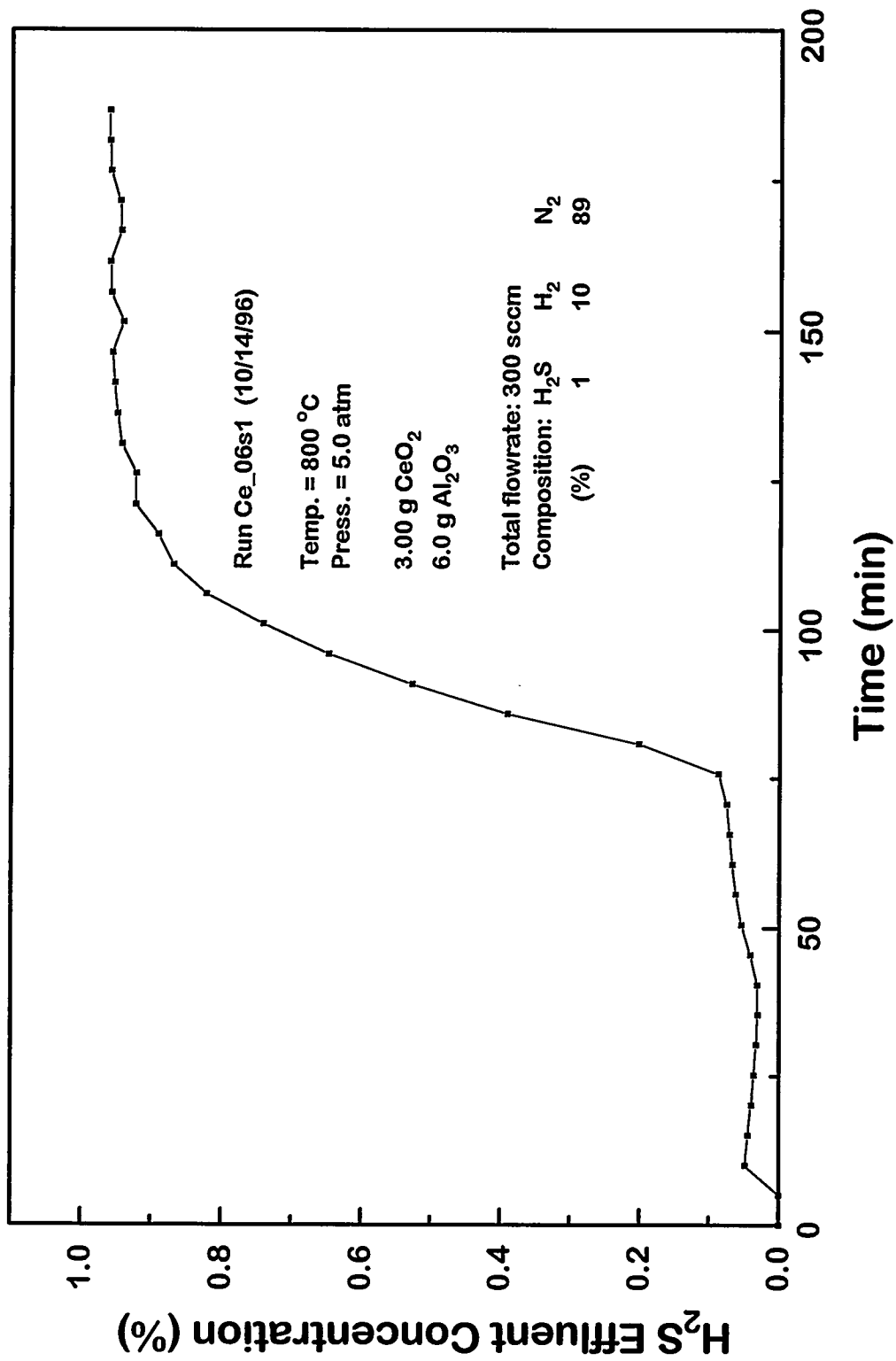


Figure 1. Sulfidation Breakthrough Curve: Run Ce-06s1

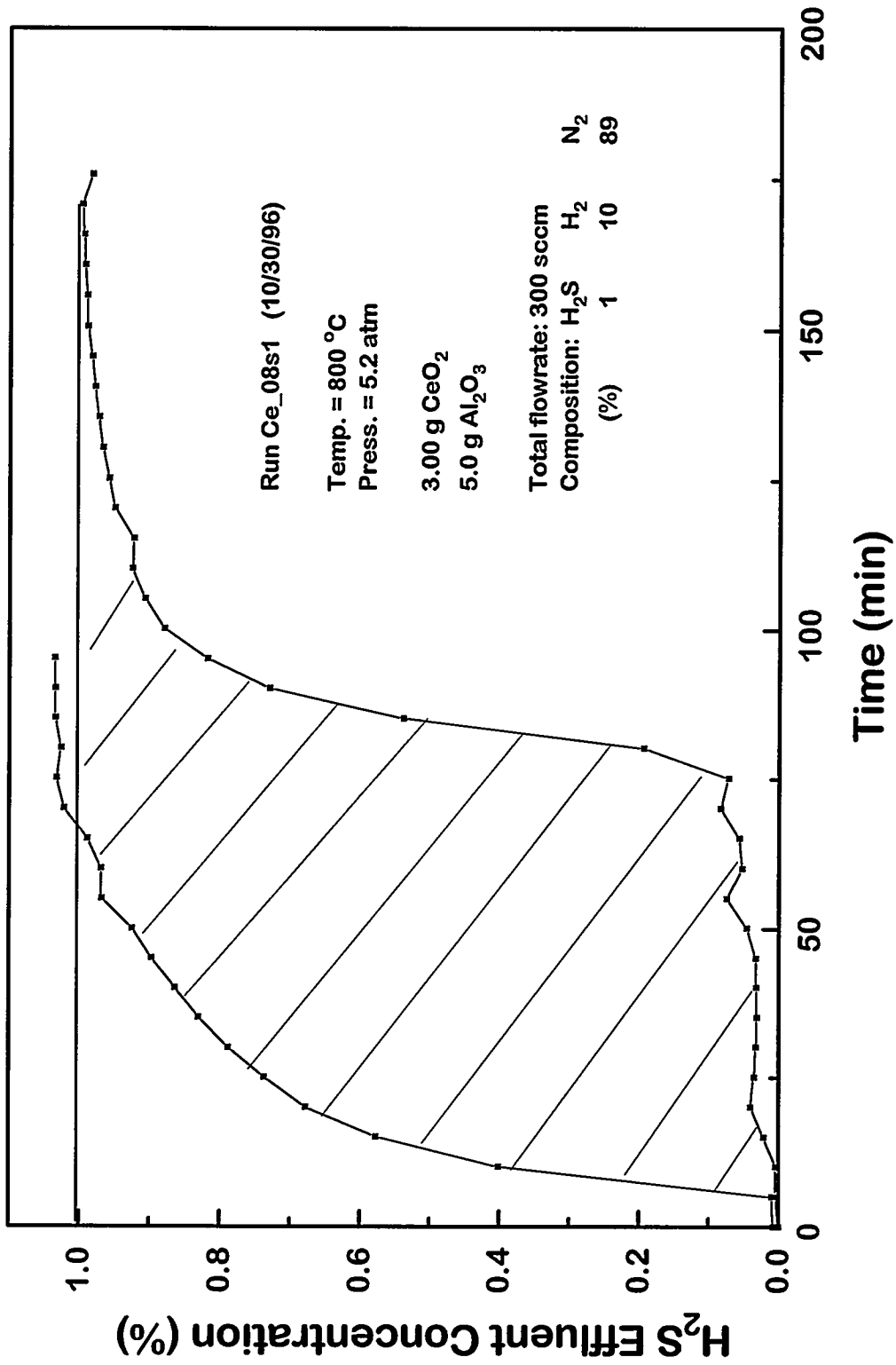


Figure 2. Sulfidation Breakthrough Curve: Run-08s1

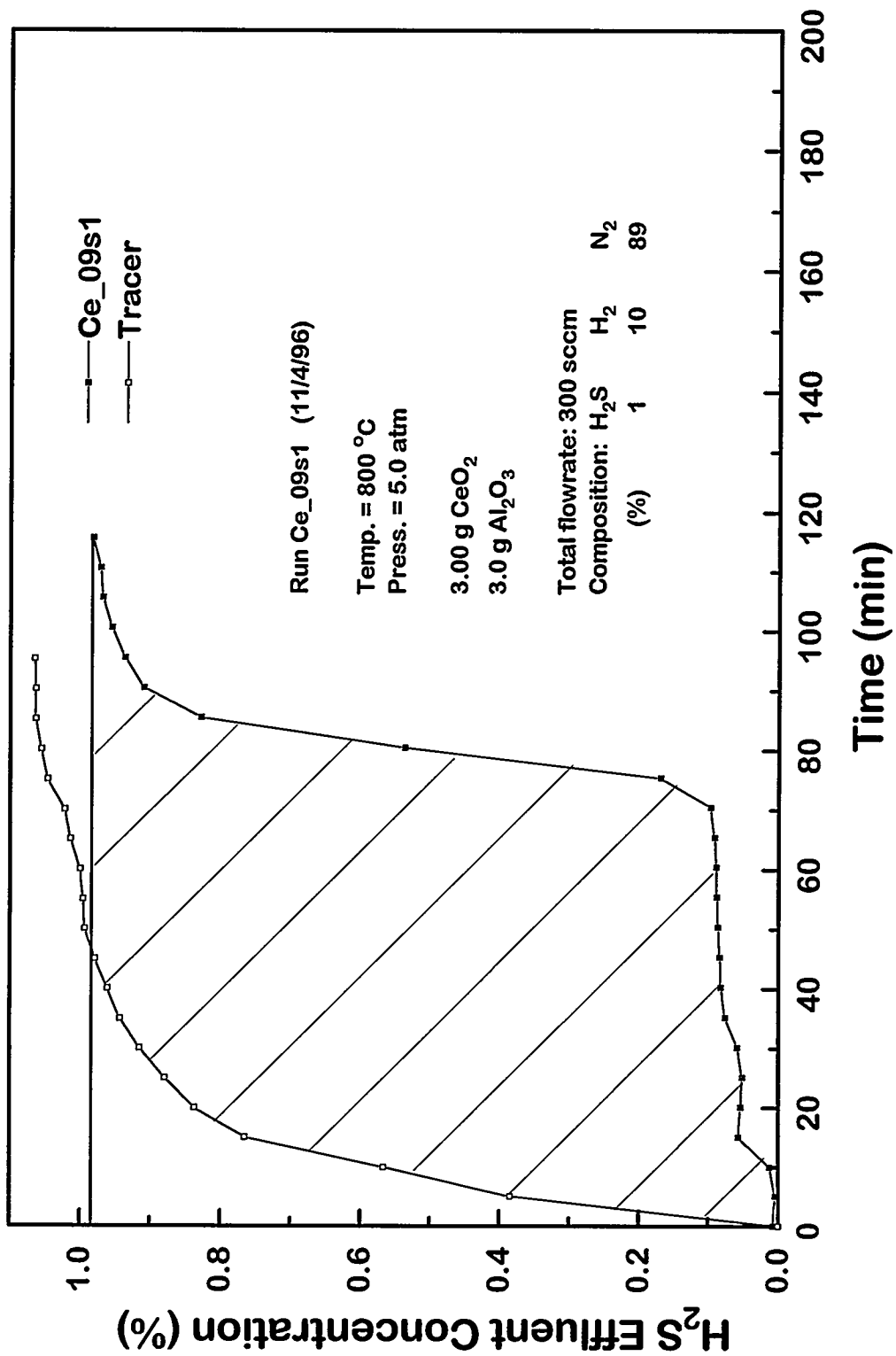


Figure 3. Sulfidation Breakthrough Curve: Run Ce-09s1

Sulfidation conditions in runs Ce-08s1 and Ce-09s1 were the same except that the particle size of the CeO_2 formed by the tableting procedure was larger and less Al_2O_3 was used in run Ce-09s1. The amount of CeO_2 was the same in both tests. The similarity of the H_2S breakthrough curves in terms of prebreakthrough plateau concentration, the time corresponding to the beginning of breakthrough, and the slope of active portion of the breakthrough curve show that tableting had little effect on the system other than to reduce bed pressure drop.

The effect of sulfidation temperature over the range of 700°C to 850°C was then investigated in a series of tests, Ce-10s1 through Ce-14s1. Resultant H_2S breakthrough curves are shown in Figure 4. There are several interesting features shown in this figure. First, although the H_2S plateau was present at each sulfidation temperature, the plateau concentration decreased as the sulfidation temperature increased. Active breakthrough began after about 60 minutes at the lower temperatures of 700 and 750°C , and after 70 to 75 minutes at 800 and 850°C . The slopes of the active portions of the breakthrough curves were approximately equal at 750 , 800 , and 850°C , but the slope was considerably smaller at 700°C . Similarly, the postbreakthrough steady-state H_2S concentration varied with temperature, ranging from 1.08% at 750°C to 1.06% at 700°C , 0.98% at 800°C , and 0.83% at 850°C . The first two values were somewhat larger than previously observed, but they, along with the 800°C result, were judged to be reasonably consistent with the error associated with the mass flow controller and analytical system. However, the 0.83% H_2S at 850°C was outside the range of acceptable error. For this reason, test Ce-13s1 was repeated in test Ce-14s1 (not shown) with effectively identical results. Thus, we must accept the reduced postbreakthrough concentration at 850°C as real. One possible explanation is the increasing importance of H_2S reaction with the stainless steel pressure vessel at this high temperature. Substitution of the quartz reactor insert for the stainless steel insert minimized, but did not eliminate, contact between hot steel surface and product gas.

The presence of the H_2S concentration plateau was not initially a source of major concern. Production of elemental sulfur during regeneration was the primary objective of the study and sulfidation tests were carried out only because $\text{Ce}_2\text{O}_2\text{S}$ could not be obtained commercially. Sulfidation was a necessary prelude to regeneration experiments. At first, we believed that the H_2S plateau was due to incomplete mixing of CeO_2 and Al_2O_3 , which allowed some H_2S to channel through the bed and avoid contact with CeO_2 . This explanation was unsatisfactory, however, because little or no change in the plateau level occurred as measures to insure more complete mixing were undertaken. Another explanation considered was that leakage of a portion of the feed gas around the O-ring seals of the reactor insert permitted some gas to totally bypass the packed bed. This explanation was also unsatisfactory in that no measurable change in the plateau concentration resulted when the bed pressure drop was reduced from about 40 psi to 5 psi upon adoption of the CeO_2 tableting procedure.

