DOE/MC/30012 -- 4021
(DE95000091)

Advanced Sulfur Control Concepts for Hot Gas Desulfurization Technology

Quarterly Report
October - December 1994

Douglas P. Harrison

January 1995

Work Performed Under Contract No.: DE-AC21-94MC30012

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
Morgantown, West Virginia

By
Louisiana State University
Baton Rouge, Louisiana

DISCLAIMER

DOE/MC/30012 -- 4021
(DE95000091)
Distribution Category UC-106

Advanced Sulfur Control Concepts for Hot Gas Desulfurization Technology

Quarterly Report
October - December 1994

Douglas P. Harrison

Work Performed Under Contract No.: DE-AC21-94MC30012

For
U.S. Department of Energy
Office of Fossil Energy
Morgantown Energy Technology Center
P.O. Box 880
Morgantown, West Virginia 26507-0880

By
Louisiana State University
Department of Chemical Engineering
Baton Rouge, Louisiana 70803

January 1995

EXECUTIVE SUMMARY

Progress during the past quarter was limited by delays in identifying an appropriate analytical instrument for measuring the concentration of sulfur species ($S_x(g)$, H_2S, and SO_2) in the regeneration product gas. The ability to carry out this analysis on a real-time basis is an important component of the overall project and we feel that a satisfactory gas analysis procedure should be available before forging ahead with other experimental activities.

The primary accomplishment, therefore, was the completion and submission of the Task 3 Project Plan. This plan, which assumed a satisfactory solution to sulfur analysis problem, is included in this quarterly report.

SULFUR ANALYSIS

Because the operation of a fixed-bed noncatalytic gas-solid reactor is intrinsically an unsteady-state process, the rate of total sulfur production during sorbent regeneration must vary with time. We also expect that the distribution of sulfur species in the regeneration product gas will be time dependent although there is no data to confirm this. In previous research involving the production of elemental sulfur during sorbent regeneration, a condenser was used to determine the total amount of sulfur produced during the duration of the regeneration test. More detailed analytical data is needed if we are to optimize elemental sulfur production.

In our original plan, the hot regenerator product gas was to be split with one portion being fed to a total oxidation unit where all sulfur would be converted to SO_2. The remainder of the regenerator product stream would be passed through a condenser where elemental sulfur would be removed leaving SO_2 and H_2S in the gas phase. IR analyzers would then provide a continuous

determination of SO_2 (equivalent to total sulfur) in the effluent from the oxidation unit, and H_2S and SO_2 in the effluent from the condenser. Elemental sulfur in the regenerator product would then be determined by difference.

Initial contacts with Perkin-Elmer led us to anticipate no problems with this analytical approach. As the discussions proceeded, however, we learned that IR cells capable of operating at the temperature and pressure of interest would cost about $5,000 each with no guarantee that the IR windows would not react with sulfur compounds.

Antek Instruments, a leader in sulfur analysis in the refining and petrochemical industries was then contacted. Professor Harrison visited Antek in early December to communicate our analytical needs and to learn about Antek's capabilities. The major problem is associated with the fact that the regenerator product gas is at high pressure; all of Antek's current instruments operate near atmospheric pressure. They use a quartz oxidation furnace operating at 800 - 1000°C to convert all sulfur species to SO_2. The quartz cannot operate at these temperatures and 15 atm pressure.

Two possible solutions were identified in the discussion. Antek is studying the use of a quartz capillary tube to expand the product gas from 15 atm to atmospheric pressure. The thick-walled capillary could presumably withstand the high temperature, high pressure combination. The product gas at atmospheric pressure would then be fed to the standard quartz oxidation chamber. The key is determining the proper combination of capillary diameter and length to achieve the required pressure drop and the gas volumetric flow rate required for analysis. The other option involves the substitution of an alonized stainless steel oxidation furnace for the

quartz furnace and carrying out the sulfur oxidation at 15 atm. Antek now prefers to work with the quartz capillary and the alonized steel option is being held in reserve.