The current explanation is that the H_2S plateau is caused by reaction between gas phase H_2 and solid elemental sulfur deposited in cooler sections of the reactor and downstream tubing during regeneration tests. Efforts to prevent condensation of sulfur before it reaches the condenser are apparently only partially successful. Evidence in support of this explanation is

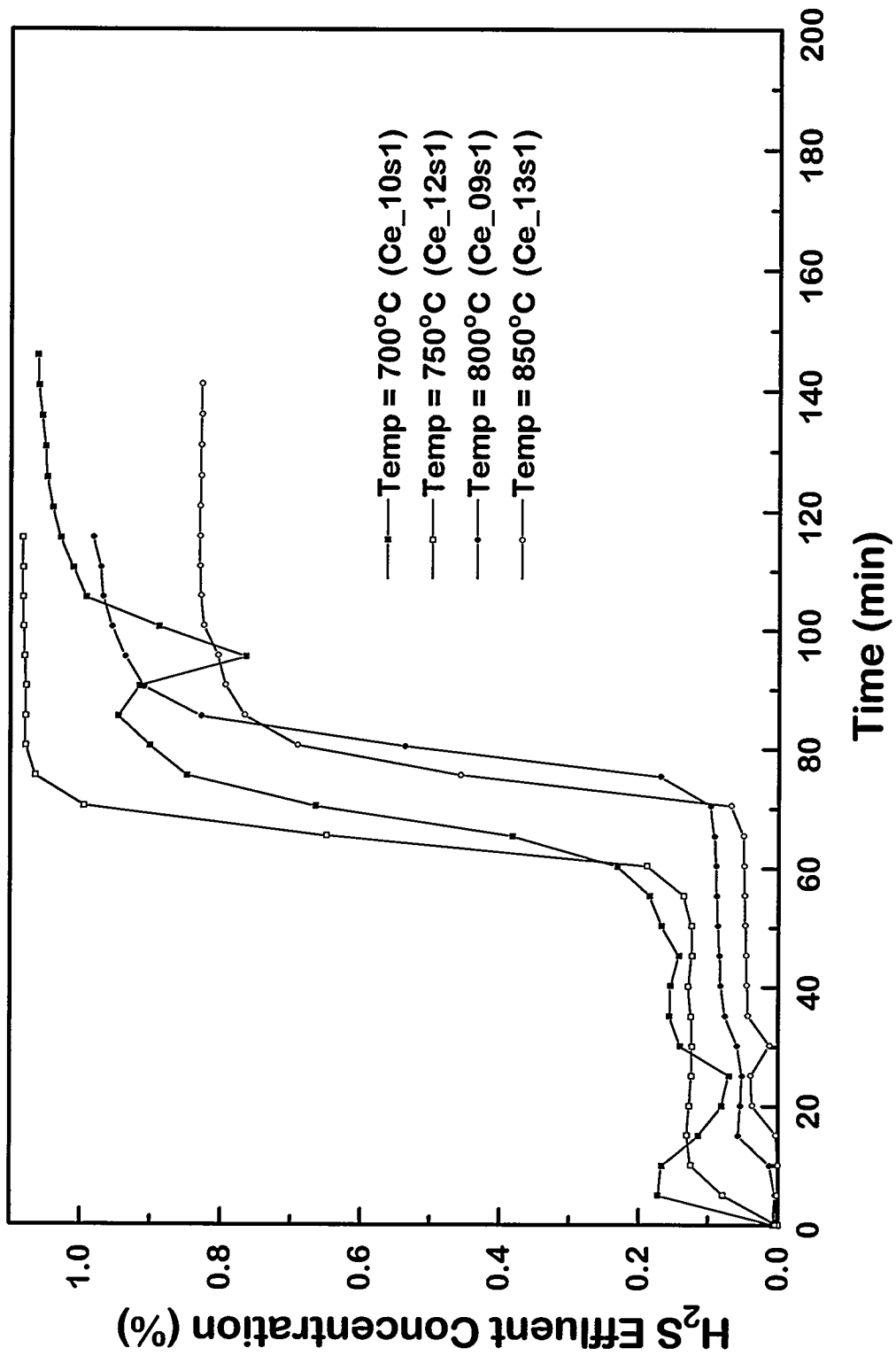


Figure 4. Sulfidation Breakthrough Curves as a Function of Sulfidation Temperature

shown in Figure 5. Following a SO₂ regeneration test, the reactor and downstream tubing were heated to 800 and 350°C, respectively, and a mixture of 10% H₂, balance N₂ was fed to the reactor. The total flow of 150 sccm provided greater residence time for the H₂-S reaction to occur. Quite large H₂S concentrations were measured in the product gas as shown in Figure 5. An initial surge of about 0.5% H₂S was followed by a gradual decrease to a relatively constant 0.2% level. After about 160 minutes the temperature of the downstream tubing was reduced to 25°C and the gas residence time decreased by increasing the flow rate to 400 sccm. The H₂S content quickly dropped to about 0.01%. Chemically cleaning the reactor and downstream tubing as well as reducing the tubing temperature to 25°C has been adopted as standard procedure in all subsequent sulfidation tests. The tubing is now heated to 350°C only during regeneration and cleaning periods.

The H₂S breakthrough curve for run Ce-16s3, the first sulfidation test following the new cleaning procedure, is shown in Figure 6. H₂S concentration during early portions of the reaction is much smaller. The 0.01% H₂S during the first 20 minutes of the test corresponds to about 99% H₂S removal, and the H₂S removal remained above 95% for 80 minutes. It is also significant that the gradual increase in H₂S concentration during the prebreakthrough period is consistent with the expected kinetics of the gas-solid reaction. Other portions of the breakthrough curve, including the time corresponding to the beginning of active breakthrough and the slope of the active portion of the breakthrough curve are similar to the results from other sulfidation tests.

Two additional features associated with the results of Ce-16s3 are worthy of mention. The first two sulfidation cycles, Ce-16s1 and Ce-16s2, were carried out before the cleaning procedure was adopted, and the H₂S plateau was present in each case. These test results are discussed in greater detail in the multicycle section of this report. It is also important to remember that the H₂S analysis was performed using a thermal conductivity detector, and 0.01% H₂S (~ 100 ppmv) corresponds approximately to the lower limit of the detector.

Regeneration Results

The first Ce₂O₃S regeneration run attempted was Ce-04r1 which followed sulfidation run Ce-04s1. With the exception of larger than desired bed pressure drop the sulfidation phase was successful; the sulfur material balance, as shown in Table 1, was a perfect 100% of stoichiometric. The regeneration feed gas in Ce-04r1 contained 1% SO₂ in N₂ at a temperature of 350°C. Little, if any, reaction occurred at this low temperature; therefore, toward the end of the test the regeneration temperature was increased. Reaction between Ce₂O₃S and SO₂ began at a temperature of about 550°C.

The next regeneration run, Ce-06r1, was carried out at 600°C. Reaction was rapid and the SO₂ breakthrough curve is shown in Figure 7. No SO₂ was present in the product gas for the first 60 minutes. SO₂ breakthrough then began and the concentration reached 0.65% after 100 minutes when the SO₂ mass flow controller malfunctioned and the run was terminated.

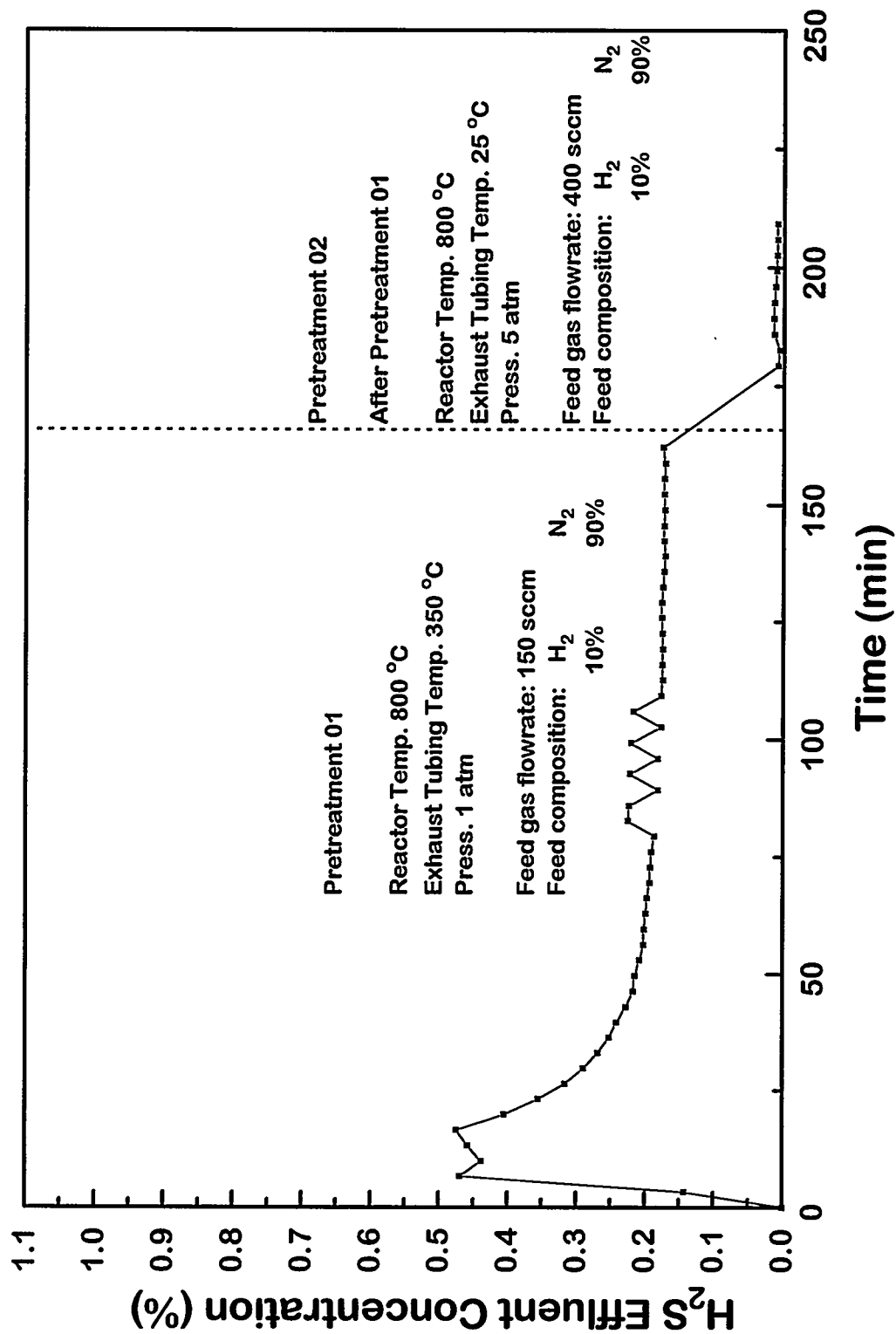


Figure 5. H₂S Formed by the Reaction Between H₂ and Elemental Sulfur

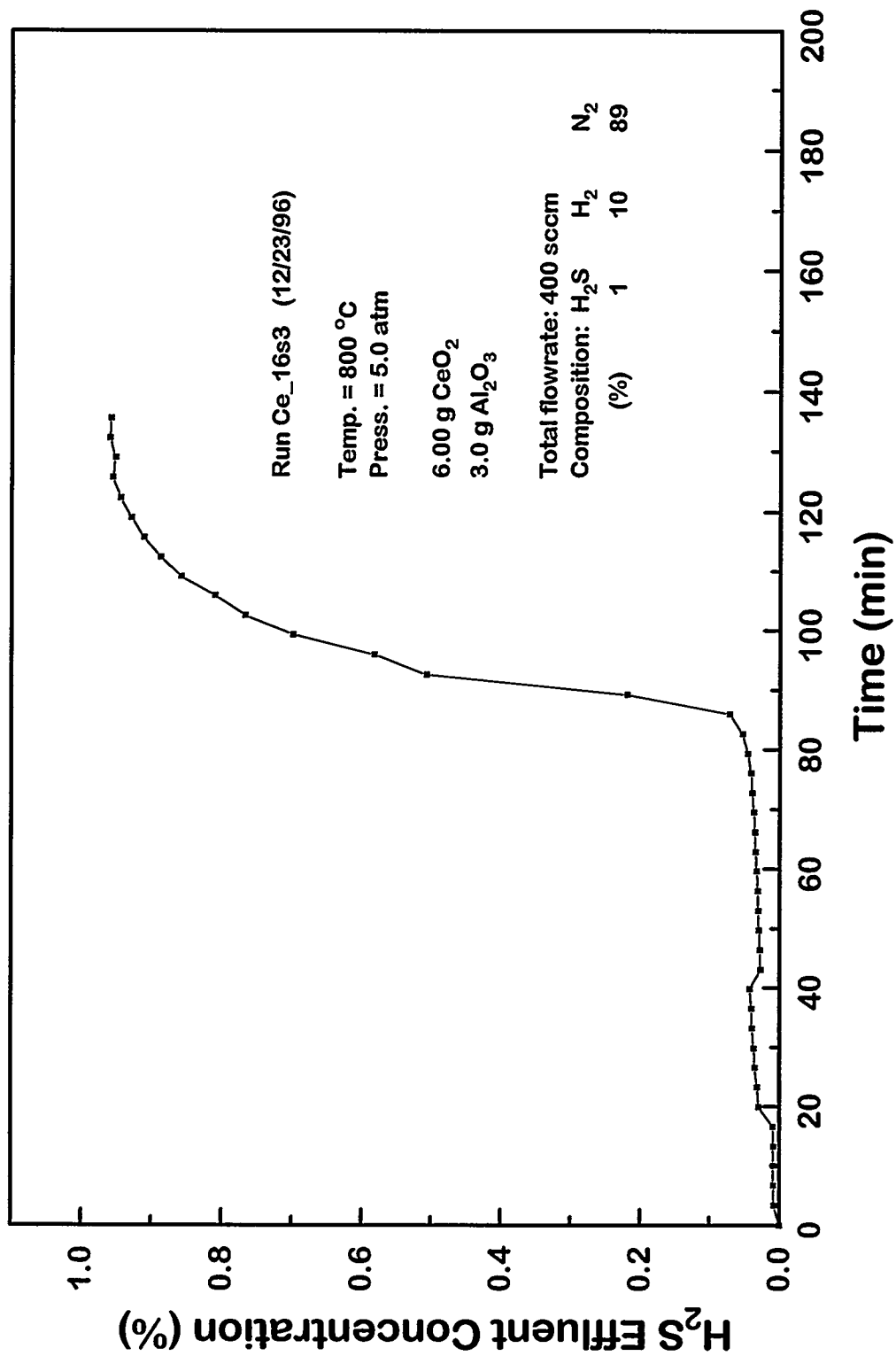


Figure 6. Sulfidation Breakthrough Curve: Run Ce-16s3

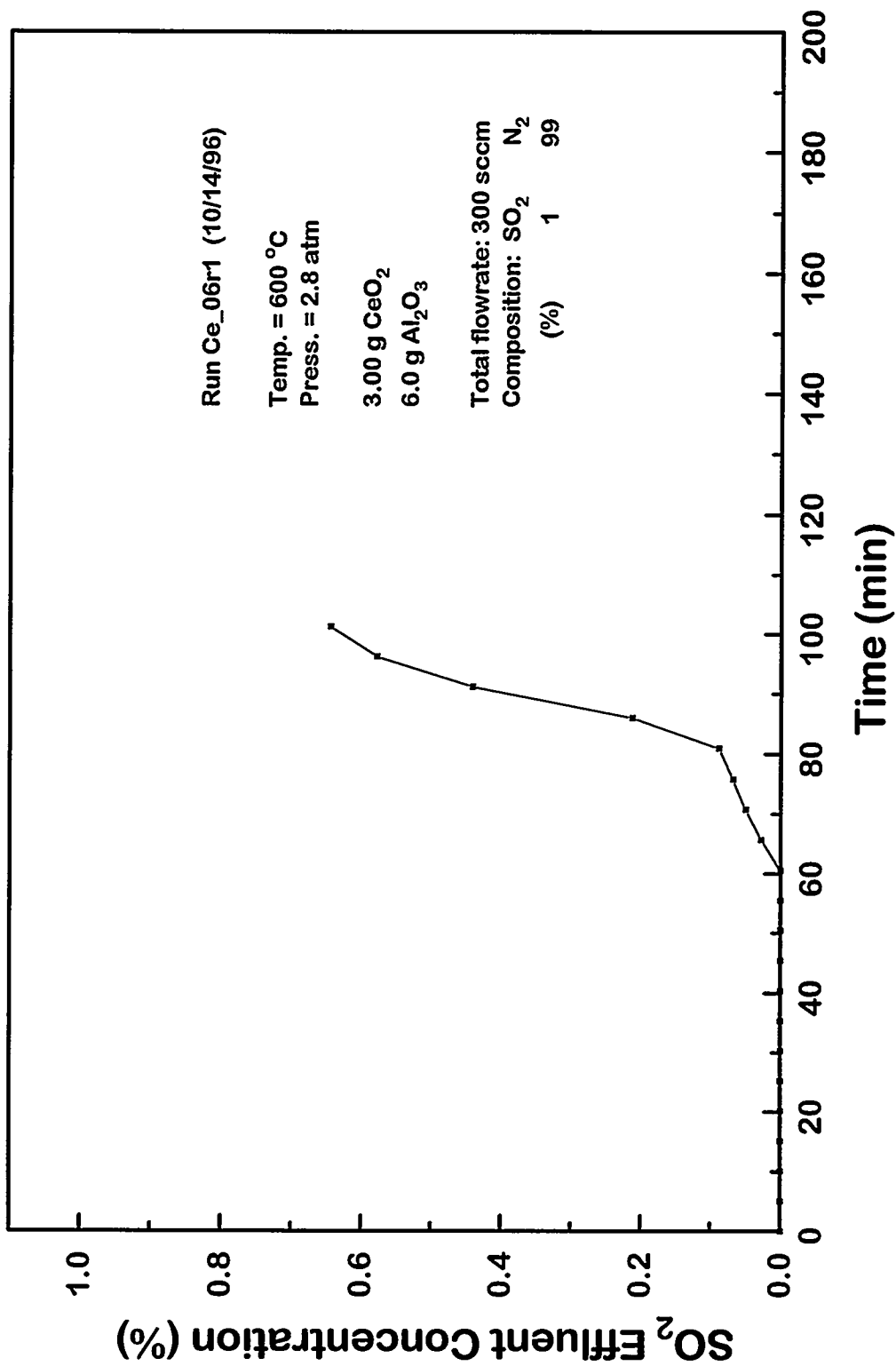


Figure 7. Regeneration Breakthrough Curve: Run Ce-06r1

However, the results to that point were encouraging. Zero SO₂ in the product gas corresponds to complete SO₂ conversion with the production of 1% elemental sulfur in the product gas. The breakthrough curve was quite steep to the point that the run was terminated.