CONDENSER OPERATING CONDITIONS

That portion of the regenerator product gas not fed to the oxidation chamber will be fed to a condenser where elemental sulfur will be condensed and separated from the permanent gases. Equilibrium sulfur condensation calculations have been made to assist in the design and operation of the condenser. The free energy minimization program CHEMQ, although primarily designed for chemical reaction equilibrium calculations, was also used for this physical equilibrium analysis. Results in the form of the temperatures required to achieve initial, 95%, and 99% sulfur condensation as a function of the initial sulfur content of the regenerator product gas and the operating pressure are presented in Figures 1 through 6, with each figure corresponding to a different initial sulfur content.

The initial mixture consisted of 1 mol S_2 and x mols of N_2, with x ranging from 0.1 for 95% sulfur to 98 for 2% sulfur. No compounds which could participate in chemical reactions such as H_2S, SO_2, O_2 or H_2O were permitted based upon the assumption that the kinetics of the chemical reactions would be negligibly slow at condenser operating temperatures. This assumption should be valid except, perhaps, at the largest sulfur contents.

If the regeneration product gas at 15 atm contains 40 mol% elemental sulfur (for example, 1 mol of S_2 per 3 mols of other gases), we see from Figure 3 that sulfur condensation would begin at 760K (487°C, 908°F), that 95% of the sulfur would be condensed at 580K (307°C, 585°F), and that 99% sulfur condensation would be expected at 520K (247°C, 477°F). This

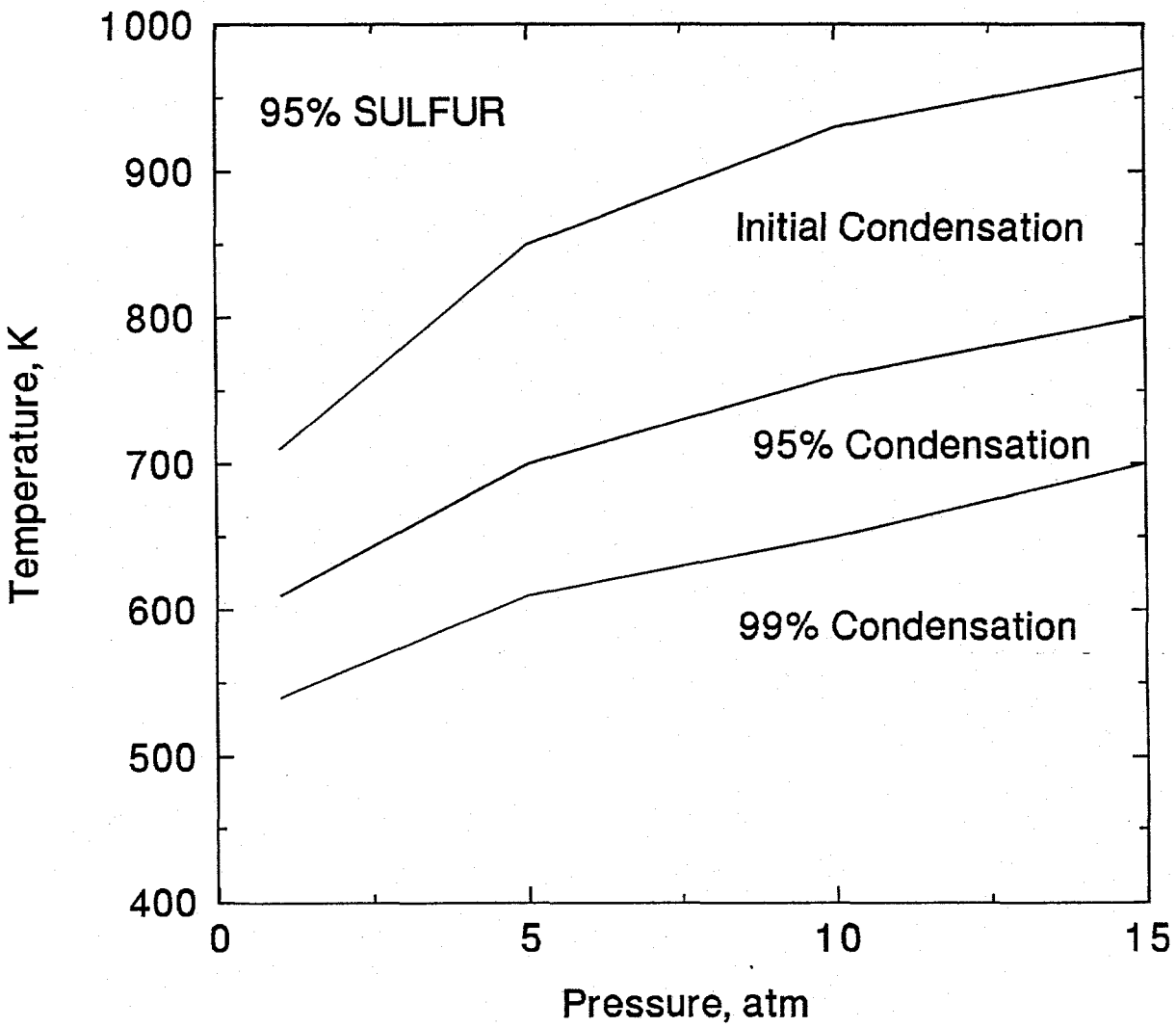


Figure 1. Elemental Sulfur Condensation Temperature as a Function of Pressure: 95% Sulfur in Inlet Gas.