Regeneration run Ce-08r1 also had to be terminated before completion, this time due to elemental sulfur plugging the entrance to the condenser. Results prior to termination were again favorable. Various changes were made downstream of the reactor in an attempt to prevent plugging and minimize sulfur deposition. Later test results indicate that plugging has been effectively eliminated but that some sulfur deposition is still occurring.

The first complete regeneration test was Ce-09r1 which again used 600°C temperature and 1% SO₂ in N₂. The SO₂ breakthrough curve along with a nonreacting SO₂ tracer test result at the same conditions are shown in Figure 8. SO₂ concentration in the reaction product was zero for the first 45 minutes, then increased slowly to 0.06% at 65 minutes, followed by rapid breakthrough thereafter. A temporary system upset occurred in the 70 to 80 minute period which was responsible for the unusual shape of the breakthrough curve during that time. Integration of the area between the breakthrough and tracer curves resulted in sulfur material balance closure of 107% of stoichiometric. Test Ce-09 was carried out with favorable results through two complete sulfidation-regeneration cycles. The multicycle aspects are discussed later in this report.

In order for the SO₂ regeneration concept to be attractive commercially, it will be necessary to produce higher concentrations of elemental sulfur in the product gas. This can be achieved by increasing the SO₂ concentration in the regeneration feed gas. The regeneration cycles of run Ce-15 (cycles 1 through 3) examined the reactor response to successively larger SO₂ contents in the feed gas (2%, 4% and 8%). Standard sulfidation conditions were used in each cycle. SO₂ breakthrough curves for the three regeneration cycles of run Ce-15 are shown in Figure 9. Results from regeneration test Ce-09r2 using 1% SO₂ are also included for comparison purposes. The y-axis in Figure 9 is the normalized SO₂ concentration, i.e., the ratio of the product and feed concentrations. Thus, all breakthrough curves in Figure 9 should approach a normalized concentration value of 1.0, which is indeed the case. The beginning of the active breakthrough period decreased from about 60 minutes using 1% SO₂ to 30 minutes with 2% SO₂, 15 minutes with 4% SO₂, and to between 5 and 10 minutes using 8% SO₂. The elemental sulfur concentration approached 8% for a short period of time in run Ce-15r3.

Note that run Ce-15r3 was effectively complete in 20 minutes, and only four product gas samples were obtained during the run. Two showed zero SO₂ concentration while the third and fourth both gave normalized concentration near 1.0. Reaction and analytical conditions were changed before the next regeneration test, which used still larger SO₂ feed gas concentrations, to extend the duration of the run and to provide more frequent product gas sampling.

Prior to run Ce-16 the chromatograph operating conditions were changed so that the sampling interval was reduced from 5 to 3.3 minutes. In addition, the amount of CeO₂ used in run Ce-16 was doubled and the regeneration gas feed rate was reduced from 300 to 200 sccm.

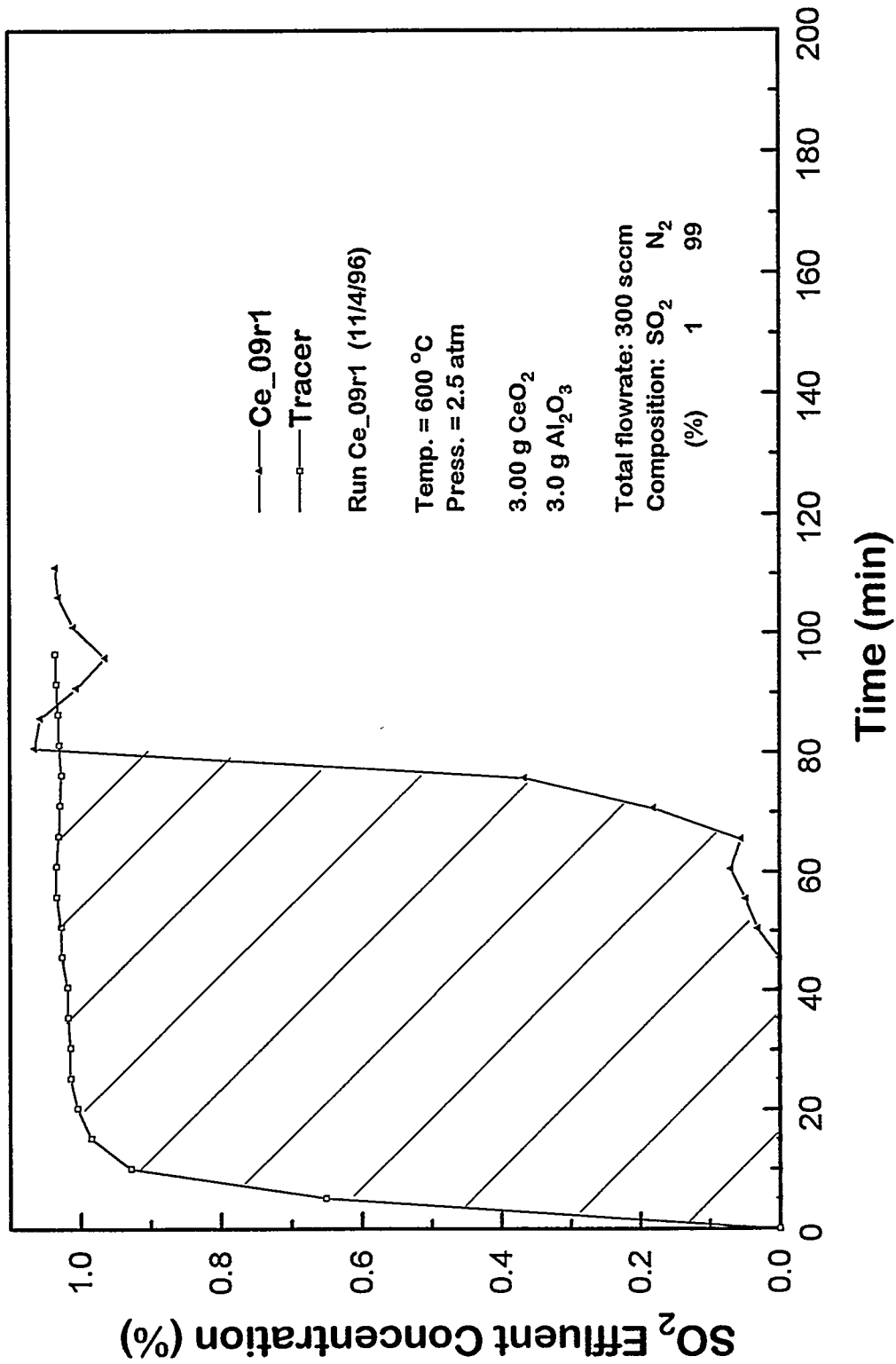


Figure 8. Regeneration Breakthrough Curve: Run Ce-09r1

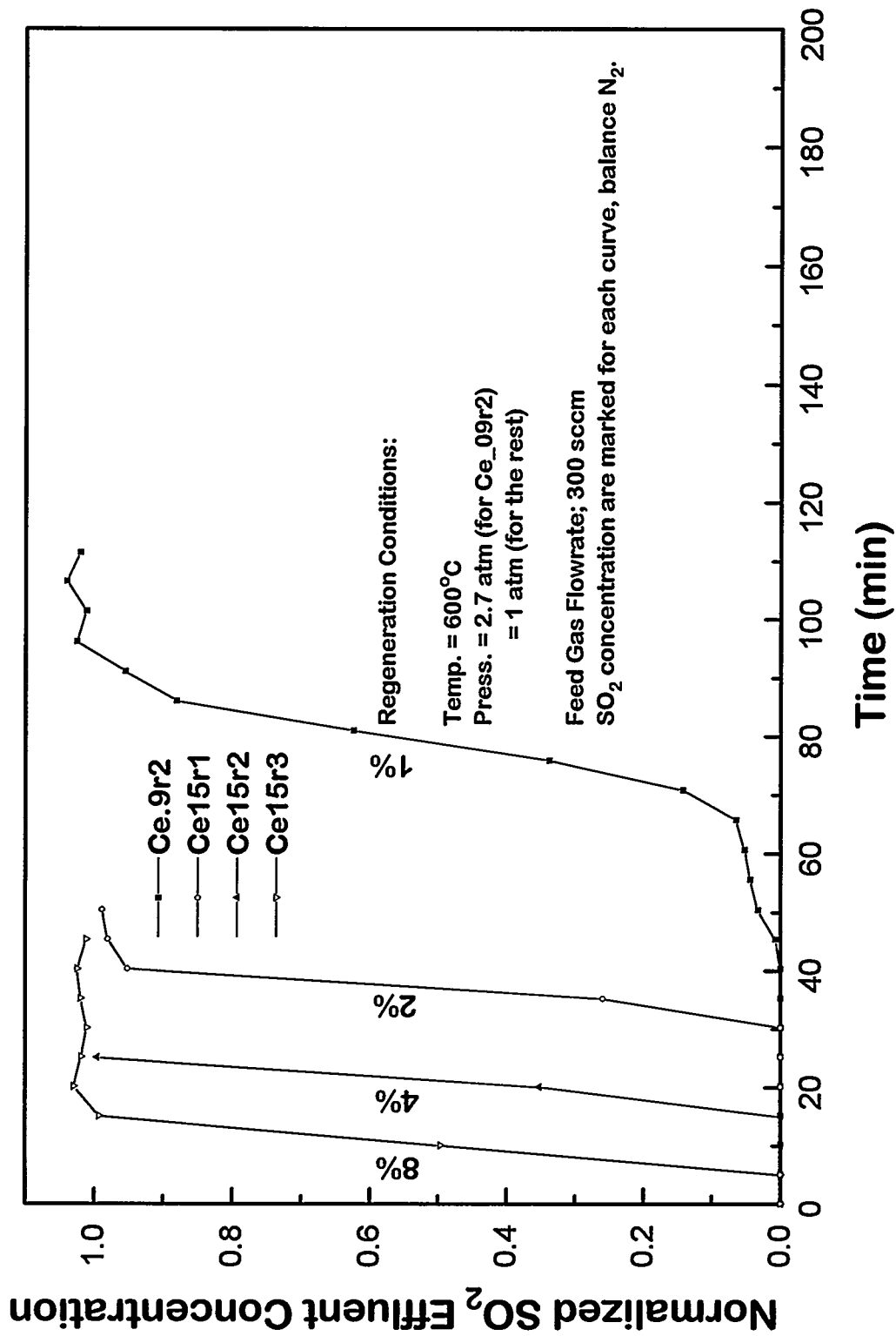


Figure 9. The Effect of SO₂ Concentration on Regeneration Breakthrough Curves

The SO₂ content of the regeneration gas was increased to 12% and the SO₂ breakthrough curve from run Ce-16r1 is shown in Figure 10. SO₂ concentration in the product gas was zero in the first three samples (~7 min), increased to about 2% over the next three samples (~17 min), and approached 12% in the seventh sample. Regeneration was again complete in about 20 minutes and the SO₂ content of the product gas approached 12% during a significant portion of that time. Preliminary results from the process modeling work (discussed in a later section of this report) suggest that the optimum sulfur content of the regeneration product gas is about 15%. Thus, operating conditions during a portion of Ce-16r1 were reasonably close to the target. Run Ce-16 was continued through three complete sulfidation and regeneration cycles; the multicycle aspects of this test are discussed in the following section.

Multicycle Results

The first successful multicycle test was Ce-09 which consisted of two complete sulfidation and regeneration cycles. H₂S breakthrough curves for the sulfidation cycles are presented in Figure 11 along with results of a nonreacting H₂S tracer test at the same conditions. Both sulfidation tests showed the familiar H₂S concentration plateau in the 10 to 70 minute time period followed by rapid H₂S breakthrough from 70 to 100 minutes. With the exception of the slightly higher H₂S plateau in cycle 2, the breakthrough curves were effectively identical. The cumulative H₂S removal was 110% and 104% of stoichiometric in cycles 1 and 2, respectively. The near stoichiometric H₂S removal in the second cycle also provides independent confirmation that complete Ce₂O₂S regeneration was achieved in the first regeneration cycle.

Results of the two regeneration cycles, Ce-09r1 and Ce-09r2, also with nonreacting SO₂ tracer tests at the same conditions are shown in Figure 12. The SO₂ concentration was effectively zero for the first 45 minutes of both cycles; active breakthrough occurred in the 45 to 90 minute time span, and the SO₂ product concentration was effectively equal to the 1.0% SO₂ concentration of the feed after 90 minutes. A temporary system upset occurred in cycle 1 in the 70 to 80 minute period which is responsible for the unusual shape of the SO₂ concentration curve during that time. The area between the curves produced sulfur material balance closure of 107% and 112% of stoichiometric in the two cycles.

Run Ce-15 was continued through three complete cycles. H₂S breakthrough curves for the three sulfidation cycles are presented in Figure 13. In addition to the extra cycle, there were two other differences in the conditions of Ce-09 and Ce-15. The CeO₂ tableting procedure for pressure drop control was adopted prior to Ce-15, and the three Ce-15 regeneration cycles used successively larger concentrations of SO₂ in the feed gas -- 2% SO₂ in Ce-15r1, 4% SO₂ in Ce-15r2, and 8% SO₂ in Ce-15r3. Results of the regeneration cycles have already been presented in Figure 9. The sulfidation breakthrough curves are similar to those previously observed except for the unexplained larger final H₂S concentration in Ce-15s2, and the initial peak in H₂S concentration at about 10 minutes and the delayed breakthrough time in Ce-15s3. The plateau H₂S concentrations were approximately equal in each cycle as were the slopes of the breakthrough. No nonreacting tracer test was made so that no sulfur material balance is reported

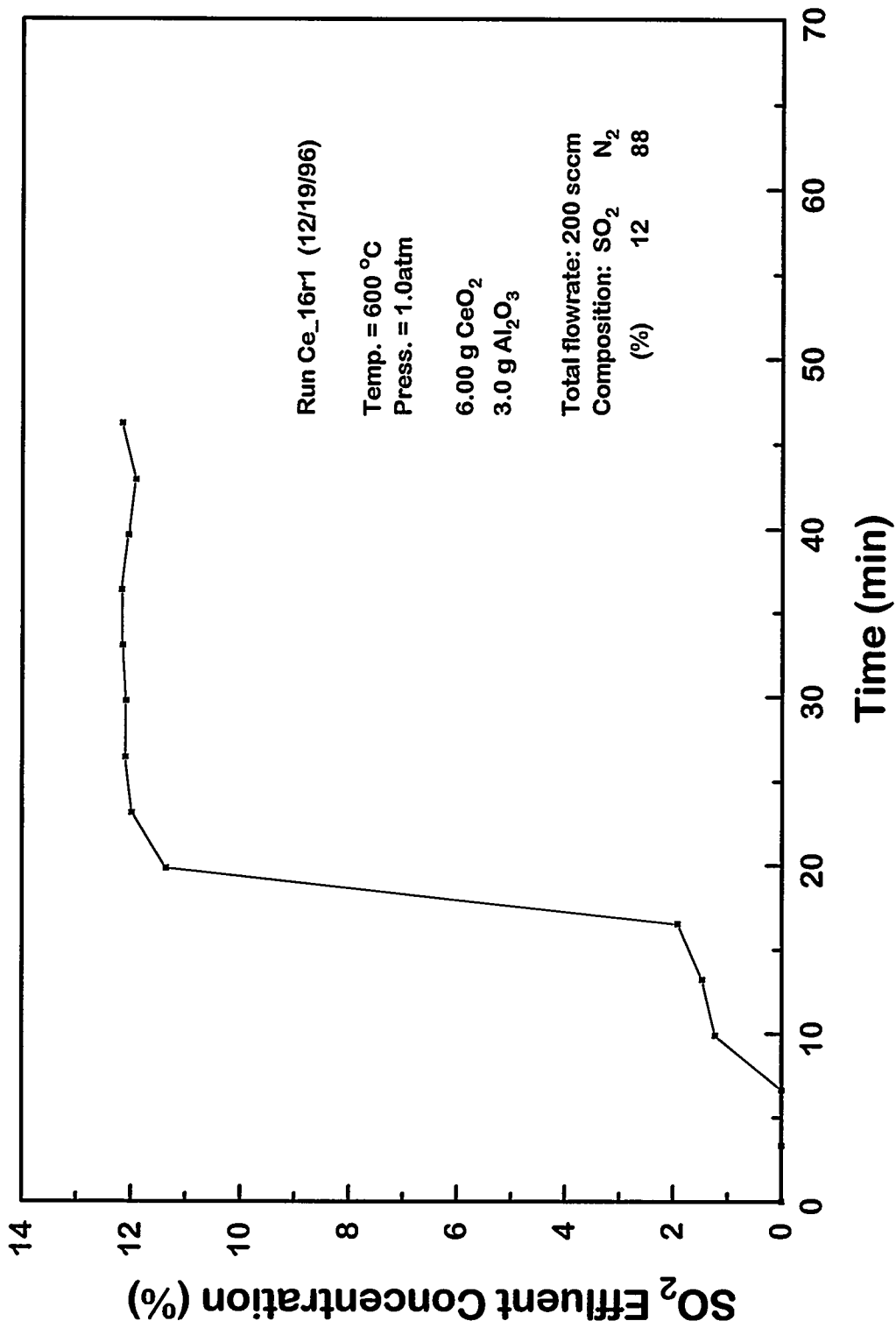


Figure 10. Regeneration Breakthrough Curve: Run Ce-16r1

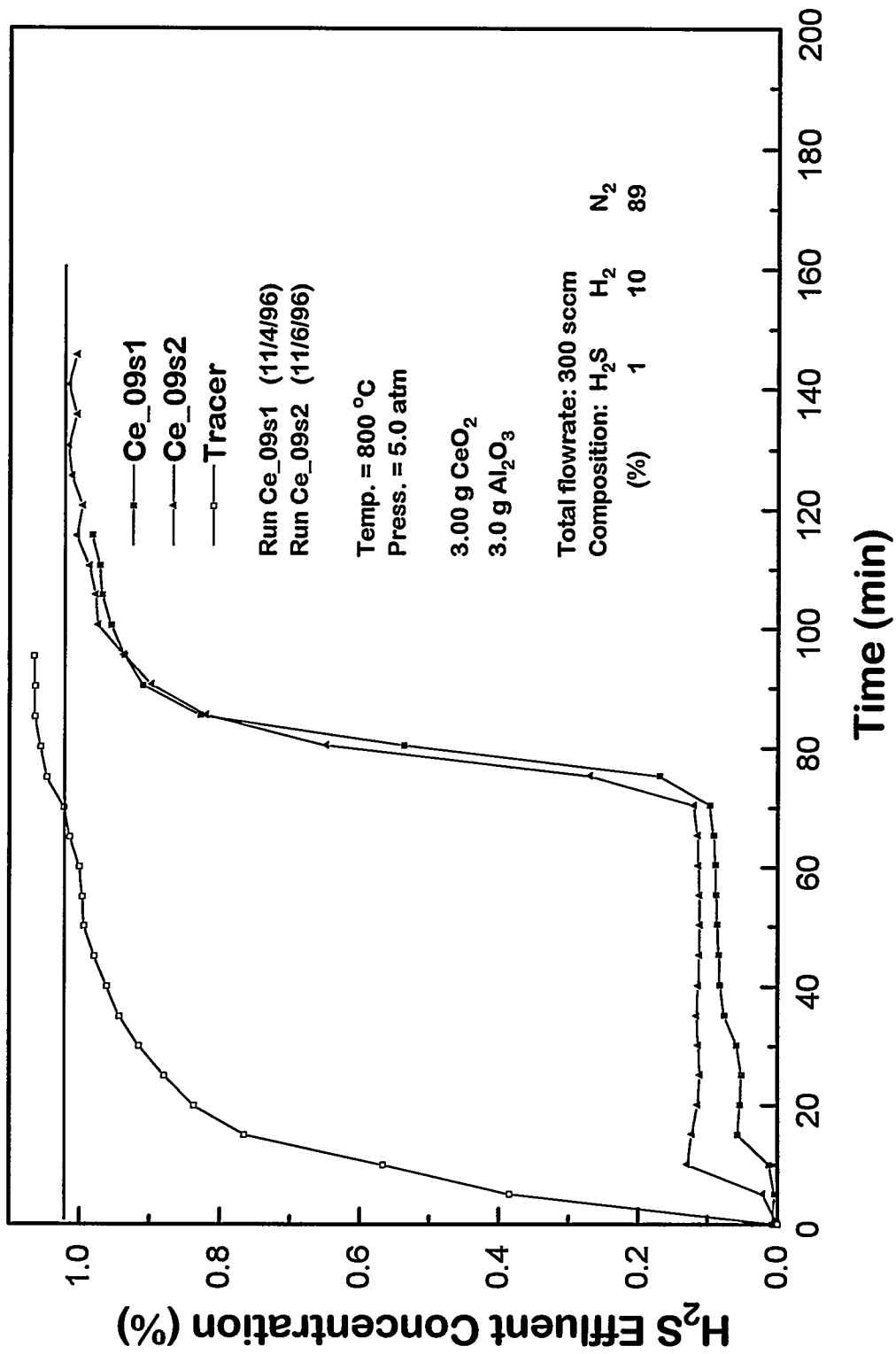


Figure 11. Sulfidation Breakthrough Curves for the Two Cycles of Run Ce-09

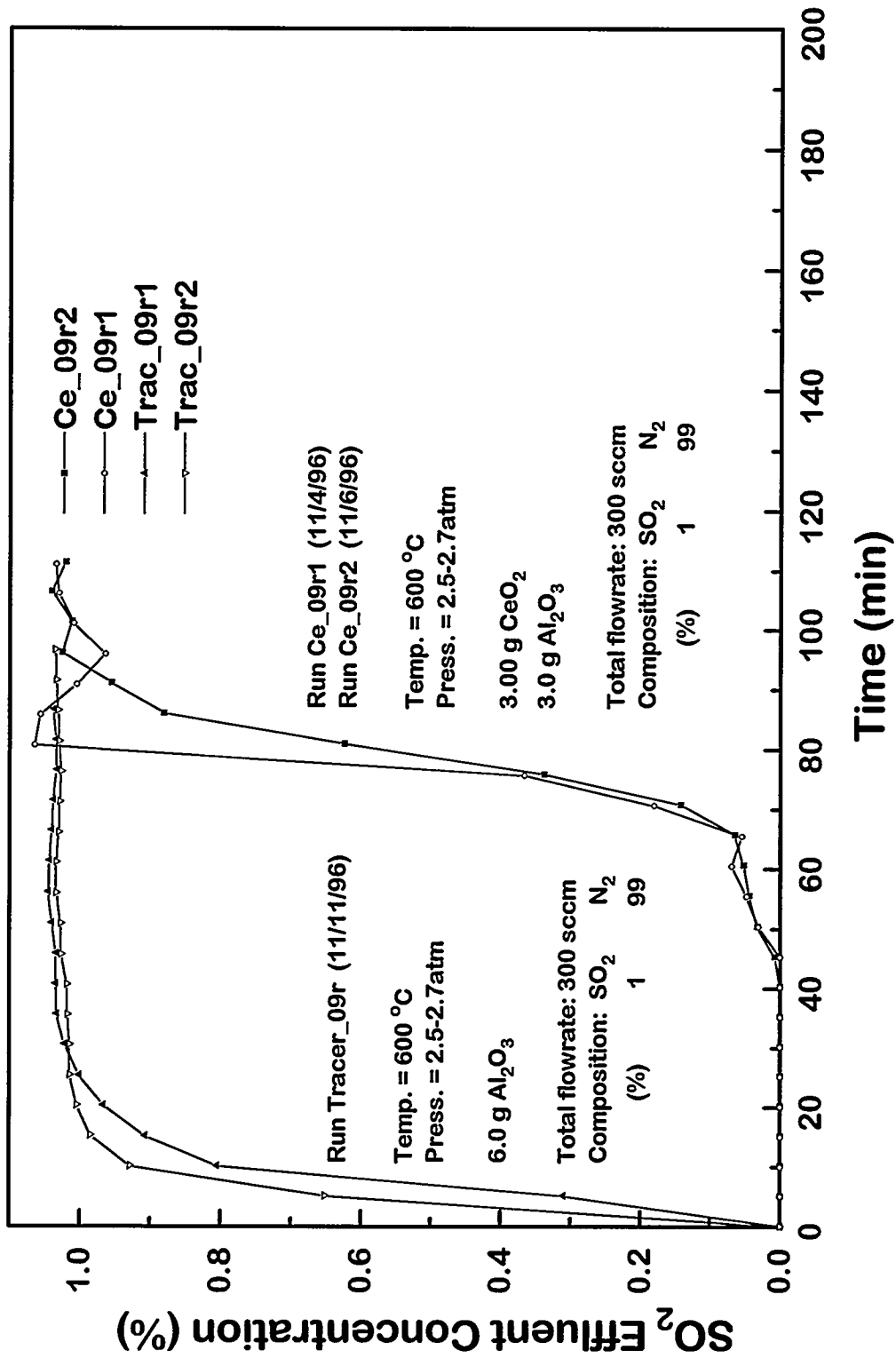


Figure 12. Regeneration Breakthrough Curves for the Two Cycles of Run Ce-09

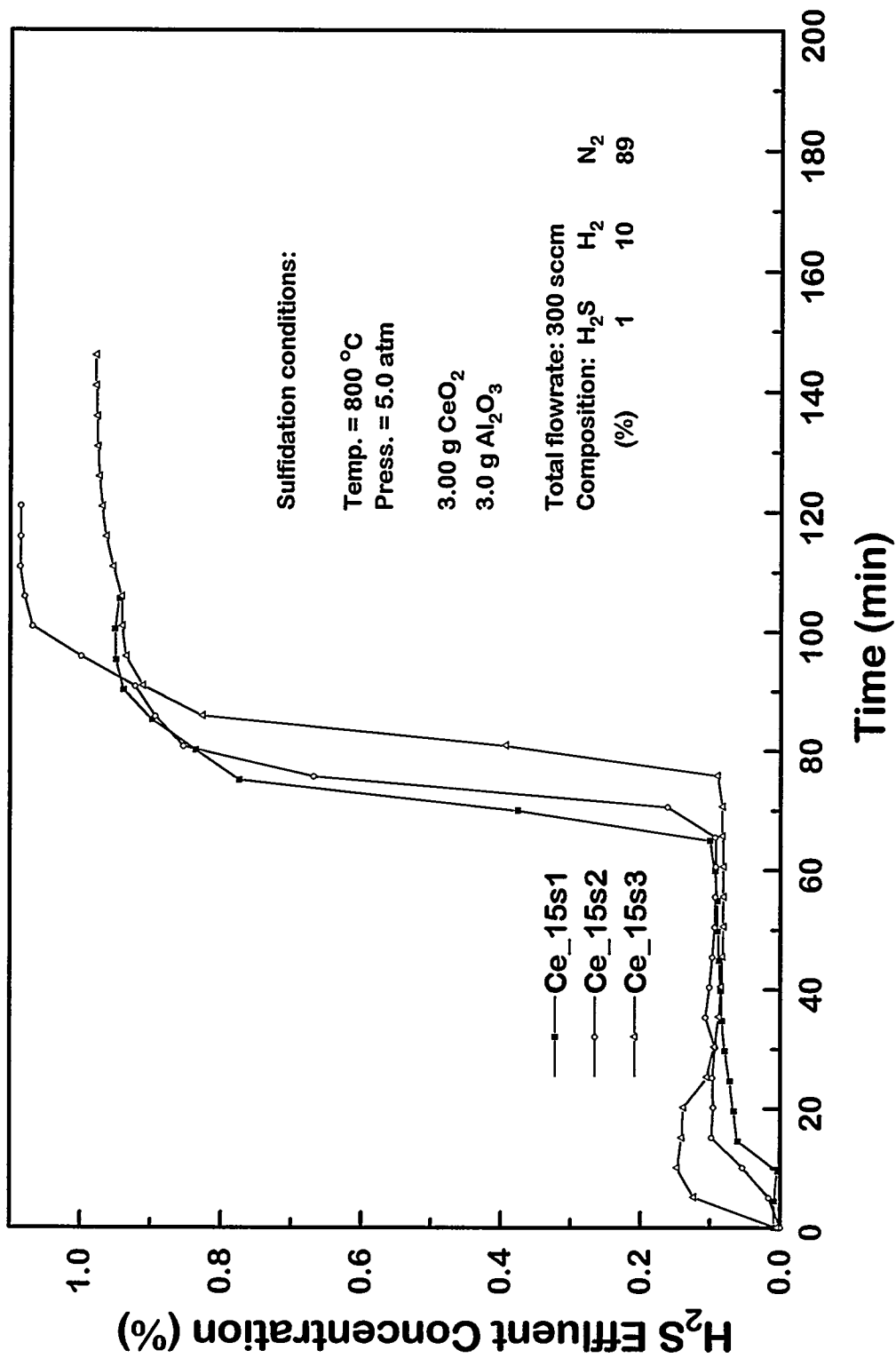


Figure 13. Sulfidation Breakthrough Curves for the Three Cycles of Run Ce-15

for the tests. However, the breakthrough curves show no sign of sorbent deterioration. If anything, the delayed breakthrough in Ce-15s3 suggests improved sorbent performance in the third cycle.

Three cycles of test Ce-16, which is planned for ten-cycles, were completed by the end of the quarter. The previously described adjustments in chromatograph and reaction conditions were made in this run to permit the SO₂ content of the regeneration feed gas to be increased to 12%. Standard sulfidation conditions were used in each cycle except that the sulfidation feed gas rate was mistakenly set at 200 sccm (instead of 400 sccm) in Ce-16s1. The duration of the first sulfidation cycle was considerably longer because of the reduced flow rate. In addition, the cleaning procedure designed to remove elemental sulfur from the bottom of the reactor and downstream tubing was instituted between Ce-16r2 and Ce-16s3. As a result, the H₂S plateau was greatly reduced in Ce-16s3 as was shown in Figure 6.