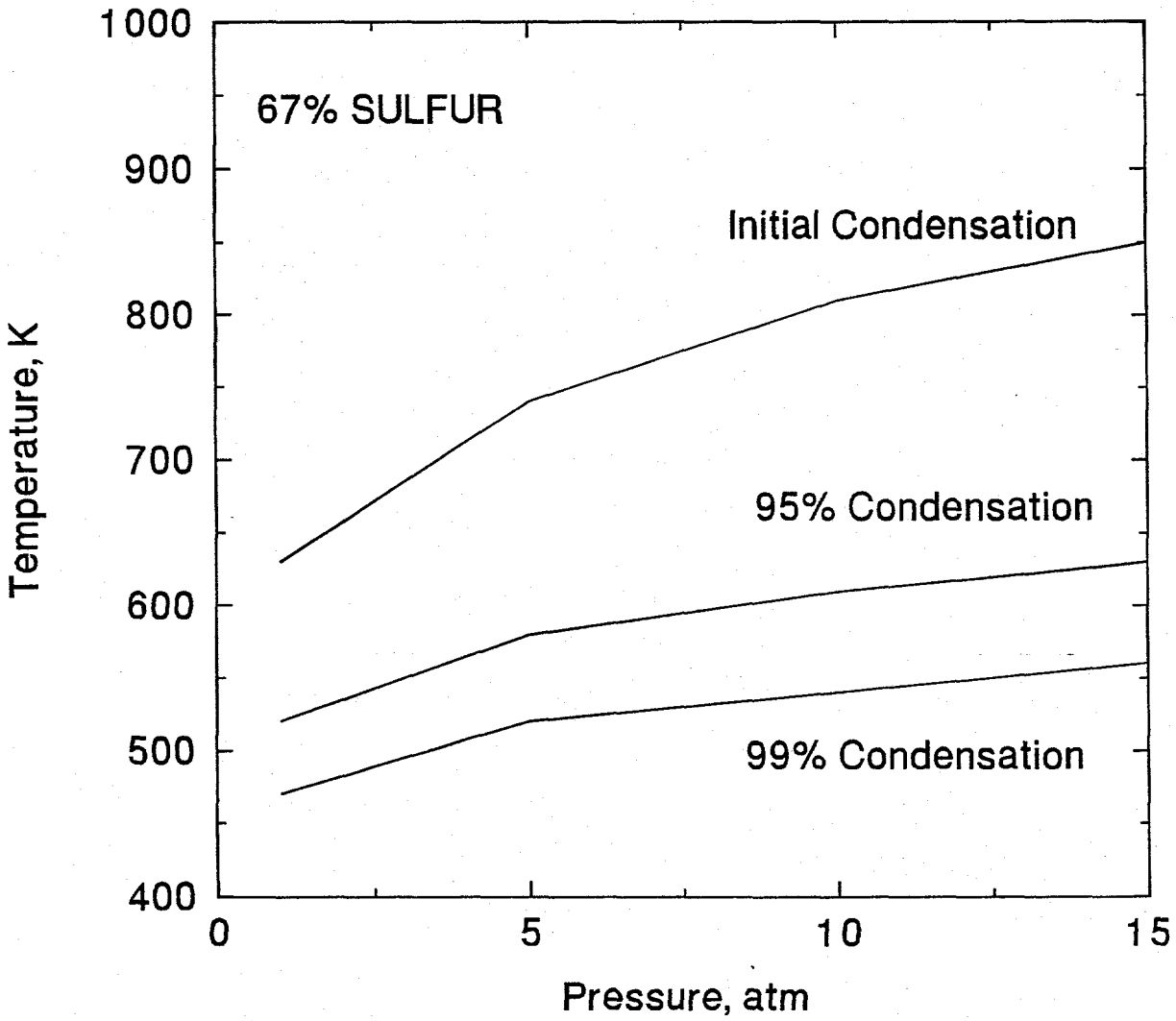


Figure 2. Elemental Sulfur Condensation Temperature as a Function of Pressure: 67% Sulfur in Inlet Gas.

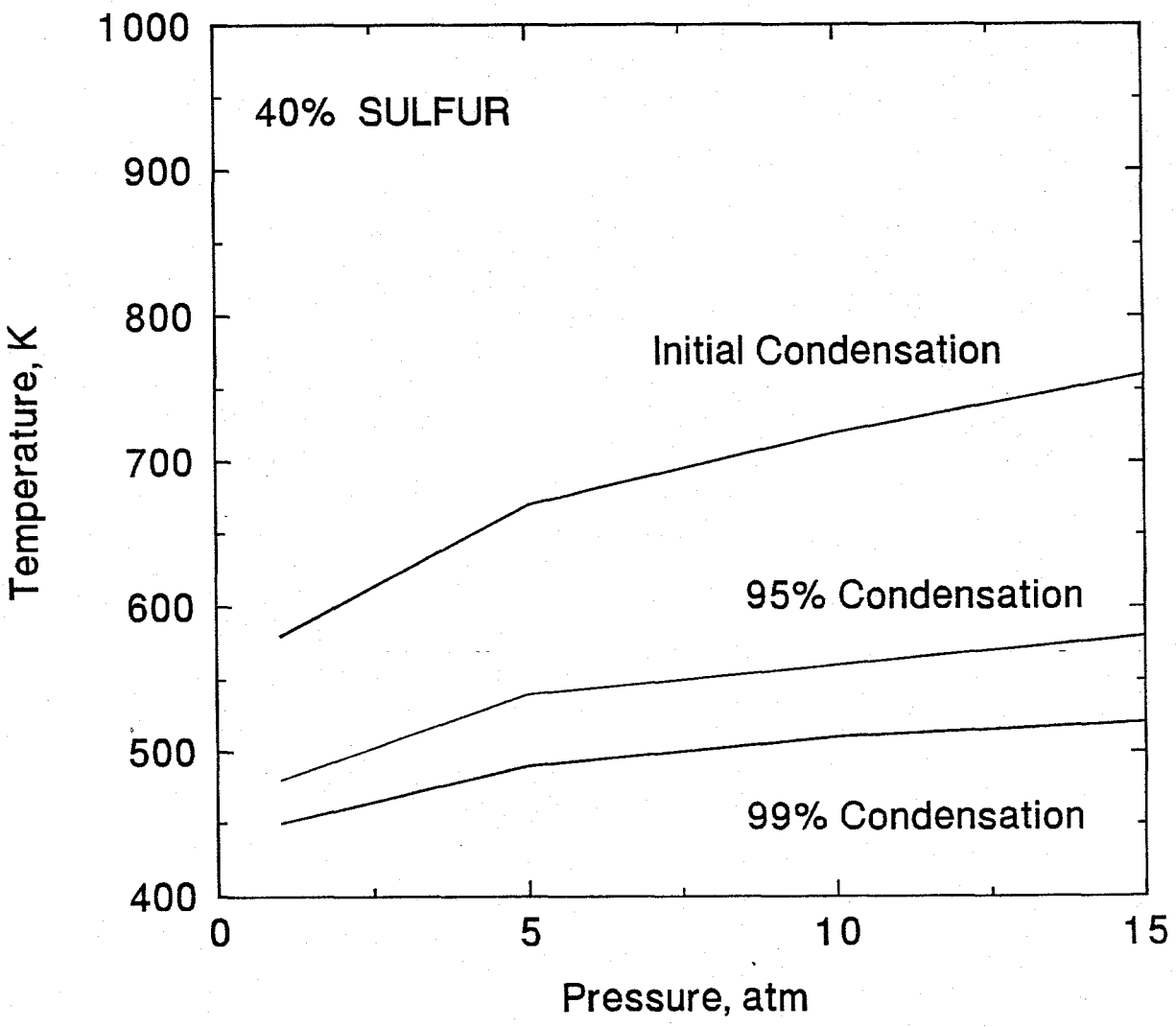


Figure 3. Elemental Sulfur Condensation Temperature as a Function of Pressure: 40% Sulfur in Inlet Gas.