The SO₂ breakthrough curves for the three regeneration cycles of Ce-16 are shown in Figure 14. The SO₂ content of the feed gas was 12% in each cycle, and the breakthrough curves were, with the exception of a single sample at about 16 minutes in cycle 3, essentially identical. No SO₂ was detected in the first three samples. The SO₂ content increased gradually to about 2% during the next three samples, and then increased rapidly to approach 12% by the seventh sample. Regeneration was effectively complete in 20 minutes with no apparent deterioration in performance over the three cycles. The elemental sulfur content of the product gas was approximately 12% during the time that the SO₂ concentration was zero.

PROCESS MODELING

At the end of the previous quarter, the process modeling work had progressed to the point that basic flow diagrams had been synthesized for the following cases:

1. Two-Stage Desulfurization With Steam Regeneration: Bulk sulfur removal is accomplished in a primary sorber by reacting H₂S with CeO₂. Final sulfur removal to meet coal gas specifications is accomplished in a secondary sorber using the H₂S-Zn₂TiO₄ reaction. Ce₂O₂S regeneration occurs in the primary regenerator by reaction with steam, and the liberated H₂S is converted to elemental sulfur in a Claus process. The sulfided zinc sorbent is regenerated by reaction with oxygen in the secondary regenerator and the off-gases are recycled to the gasifier.

2. Two-Stage Desulfurization With SO₂ Regeneration: Sulfur removal in the primary and secondary sorbers as well as the regeneration of sulfided zinc sorbent in the secondary regenerator are identical to the previous case. However, Ce₂O₂S is regenerated by reaction with SO₂ in the primary regenerator to form elemental sulfur directly.

3. Single-Stage Desulfurization With DSRP: Complete desulfurization is achieved in a single sorber using a zinc titanate sorbent. The sulfided sorbent is regenerated with air to

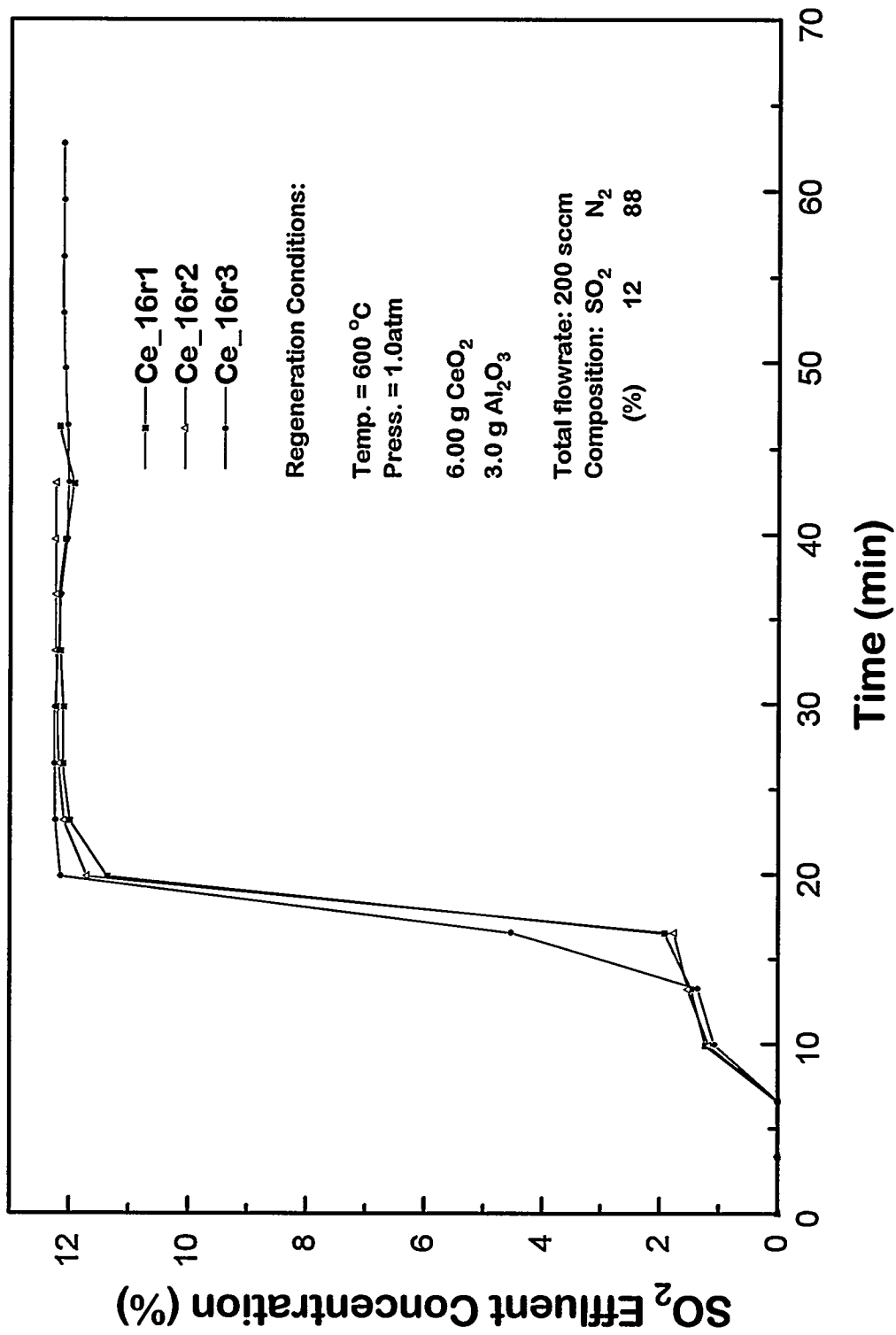


Figure 14. Regeneration Breakthrough Curves for the Three Cycles of Run Ce-16

form SO_2 which is subsequently reduced to elemental sulfur in the DSRP reactor by reaction with a slip stream of coal gas.

Results of twelve preliminary material and energy balance case studies for the two-stage desulfurization processes were presented in the previous quarterly report based upon a number of realistic, but sometimes arbitrary, operating conditions, such as the following:

i. The feed to the desulfurization process consisted of 17,000 lbmol/hr of Shell gas containing 1% H_2S at 800K and either 15 or 25 atm. The low temperature implied that the coal gas had been cooled for particulate removal prior to entering the desulfurization block.

ii. Primary and secondary sorber temperatures were taken as 800K, 900K, and 1000K at both 15 atm and 25 atm. These conditions specified the twelve case studies (2 processes x 3 temperatures x 2 pressures). With three exceptions, the temperatures of the regenerators were set equal to the sorber temperatures.

iii. H_2S removal in both sorbers was based on thermodynamic equilibrium at the specified temperature and pressure. No reactions other than desulfurization were allowed to occur.

iv. All reactors were assumed to operate isothermally at the specified temperature. Thus, the energy balance determined the quantity of heat which must be added or removed in each reactor to maintain the specified temperature.

v. Spent sorbent discharge and fresh sorbent make-up from the primary and secondary desulfurization steps were fixed at 1% of the respective sorbent circulation rate.

Other conditions and assumptions are itemized in the previous quarterly report. During the current quarter the scope of process conditions considered was narrowed. The operating pressure was fixed at 25 atm since the preliminary studies showed that pressure had little effect on material and energy balance results. In addition, cases involving 800K primary desulfurization temperature were dropped because only about 80% of the H_2S can be removed by reaction with CeO_2 at this temperature. Therefore, four of the twelve preliminary case studies were used in the refined case studies completed during the present quarter. In addition, temperatures of the secondary sorber and regenerator were changed to 873K and 1000K, in closer agreement with current operating practice. The temperature of the coal gas was increased from 800K to 1283K, which more closely approximates the actual temperature from a Shell gasifier. This change permitted us to better incorporate the sensible energy of the coal gas into the process energy balance. Finally, the restriction that the sorbers and regenerators operate isothermally was changed. Heat effects associated with the primary and secondary sorbers and secondary regenerator are relatively small. Flow rates and temperatures of the feed streams were adjusted so that product temperatures would be near the nominal temperatures under adiabatic operation. The primary regenerator operates nonadiabatically because the heat released by the reaction between $\text{Ce}_2\text{O}_2\text{S}$ and SO_2 is of such magnitude that adiabatic operation is not

feasible. Sufficient heat transfer capability is included so that the product temperature is near the nominal value.

Three additional cases studies were added to the four described above. One case involved single-stage desulfurization and regeneration with elemental sulfur recovery using the DSRP reaction. Reactor temperatures in this case were based on expected operating conditions reported by RTI (Gangwal et al., 1993). Both the sorber and regenerator operate adiabatically while the DSRP reactor is nonadiabatic with heat transfer capability included. Both of the other new cases involve two-stage desulfurization with SO₂ regeneration. Temperatures of the primary sorber (adiabatic) and regenerator (nonadiabatic) were changed to 1073K and 873K, respectively, to conform to present laboratory test temperatures.

The rate of SO₂ recycle to the primary regenerator determines the heat transfer requirements in that reactor. A small recycle rate results in large elemental sulfur concentrations in the regenerator product and relatively small loads on the sulfur condenser and SO₂ recycle compressor. However, the SO₂ recycle rate originally used was not consistent with the reactor heat transfer requirements. Therefore, in the second additional case study involving two-stage desulfurization with SO₂ regeneration, the recycle rate was increased to a level consistent with the heat transfer requirements of the primary regenerator.

In summary, seven case studies are discussed in the following. Two involve two-stage desulfurization with steam regenerator, four are based on two-stage desulfurization with SO₂ regeneration, and the seventh assumes single-stage desulfurization with elemental sulfur recovery using DSRP.

Two-Stage Desulfurization With Steam Regeneration

Separate process flow diagrams for the nominal 900K and 1000K primary sorber case studies are shown in Figures 15 and 16, respectively. The diagrams differ only in the heat transfer portions of the process. All process streams (but not necessarily flow rates) are the same as previously reported in the simplified cases. In the 900K case (Figure 15), three exchangers are used to remove heat from the cool gas prior to the primary sorber and one exchanger is used to cool the gas between the primary and secondary sorbers. Two exchangers are used both before and after the primary sorber in the 1000K case. Water and steam flows in Figures 15 and 16 are represented by dashed lines while process flows are indicated by solid lines. Note that the dashed steam line (stream 162) becomes two solid lines (streams 8 and 16) to indicate process steam fed to both the primary and secondary regenerators.

In both options, feed water (stream 207) is pumped to 1250 psia and used in a variety of condensers, reboilers, heat exchangers, and heat recovery steam generators. All water/steam flows enter or exit the steam drum which contains saturated liquid and vapor at 1250 psia and 572°F (573K). Excess steam, represented by stream 210, may be "sold" to supplement power generation, while a small bleed of liquid is represented by stream 213.

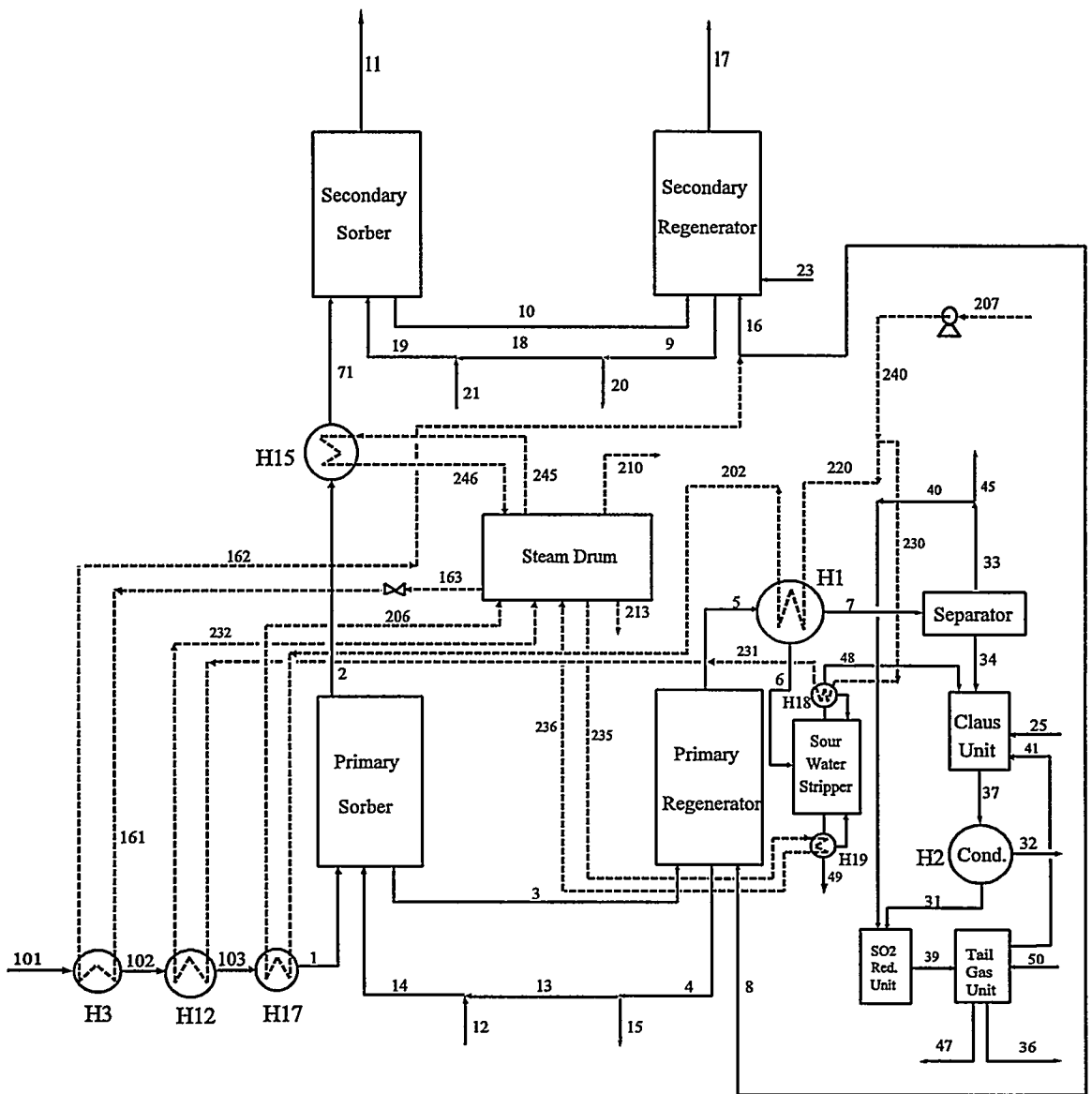


Figure 15. Two-Stage Desulfurization With Steam Regeneration, 900K
 Primary Sorber Temperature