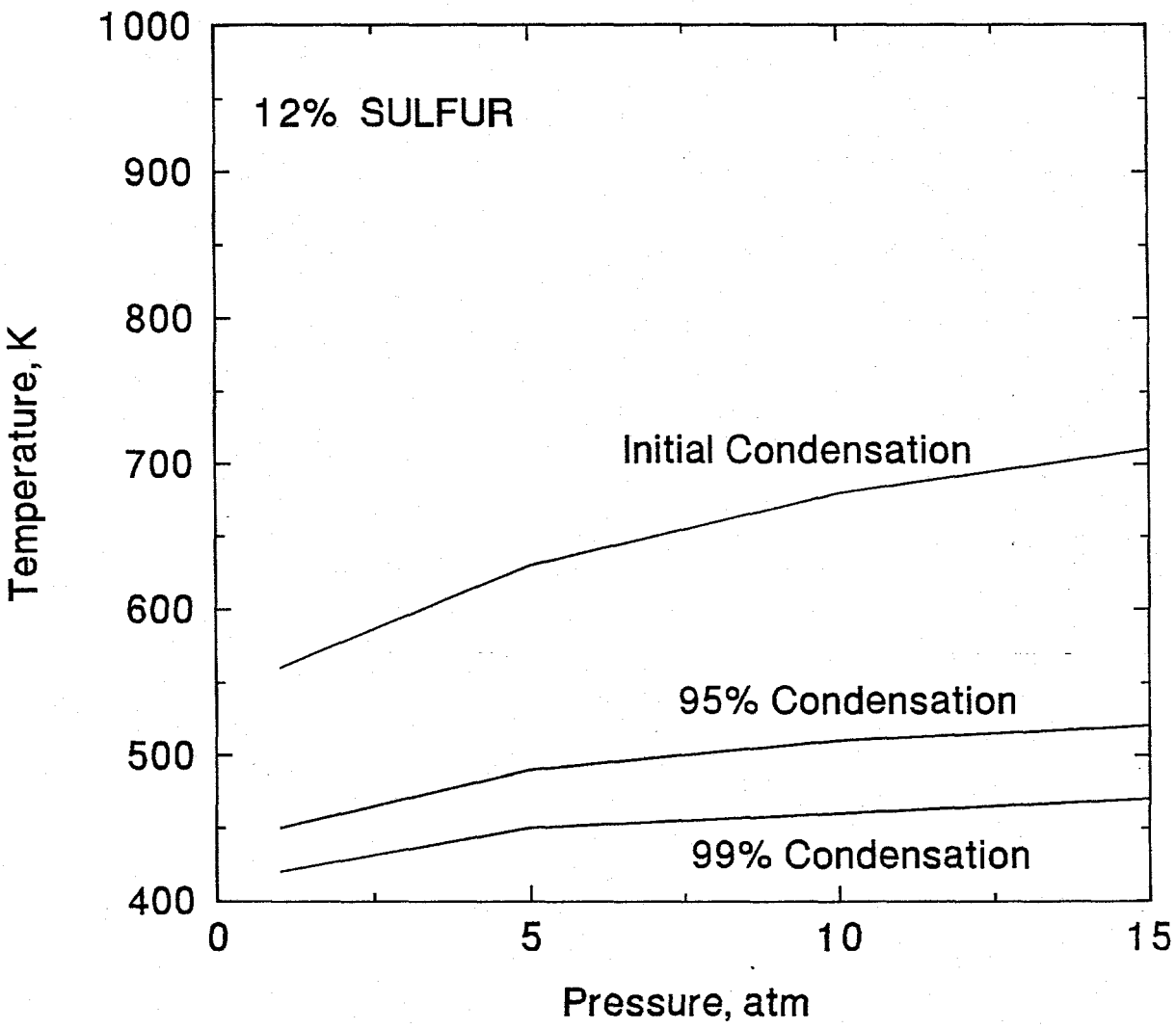


Figure 4. Elemental Sulfur Condensation Temperature as a Function of Pressure: 12% Sulfur in Inlet Gas.