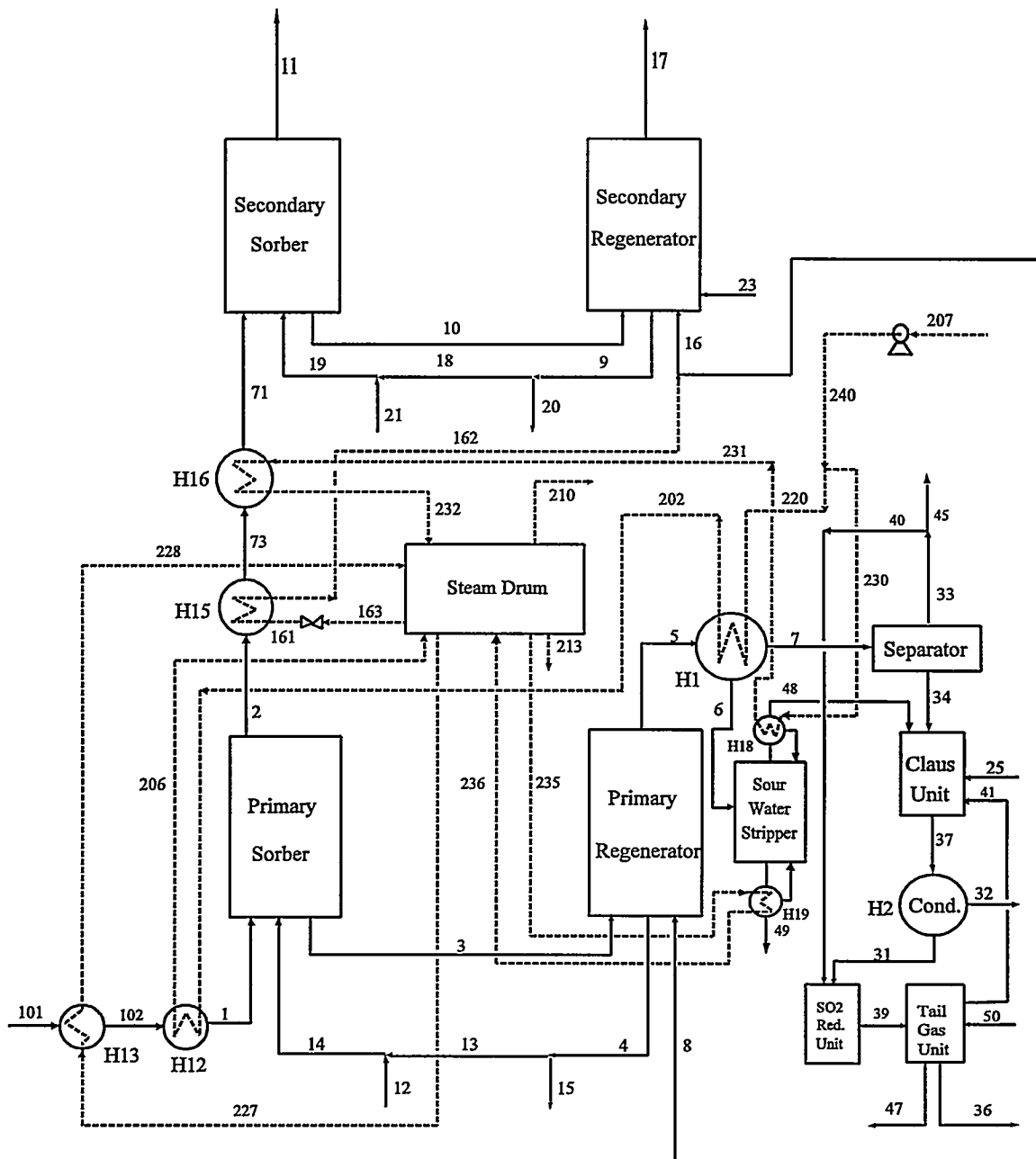


Figure 16. Two-Stage Desulfurization with Steam Regeneration, 1000K
Primary Sorber Temperature

Adiabatic operation of the sorbers and regenerators means that reactor temperatures are no longer independent. The reaction temperature, which is assumed equal to the product temperature, is determined by the flow rates and temperatures of all feed streams coupled with the energy released or absorbed by the reaction. The actual outlet temperatures are near the nominal values as shown in Table 2.

The Claus reactor and downstream tail gas unit have not been heat integrated with the remainder of the desulfurization process. Since these are common processes, we assume that they would serve as an independent, modular add-on process.

A summary of overall process input and output flow rates for these two cases is presented in Table 3. Input and output coal gas flow rates (streams 101 and 11) are equal, as are the make-up and discharge rates of primary sorbent (streams 12 and 15) and secondary sorbent (stream 21 and 20). Sorbent rates are based on the assumption that the replacement rate was 1% of the circulation rate. Primary sorbent rate is independent of temperature while the secondary sorbent rate is increased in case H₂O-B because less H₂S is removed in the primary sorber at the lower temperature. The flow of steam to the primary regenerator is lower in case H₂O-B for the same reason. The increased sulfur removal in the secondary sorber at the lower temperature (case H₂O-B) is responsible for the increased flow rates of steam (stream 16) and oxygen (stream 23) to the secondary regenerator as well as the increased off-gas flow rate (stream 17) from the secondary regenerator.

The flow rates of all input and output streams downstream of the primary regenerator (streams 25, 50, 49, 45, 32, 36, and 47) are associated with elemental sulfur recovery. All of these rates are smaller in case H₂O-B, again because less H₂S is removed in the primary sorber at 900K. Finally, less make-up water (stream 207) is required and less excess steam (stream 210) is produced in low temperature case H₂O-B. The difference between the make-up water and product steam flow rates is due primarily to the quantity of process steam fed to the primary (stream 8) and secondary (stream 16) regenerators.

The sulfur balance for the steam regeneration cases is summarized in Table 4. 5500 lb/hr of sulfur is present in the coal gas (stream 101). Most of this is recovered as elemental sulfur product (stream 32) or recycled to the gasifier in the secondary regenerator off-gas (stream 17). The total sulfur in these two streams is effectively equal in both cases and amounts to 99.8% of the sulfur fed. Less elemental sulfur is produced directly in case H₂O-B because less sulfur is removed in the primary sorber at the lower temperature. However, since the off-gas from the secondary regenerator (stream 17) is recycled to the gasifier, the sulfur present in this stream would eventually be captured and recovered as elemental sulfur. Thus, for practical purposes the elemental sulfur recovery is approximately equal to 99.8% in both cases. Full incorporation of the recycle stream into the material and energy balance calculations would require inclusion of the gasifier, which is beyond the scope of this study.

Table 2. Inlet and Outlet Temperatures of the Primary and Secondary Sorbers and Regenerators for Two-Stage Desulfurization with Steam Regeneration.

	Temperature, K		
	<u>Nominal</u>	<u>Avg. Inlet</u>	<u>Outlet</u>
Case H ₂ O-A			
Primary Sorber	1000	1012	1001
Primary Regenerator	1000	914	979
Secondary Sorber	873	872	874
Secondary Regenerator	1000	811	1005
Case H ₂ O-B			
Primary Sorber	900	916	902
Primary Regenerator	900	867	932
Secondary Sorber	873	873	875
Secondary Regenerator	1000	811	998

Table 3. Summary of Material Balance Results for Two-Stage Desulfurization with Steam Regeneration

Input Stream, Description	Stream Flow Rate, lb-mol/hr	
	Case H ₂ O-A	Case H ₂ O-B
101. Coal Gas From Gasifier	17000	17000
12. Primary Sorbent Make-up	6.9	6.9
21. Secondary Sorbent Make-up	0.1	0.2
8. Steam to Primary Regenerator	1150	1030
16. Steam to Secondary Regenerator	550	970
23. Oxygen to Secondary Regenerator	17	30
25. Air to Claus Reactor	530	500
50. Make-up Solvent to Tail Gas Unit	0.1	0.1
207. Make-up Water	5380	5180
Output Stream, Description		
11. Desulfurized Coal Gas	17000	17000
15. Primary Sorbent Discharge	6.9	6.9
20. Secondary Sorbent Discharge	0.1	0.2
17. Secondary Regenerator Off-Gas	560	990
49. Water From Sour Water Stripper	820	720
45. Hydrogen Product	110	100
32. Elemental Sulfur Product	250	240
36. Gas Discharge from Sulfur Tail Gas Unit	430	410
47. Liquid Discharge from Sulfur Tail Gas Unit	120	120
210. Product Steam	3670	3170

Table 4. Summary of Sulfur Material Balance Results for
Two-Stage Desulfurization with Steam Regeneration.

	Sulfur Flow Rate, lb/hr	
	Case H ₂ O-A	Case H ₂ O-B
Input Stream (Sulfur Form)		
101. Coal Gas from Gasifier (H ₂ S)	5500	5500
Output Stream (Sulfur Form)		
11. Desulfurized Coal Gas (H ₂ S)	5.9	5.9
17. Secondary Regenerator Off-Gas (SO ₂)	340	582
32. Elemental Sulfur Product (S)	5150	4910
36. Sulfur Tail Gas Unit Off-Gas (H ₂ S)	<0.1	<0.1
47. Water discharge from Tail Gas Unit (H ₂ S)	2	2
49. Water Discharge from Sour Water Stripper (H ₂ S)	<0.1	<0.1

Approximately 0.1% of the entering sulfur leaves in the desulfurized coal gas (stream 11) which corresponds to an H₂S concentration of about 10 ppmv. The remaining sulfur, approximately 2 lb/hr, is discharged to the environment from the sulfur recovery process (streams 36, 47, and 49).

Heat duties associated with the condensers, exchangers, and heat recovery steam generators along with the horsepower of the water feed pump are presented in Table 5. The pump is the only mechanical device in the process and the horsepower is based on a pump efficiency of 65%.

In case H₂O-B the three heat exchangers upstream of the primary sorber (H3, H12, and H17) remove a total of 90.9×10^6 Btu/hr in cooling the coal gas from 1283K to near 900K. An additional 8.2×10^6 Btu/hr is removed in H15 in cooling the partially desulfurized coal gas between the primary and secondary sorbers. In case H₂O-A, the two exchangers (H13 and H12) upstream of the primary sorber remove 66.8×10^6 Btu/hr while the two exchangers (H15 and H16) between the sorbers remove an additional 31.2×10^6 Btu/hr. The total heat transferred from the coal gas is approximately equal in both cases; the distribution is different because of the different operating temperatures of the primary sorber.

Exchanger H1 condenses excess steam from the primary regenerator product. The amount of heat removed in case H₂O-B is somewhat less because less sulfur is removed in the primary regenerator at the lower temperature. Exchanger H2 condenses elemental sulfur from the Claus reactor product. Again the heat duty is somewhat less in case H₂O-B because less sulfur is produced. Exchangers H18 and H19 serve as condenser and reboiler, respectively, for the sour water stripper. These heat duties are also somewhat smaller in case H₂O-B because of the reduced sulfur removal in the primary sorber at the lower temperature.

Two-Stage Desulfurization With SO₂ Regeneration

Four case studies involving two-stage desulfurization with SO₂ regeneration are discussed in this section. The process flow diagram shown in Figure 17 is applicable to both. Process flows (although not necessarily flow rates) associated with the primary and secondary sorbers and the secondary regenerator are the same as with two-stage desulfurization with steam regeneration. However, the direct production of elemental sulfur in the primary regenerator simplifies the remainder of the process. The sulfur condenser (H1) and recycle compressor replace the steam stripper, Claus reactor, tail gas unit, and related equipment.

As with steam regeneration, feed water (stream 207) is distributed through heat exchangers, condensers, and heat recovery steam generators to produce saturated steam at 1250 psia. The primary and secondary sorbers and the secondary regenerator operate adiabatically. However, because of the highly exothermic reaction between C₂O₂S and SO₂, adiabatic

Table 5. Summary of Heat Duties and Process Power Requirements for for Two-Stage Desulfurization with Steam Regeneration.

Unit	Heat Duty x 10 ⁶ , Btu/hr	
	Case H ₂ O-A	Case H ₂ O-B
H1	26.0	22.1
H2	14.6	13.9
H3	--	10.9
H12	64.8	17.6
H13	2.0	--
H15	9.3	8.2
H16	21.9	--
H17	--	62.4
H18	3.2	2.8
H19	4.7	4.1
Pump Horsepower	220	212

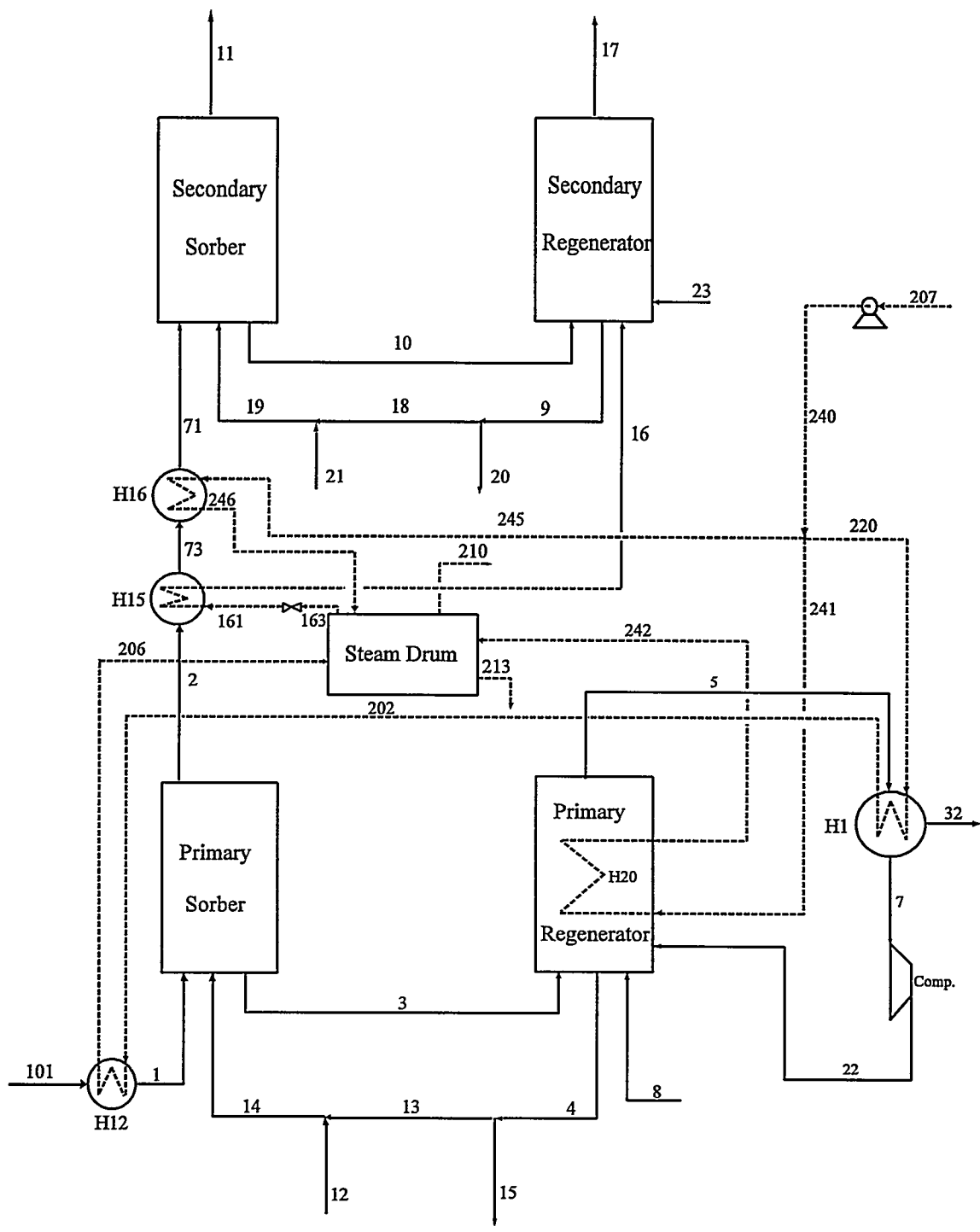


Figure 17. Two-Stage Desulfurization With SO₂ Regeneration

operation of the primary regenerator is not practical. Sufficient heat exchange capability is included to produce the desired reaction temperature.

The four case studies are defined in Table 6 in terms of the temperatures of the reactors. In cases SO₂-A and SO₂-B the primary sorber and regenerator temperatures are set at 1000K and 900K, respectively, while in case SO₂-C and SO₂-D, the primary sorber is at 1073K and primary regenerator at 873K. These temperatures are equal to those presently being used in the laboratory experimental program. The temperatures of the secondary sorber and regenerator are 873K and 1000K, respectively, in all cases. The temperatures and flow rates of feeds to the adiabatic reactors are such that, when coupled with the heat of reaction, the product temperature is approximately equal to the nominal temperature. Sufficient heat transfer capability is included in the primary regenerator to control the product temperature to the desired level.

All temperatures for cases SO₂-C and SO₂-D in Table 6 are equal, and these cases differ in the recycle rate of SO₂ to the primary regenerator. It was impossible to accomplish the required primary regenerator heat transfer using the low SO₂ recycle rate associated with case SO₂-C. Therefore, the recycle rate was increased in Case SO₂-D in order to accommodate the heat transfer needs. The distinctions in these cases are discussed in more detail in the later section on vessel design.

Material balance results for the SO₂ regeneration cases are summarized in Tables 7 and 8. Table 7 includes all overall process input and output streams while Table 8 shows the overall sulfur balance. Comparison of Table 3 for steam regeneration and Table 7 for SO₂ regeneration shows that several of the stream flows are independent of both temperature and the process used in the primary regenerator. Included in the category are the coal gas feed (stream 101), primary sorbent make-up (stream 12), desulfurized coal gas (stream 11), and primary sorbent discharge (stream 15). The flow rates of additional streams are independent of the process but vary with temperature. This group includes the secondary sorbent make-up (stream 21), the steam to the secondary regenerator (stream 9), oxygen to the secondary regenerator (stream 23 in Table 3 and stream 10 in Table 7), secondary sorbent discharge (stream 20), and secondary regenerator off-gas (stream 17). The flow rates of all these streams decrease with increasing temperature of the primary sorber since more sulfur is removed in the primary sorber at higher temperature.

The total flow rate of the elemental sulfur product (stream 32 in Table 7) is smaller in the SO₂ regeneration case, but the sulfur content of this stream is equal at equal primary sorber temperature (stream 32 in Table 8). The sulfur product in steam regeneration contains a large quantity of water impurity. In contrast, the sulfur product stream from SO₂ regeneration is essentially pure. The flow rate of sulfur product increases with increasing primary sorber temperature. Oxygen to the primary regenerator (stream 8 in Table 7) does not exist in the steam regenerator case. The oxygen is supplied to react with elemental sulfur and maintain a constant flow of SO₂ to the primary regenerator. The flow rate of oxygen increases with increasing primary sorber temperature.

Table 6. Inlet and Outlet Temperatures of the Primary and Secondary Sorbers and Regenerators for Two-Stage Desulfurization with SO₂ Regeneration.

	Temperature, K		
	Nominal	Avg. Inlet	Outlet
Case SO ₂ -A			
Primary Sorber	1000	1014	1003
Primary Regenerator	1000	866	1000
Secondary Sorber	873	872	874
Secondary Regenerator	1000	806	1000
Case SO ₂ -B			
	900	914	903
Primary Sorber	900	791	900
Primary Regenerator	873	873	875
Secondary Sorber	1000	813	1001
Secondary Regenerator			
Case SO ₂ -C			
Primary Sorber	1073	1085	1074
Primary Regenerator	873	857	873
Secondary Sorber	873	872	873
Secondary Regenerator	1000	820	1004
Case SO ₂ -D			
Primary Sorber			
Primary Regenerator	1073	1085	1074
Secondary Sorber	873	711	873
Secondary Regenerator	873	872	873
	1000	820	1004

Table 7. Summary of Material Balance Results for Two-Stage Desulfurization with SO₂ Regeneration

Case	Stream Flow Rate, lb mol/hr			
	SO ₂ -A	SO ₂ -B	SO ₂ -C	SO ₂ -D
Input Stream, Description				
101. Coal Gas From Gasifier	17,000	17,000	17,000	17,000
12. Primary Sorbent Make-up	6.9	6.9	6.9	6.9
21. Secondary Sorbent Make-up	0.1	0.2	0.1	0.1
8. Oxygen to Primary Regenerator	160	150	164	164
9. Steam to Secondary Regenerator	550	970	424	424
10. Oxygen to Secondary Regenerator	17	30	13	13
207. Make-up Water	13,600	13,000	11,400	6,500
Output Stream, Description				
11. Desulfurized Coal Gas	17,000	17,000	17,000	17,000
15. Primary Sorbent Discharge	6.9	6.9	6.9	6.9
20. Secondary Sorbent Discharge	0.1	0.2	0.1	0.1
17. Secondary Regenerator Off-Gas	560	990	430	430
32. Elemental Sulfur Product	160	150	165	165
210. Product Steam	13,100	12,000	11,000	6,050

Table 8. Summary of Sulfur Material Balance Results for Two-Stage Desulfurization with SO₂ Regeneration.

Case	Sulfur Flow Rate, lb/hr			
	SO ₂ -A	SO ₂ -B	SO ₂ -C	SO ₂ -D
Input Stream (Sulfur Form)	5500	5500	5500	5500
101. Coal Gas From Gasifier (H ₂ S)				
Output Stream (Sulfur Form)				
11. Desulfurized Coal Gas (H ₂ S)	5.9	5.9	5.9	5.9
17. Secondary Regenerator Off-Gas (SO ₂)	340	580	252	252
32. Elemental Sulfur Product (S)	5150	4910	5240	5240

The flow rates of feed water (stream 207) and product steam (stream 210) decrease with increasing primary sorber temperature. The only streams whose flow rates differ between cases SO₂-C and SO₂-D are the feed water and product steam. The increased SO₂ recycle rate to the primary regenerator in case SO₂-D reduces the heat transfer requirement in that reactor, which results in the large decrease in water demand and steam production. Since the only process steam in the SO₂ regeneration option is that required in the secondary regenerator, most of the make-up water (stream 207) is available as product steam (stream 210). Even in case SO₂-D the amount of product steam is larger than in the steam regeneration cases because of the large amount of heat liberated by the exothermic primary regeneration reaction.

Sulfur flow rates for the two regeneration concepts are similar except that the small discharges from the Claus and tail gas units are not present in the SO₂ regeneration case. The amount of sulfur in the coal gas feed (stream 101) and in the desulfurized coal gas (stream 11) is equal in all cases. As with steam regeneration, the H₂S content of the desulfurized coal gas is about 10 ppmv. The amount of elemental sulfur product (stream 32) increases with increasing primary sorber temperature while the sulfur in the secondary regenerator off-gas (stream 17) decreases by a corresponding amount. As before, the secondary regenerator off-gas is recycled to the gasifier so that this sulfur will ultimately be captured and removed as elemental sulfur. In all of the SO₂ regeneration cases the overall sulfur recovery is greater than 99.8%.

Heat duties of the exchangers and power requirements for the pump and recycle compressor are presented in Table 9. The duty of the elemental sulfur condenser, H1, increases as the temperature of the primary sorber increases since more H₂S is removed at high temperature. The heat duty also increases as the SO₂ recycle rate is increased as shown by comparison of cases SO₂-C and SO₂-D. Heat removed from the coal gas feed in H12 decreases at higher primary sorber temperature but is independent of the SO₂ recycle rate to the primary regenerator. In contrast, heat removed from the partially desulfurized coal gas in H15 and H16 decreases as the primary sorber temperature decreases, but is independent of SO₂ recycle rate. Increasing the SO₂ recycle rate has the strongest effect on the heat which must be removed from the primary regenerator (H20). The large values ranging from 137.1 x 10⁶ to 181.9 x 10⁶ Btu/hr associated with cases SO₂-A, SO₂-B and SO₂-C are not compatible with the design criteria for the fluidized-bed reactor. It was necessary to increase the SO₂ recycle rate by a factor of 3, from 330 lbmol/hr in case SO₂-C to 1000 lbmol/hr in case SO₂-D in order to satisfy the heat transfer requirements. This topic is addressed in greater detail in the reactor design section of this report.

The increased SO₂ recycle rate was responsible for the increased heat duty of H1, the reduced feed rate of water, the corresponding reduction in pump horsepower, and the increased power required for the recycle compressor. The water flow rate and pump horsepower also decrease with increasing primary sorber temperature while the recycle compressor horsepower requirement increases with increasing temperature.

Table 9. Summary of Heat Duties and Process Power Requirements for Two-Stage Desulfurization with SO₂ Regeneration.

Unit	Heat Duty x 10 ⁻⁶ , Btu/hr			
	SO ₂ -A	SO ₂ -B	SO ₂ -C	SO ₂ -D
H1	9.0	8.6	10.3	17.7
H12	66.8	91.0	44.7	44.7
H15	3.0	5.3	2.4	2.4
H16	28.6	2.1	46.8	46.8
H20	181.9	171.8	137.1	27.2
Pump Horsepower	557	532	465	265
Recycle Compressor Horsepower	16	20	33	118

Single-Stage Desulfurization with DSRP

A single material and energy balance case study has been completed for single-stage desulfurization with elemental sulfur recovery using the DSRP process. The flow diagram for this process is shown in Figure 18. Coal gas (stream 101) passes successively through H2, H4, and H1 and is then split (streams 2 and 3). The majority of the coal gas is fed to the sorber (stream 2) where H₂S is removed in a single-stage by reaction with Zn₂TiO₄. A small side stream of the coal gas (stream 3) is fed directly to the DSRP reactor where reducing components (H₂ and CO) react with SO₂ to produce elemental sulfur. Spent sorbent is transferred to the regenerator where it reacts with air diluted with nitrogen for temperature control. SO₂ in the regenerator off-gas (stream 12) is reduced to elemental sulfur in the DSRP reactor. The product sulfur (stream 32) is removed in the condenser, H9, and the remaining gases (stream 17) are recycled to the regenerator (stream 14) after passing through heat exchanger H3, or purged (stream 24) to prevent build-up of carbon oxides in the recycle gas.

Regenerated sorbent (stream 8) is recycled to the sorber after a portion is discharged (stream 6) and replaced with make-up (stream 5) sorbent to maintain activity.

The feed water (stream 207) is pumped to 1250 psig (stream 240) and split into four streams. Streams 251 and 252 are used in the interstage coolers of the three-stage air compressor. From the coolers, the streams rejoin (stream 255) and are vaporized in the waste heat boiler H2 (stream 256) before flowing to the steam drum. Water stream 221 exits the sulfur condenser as superheated steam (stream 204), which is used to preheat the make-up nitrogen diluent (stream 151) in H8. From H8, stream 200 flows to the steam drum. The remainder of the water (stream 222) is used to remove heat from the DSRP reactor, after which it (stream 223) goes to waste heat boiler, H1, where it becomes superheated steam (stream 224). This superheated steam is used to preheat the nitrogen-rich recycle (stream 18) to the regenerator, and then it (stream 202) enters the steam drum. Additional heat available from the coal gas feed is used to convert saturated water (stream 211) to steam (stream 212) in heat recovery boiler H4. Excess product steam (stream 210) is available for supplemental power generation while stream 213 represents a small flow of discharge liquid from the steam drum.

Process conditions for the material and energy balance were based on reports from RTI (Gangwal, et al., 1993). Reactor inlet and outlet temperatures are compared to the nominal values in Table 10. The sorber and regenerator are assumed to operate adiabatically while the DSRP reactor includes heat transfer capability to achieve the desired outlet temperature of 873K.

Sorbent discharge (stream 6) and make-up (stream 5) rates are arbitrarily set at 1% of the sorbent circulation rate. Sufficient diluent N₂ (stream 151) is added to the regenerator to produce a combined regenerator feed containing 3% oxygen, with the air feed rate (stream 16) 10% above the stoichiometric quantity required for complete sorbent regeneration and oxidation of any residual reducing gas from the coal gas fed to the DSRP reactor. The flow rate of reducing gas to the DSRP reactor (stream 3) is the stoichiometric amount required to react with the SO₂ and excess O₂ in the regenerator product stream (stream 12).

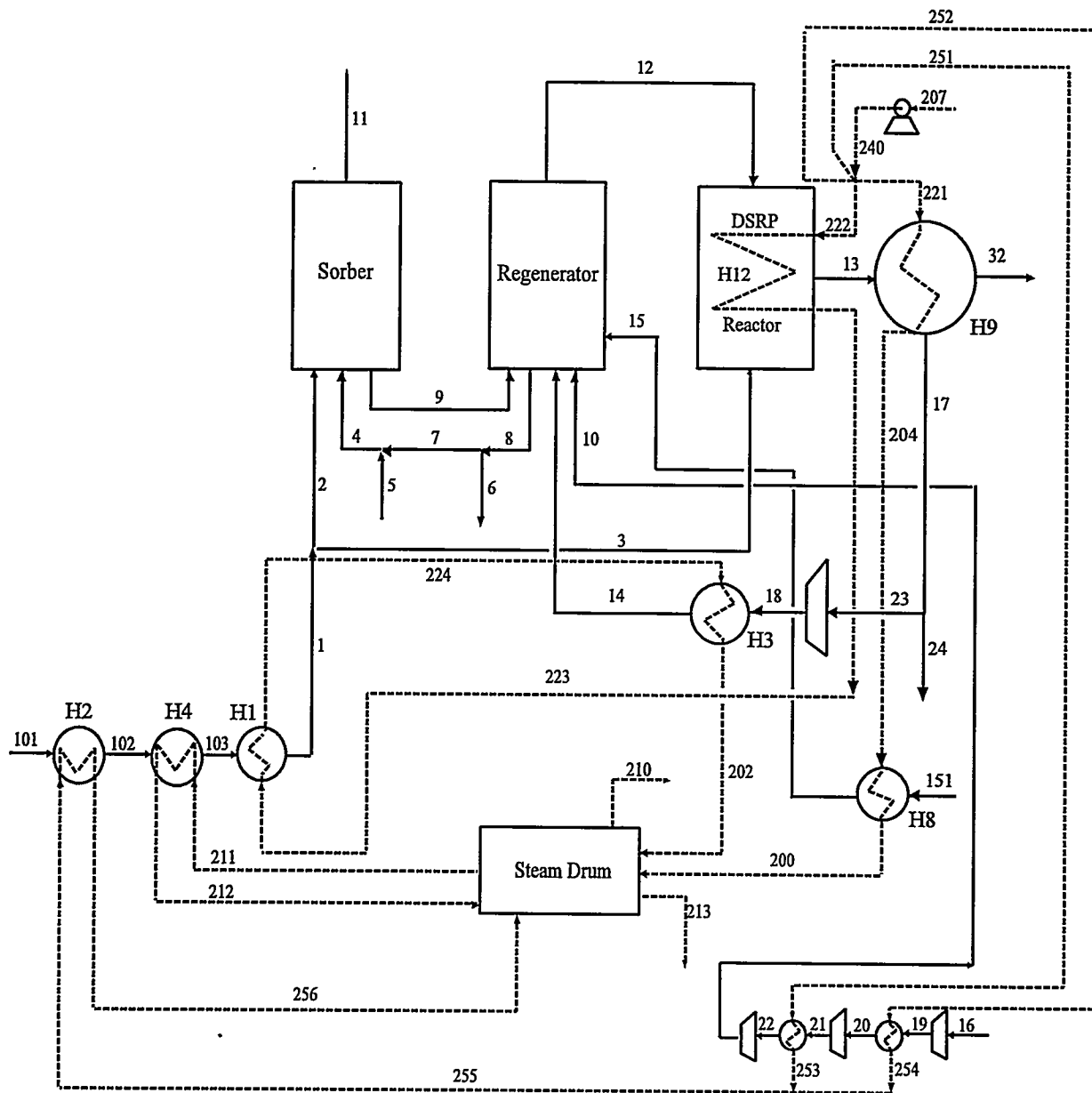


Figure 18. Two-Stage Desulfurization with DSRP

Table 10. Inlet and Outlet Temperatures of the Reactors for Single-Stage Desulfurization with DSRP.

Reactor	Nominal	Temperature, K	
		Avg. Inlet	Outlet
Sorber	873	783	887
Regenerator	1000	783	996
DSRP Reactor	873	990	873

The overall mass balance, sulfur balance, and vessel heat duties are summarized in Tables 11, 12, and 13, respectively. The coal gas feed rate (stream 101) of 17,000 lbmol/hr is the equal to the rate used for the other processes. However, the flow rate of desulfurized coal gas (stream 11) is reduced by about 3% to 16,500 lbmol/hr because of the side stream to the DSRP reactor (stream 3). Make-up nitrogen (stream 151) is used as diluent for temperature control in the regenerator, while the oxygen for sorbent regeneration is obtained by compressing air (stream 16). Sorbent make-up (stream 5) and discharge (stream 6) are based on 1% of the sorbent circulation rate. The elemental sulfur product (stream 32) from the condenser contains both sulfur and water (about 62 mol% sulfur). The gas purge (stream 24) is required to prevent the build-up of carbon oxides in the recycle gas. Finally, since no process water is required, the flow rates of make-up water (stream 207) and product steam (stream 210) are effectively equal.

From Table 12, we see that approximately 99.8% of the sulfur fed to the process (stream 101) is recovered as elemental sulfur (stream 32). About 0.1% of the sulfur exits in the clean coal gas (stream 11) which has a H₂S concentration of about 10 ppmv. The remaining 0.1% of the sulfur is discharged in the recycle purge gas (stream 24).

Process heat duties and power requirements are presented in Table 13. 104.1×10^6 Btu/hr are removed from the coal gas in H1, H2, and H4. This value is somewhat larger than in either of the two-stage desulfurization cases because the operating temperature of the single sorber is lower than the temperature of the primary sorber. There are no equivalents to the interstage coolers, H6 and H7, the recycle heater, H3, and the nitrogen preheater, H8, in the two-stage desulfurization options. The 23.9×10^6 Btu/hr removed from the DSRP reactor is only marginally less than the 27.2×10^6 Btu/hr removed from the primary regenerator in the SO₂ regeneration case SO₂-D.

Process power requirements are significantly larger than in the two stage desulfurization cases, due primarily to the three-stage air compressor.

Process Comparison

While the final evaluation of process options must be on an economic basis, it is possible at this stage to develop qualitative comparisons. One comparison is based on process complexity as measured by the number of processing steps required. The three processes are compared on this basis in Table 14, which does not include secondary processing steps such as condensers, heat exchangers, waste heat boilers, compressors, and pumps. Two-stage desulfurization with steam regeneration is clearly the most complex option because of the additional processing steps associated with sulfur recovery using Claus technology. Although this technology is well-developed, it is almost certain to result in higher capital cost. Single-stage desulfurization with DSRP is somewhat simpler than two-stage desulfurization with SO₂ regeneration because a single DSRP reactor takes the place of the secondary sorber and regenerator.

Table 11. Summary of the Material Balance Results for Single-Stage Desulfurization with DSRP (Case DSRP-A).

Input Stream, Description	Stream Flow Rate, lbmol/hr
101. Coal Gas from Gasifier	17,000
151. Make-up N ₂	1100
16. Air to Regenerator	1320
5. Sorbent Make-up	1.7
207. Make-up Water	7590
Output Stream, Description	
11. Desulfurization Coal Gas	16500
32. Elemental Sulfur Product	276
24. Recycle Gas Purge	2470
6. Sorbent Discharge	1.7
210. Product Steam	7590

Table 12. Summary of Sulfur Material Balance Results for Single-Stage Desulfurization with DSRP (Case DSRP-A).

Input Stream (Sulfur Form)	Sulfur Flow Rate, lb/hr
101. Coal Gas from Gasifier	5500
Output Stream (Sulfur Form)	
11. Desulfurized Coal Gas (H_2S)	5.4
32. Elemental Sulfur Product (S)	5488
24. Recycle Purge Gas (H_2S)	5.1

Table 13. Summary of Heat Duties and Process Power Requirements for Single-Stage Desulfurization with DSRP (Case DSRP-A).

Unit	Heat Duty x 10 ⁻⁶ , Btu/hr
H1	70.4
H2	13.8
H3	48.7
H4	19.9
H6	1.1
H7	2.6
H8	3.1
H9	79.4
H12	1.1
	2.6
Pump Horsepower	310
Recycle Compressor Horsepower	390
Three-Stage Air Compressor Horsepower	2840

Table 14. Major Process Steps Required for Coal Gas
Desulfurization With Elemental Sulfur Recovery
Using Three Process Concepts

Two-Stage Desulfurization, <u>Steam Regeneration</u>	Two-Stage Desulfurization, <u>SO₂ Regeneration</u>	Single-Stage Desulfurization, <u>DSRP</u>
Primary Sorber Primary Regenerator Secondary Sorber Secondary Regenerator H ₂ /H ₂ S Separator Claus Reactor Tail Gas Unit	Primary Sorber Primary Regenerator Secondary Sorber Secondary Regenerator	Primary Sorber Primary Regenerator DSRP Reactor

Power requirements for pumps and compressors for the three processes are compared in Table 15. For this comparison, results associated with two-stage desulfurization with stream regeneration, case H₂O-A, two-stage desulfurization with SO₂ regeneration, cases SO₂-C and SO₂-D, and single-stage desulfurization with DSRP, case DSRP-A, have been selected. The three stage air compressor used in single-stage desulfurization with DSRP is easily the largest power user. Neither of the two-stage desulfurization options require an air compressor. Instead, high purity oxygen, which we assume is obtained from the air separation plant associated with the gasifier, is required in both two stage options. Both two-stage desulfurization with SO₂ regeneration and single-stage desulfurization with DSRP require recycle compressors. However, the quantity of recycle gas in DSRP-A is much larger. Feed water pumps are required for all options and the horsepower is directly proportional to the feed water rate.

The differences between cases SO₂-C and SO₂-D are associated with the SO₂ recycle rate. A small recycle rate is used in SO₂-C which, as will be shown subsequently, is not consistent with the heat transfer requirements in the primary regenerator. Increasing the SO₂ recycle rate to the point that the heat transfer requirements can be satisfied results in an increase in the recycle compressor horsepower and a decrease in the feed water pump horsepower. The production rate of high pressure steam is also reduced when the SO₂ recycle rate is increased. The power requirements of the pump and compressor are relatively insensitive to the operating temperature of the primary sorber. Thus the power requirements associated with the other case studies are similar to those shown in Table 15.

The flow rates of "purchased" raw materials and by-products to be "sold" are compared in Table 16. The case studies listed in Tables 15 and 16 are the same. The composition and feed rate of coal gas are equal in each of the cases, as is the sulfur content and temperature of the desulfurized coal gas. The flow rate of desulfurized coal gas is the same as the coal gas feed rate in the two-stage desulfurization options, but is reduced by about 3% in the single-stage desulfurization process since a portion of the coal gas is diverted to the DSRP reactor.

All sorbent replacement rates in Table 16 are arbitrary and are based on an average sorbent lifetime of 100 cycles, i.e., the replacement rate is 1% of sorbent circulation rate. This replacement rate provides a large advantage to single-stage desulfurization with DSRP as the total sorbent replacement rate is smaller and zinc titanate sorbent is expected to be less expensive per pound than CeO₂. However, even single-stage desulfurization with DSRP may not be economical at these conditions since an average sorbent lifetime well in excess of 100 cycles is believed to be necessary. The overall process material and energy balances are effectively independent of the sorbent replacement rate. Thus, in the final economic analysis the sorbent replacement rate will be varied as a sensitivity parameter.

Oxygen is required in both two-stage desulfurization processes with the total oxygen use considerably greater in the SO₂ regeneration option. As previously indicated, we assume that oxygen will be purchased from the air separation plant associated with the gasifier. Nitrogen, rather than oxygen, is required in the single-stage desulfurization option, with the nitrogen also to be purchased from the gasifier air separation plant.

Table 15. Comparison of Pump and Compressor Horsepower Requirements for Three Desulfurization Processes With Elemental Sulfur Recovery.

Case	Two-Stage Desulfurization, Steam Regeneration	Two-Stage Desulfurization, SO ₂ Regeneration		Single-Stage Desulfurization, DSRP
	H ₂ O-A	SO ₂ -C	SO ₂ -D	DSRP-A
Feed Water Pump	220	465	265	310
Primary Regenerator Recycle Compressor	--	33	118	390
Regenerator Air Compressor	--	--	--	2840

Table 16. Comparison of Raw Material and Product Rates for Three Desulfurization Processes with Elemental Sulfur Recovery.

Case	Two-Stage Desulfurization, Steam Regeneration	Two-Stage Desulfurization, SO ₂ Regeneration		Single-Stage Desulfurization DSRP
	H ₂ O-A	SO ₂ -C	SO ₂ -D	DSRP-A
Input Streams				
Coal Gas from Gasifier lb mol/hr	17,000	17,000	17,000	17,000
Sorbent Replacement, lb/hr				
CeO ₂	1200	1200	1200	--
Zn ₂ TiO ₄	48	48	48	413
Oxygen, lbmol/hr				
Primary Regenerator	--	164	164	--
Secondary Regenerator	30	13	13	--
Nitrogen, lbmol/hr				
Primary Regenerator	--	--	--	1100
Water, lb/hr	93,000	205,200	117,000	137,000
Product Streams				
Desulfurized Coal Gas, lbmol/hr	17,000	17,000	17,000	16,500
H ₂ S Content, ppmv	10	10	10	10
Hydrogen, lbmol/hr	100	--	--	--
Elemental Sulfur, lb/hr	5490	5490	5490	5490
Sulfur Purity (mass%)	76	>99	>99	74
High Pressure Steam, lb/hr	57,100	198,000	108,900	136,000

All options result in the production of excess high pressure steam which can be "sold" to the power generation plant. The water input rate is sufficient to satisfy process requirements and excess steam production. The largest feed water requirement and steam production rate is associated with case SO₂-C. However, this case is not practical because of heat transfer limitations, and case DSRP-A shows the next largest water and steam flows. The water and steam rates associated with SO₂-D are intermediate in magnitude. The largest difference between the water feed rate and steam production rate is associated with case H₂O-A since more process steam is required for steam regeneration.

Elemental sulfur production rates are effectively equal in all cases. This is based on the assumption that SO₂ from the secondary regenerator in all two-stage regeneration cases is recycled to the gasifier and then back to the primary sorber where all of the sulfur is eventually recovered as elemental sulfur product. An additional disadvantage associated with two-stage desulfurization with steam regeneration includes the fact that aqueous waste streams containing sulfur are produced. The two-stage desulfurization with SO₂ regeneration and single-stage regeneration with DSRP processes are totally dry with no aqueous discharge. However, a small amount of sulfur is present in the gas purge stream in the single-stage process with DSRP. The purity of the elemental sulfur produced by two-stage desulfurization with SO₂ regeneration is considerably larger than in either of the other processes where significant amounts of water are also condensed with the sulfur product. The only other by-product is the relatively small amount of high purity hydrogen produced in the steam regeneration option. This hydrogen may either be sold or added to the clean coal gas.

At this stage of the process modeling effort, it appears that both two-stage desulfurization with SO₂ regeneration and single-stage desulfurization with DSRP are preferable to two-stage desulfurization with steam regeneration. This is based on the simpler processing schemes and the fact that no aqueous waste streams are produced. There is no clear advantage between two-stage desulfurization with SO₂ regeneration and single-stage desulfurization with DSRP. The latter involves one less major processing step but this is somewhat offset by the large air compressor required in the single-stage process. Also important are the relative costs of replacement sorbent and the purchased cost of oxygen for two-stage desulfurization compared to the cost of purchased nitrogen for single-stage desulfurization. Finally, there may be additional by-product credit which can be claimed for the higher purity sulfur produced in the two-stage process with SO₂ regeneration. The relative importance of these factors will be developed in the process economic analysis.

Reactor Design

Preliminary reactor vessel design for two-stage desulfurization with SO₂ regeneration, case SO₂-C and case SO₂-D, and for single-stage desulfurization with DSRP, case DSRP-A, were completed during the quarter. Sorber and regenerator design was based on bubbling fluidized beds, while the DSRP reactor was assumed to operate in a fixed-bed mode. Four parallel reactor trains were assumed in each case. As previously stated, the primary and secondary

sorbers and the secondary regenerator in two-stage desulfurization, and the sorber and regenerator in the single-stage process operate adiabatically. However, the energy liberated by the exothermic $\text{Ce}_2\text{O}_2\text{S-SO}_2$ and DSRP reactions is too large for adiabatic operation. Instead, sufficient heat transfer capability is included within the reactors so that the temperatures of the outlet streams are equal to the desired nominal temperature.

The design of the fluidized-bed reactors followed the procedure described by Chen et al. (1991). The open area of the reactor was fixed to provide a superficial gas velocity of 1 ft/s, which corresponds to 5 to 7 times the minimum fluidization velocity. The height of the fluidized bed was then fixed at 6 feet to provide 6 seconds of contact time. The total vessel height was increased to 9 feet to provide 50% freeboard. Because of the high reactor temperatures, 6 inches of refractory lining was included in each reactor. An approximate heat transfer calculation was then used to estimate the temperature of the steel pressure vessel, and the required vessel wall thickness was calculated from ASTM specifications (Peters and Timmerhaus, 1991).

The fixed-bed DSRP reactor was designed on the basis of conditions reported by Gangwal et al. (1993). The packed volume was based on a space velocity of 4560 hr^{-1} (STP), and this volume was distributed appropriately between cross-sectional area and packed height. Again, 6 inches of refractory lining was added and the vessel wall thickness calculated using the method used for the fluidized-bed reactors.

When this procedure was applied to two-stage desulfurization with SO_2 regeneration (case $\text{SO}_2\text{-C}$), the resulting vessel could not accommodate the required heat transfer area. 1.37×10^8 Btu/hr must be removed from the primary regenerator, and, using an overall heat transfer coefficient of $10 \text{ Btu/hr ft}^2 \text{ }^\circ\text{F}$, approximately 1550 heat transfer tubes would be required in each of the four primary regenerators. This number of tubes could not be accommodated in the vessel without reducing the gas velocity below 1 ft/s.

Increasing the SO_2 recycle rate reduces the quantity of heat which must be removed from the regenerator and increases the cross-sectional area required for a gas velocity of 1 ft/s. The larger cross-sectional area provides greater flexibility in accommodating heat transfer tubes. A trail-and-error procedure was used to determine a SO_2 recycle rate consistent with heat transfer requirements. The rate selected corresponded to an increase in the SO_2 recycle rate by a factor of 3, from 330 to 1000 lbmol/hr. This reduced the heat transfer requirement to 2.72×10^7 Btu/hr which could be accomplished by adding 300 tubes to each reactor, again based on an overall heat transfer coefficient of $10 \text{ Btu/hr ft}^2 \text{ }^\circ\text{F}$.

The final vessel dimensions are shown in Table 17. The vessel outside diameter is based on an open cross-sectional area required to produce a gas velocity of 1 ft/s, plus sufficient area for heat transfer tubes (when required), plus 6 inches of refractory lining, plus the indicated pressure vessel wall thickness.

Table 17. Preliminary Reactor Dimensions* for Selected Design Cases (Including Heat Transfer Capability in Primary Sorber and DSRP Reactor).

	Height, ft	Number of Heat Transfer Tubes	Pressure Vessel OD, ft	Wall Thickness, in
Two-Stage Desulfurization With SO₂ Regeneration (Case SO₂-D)				
Primary Sorber	9.0	0	10.1	2.0
Primary Regenerator	9.0	300	4.0	0.9
Secondary Sorber	9.0	0	9.3	1.9
Secondary Regenerator	9.0	0	2.5	0.6
Single-Stage Desulfurization With DSRP (Case DSRP-A)				
Sorber	9.0	0	9.2	1.8
Regenerator	9.0	0	7.8	1.6
DSRP Reactor	11.5	200	6.3	1.3

*based on heat transfer coefficient of 10 Btu/hrft²°F.

The largest vessel in the two-stage desulfurization with SO₂ regeneration case is the primary sorber with an outside diameter of 10.1 ft and height of 9 ft. The diameter of the secondary sorber is somewhat smaller because of the lower operating temperature. Both regenerators have considerably smaller diameters because of different operating temperatures and gas volumetric flow rates. The size of the single sorber used in single-stage desulfurization is comparable to that of the secondary sorber in the two-stage desulfurization case while the single regenerator is larger than either of the two-stage regenerators. Finally, the DSRP reactor has an intermediate diameter but greater height.

REFERENCES

- Chen, H.T., et al., 1991, Sensitivity Effects of Sulfur Control on IGCC Cost of Electricity, Task Assignment 2, Final Report, DE-AC21-88F361662.
- Gangwal, S.K., et al., 1993, Slipstream Testing of Hot Gas Desulfurization With Sulfur Recovery, Proceedings of the Advanced Coal-Fired Power Systems '95 Review Meeting, Vol. I, p. 220, DE-AC21-93MC30010.
- Peters, M.S., and Timmerhaus, K.D., 1991, Plant Design and Economics for Chemical Engineering, 4th ed., McGraw-Hill, New York.