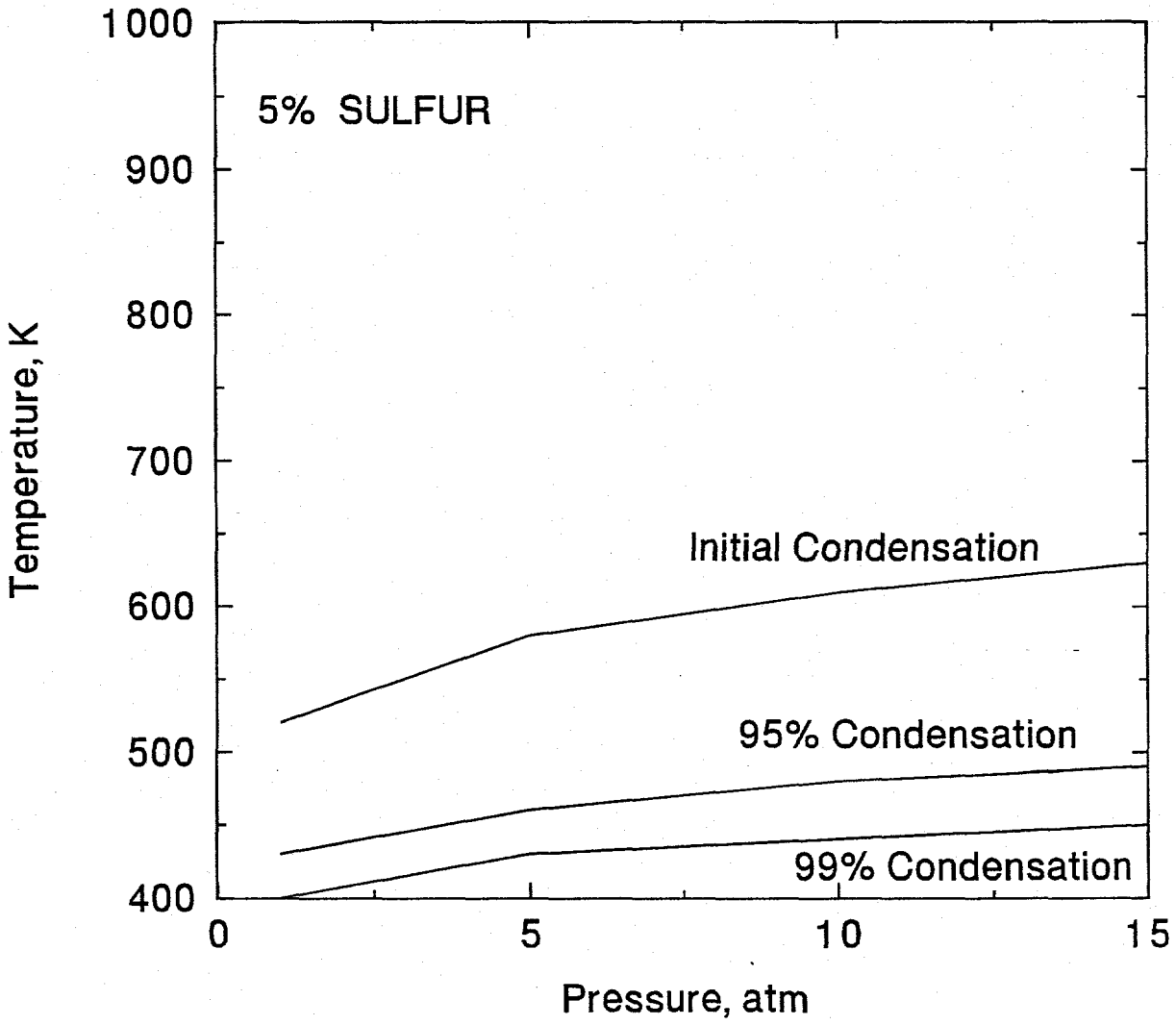


Figure 5. Elemental Sulfur Condensation Temperature as a Function of Pressure: 5% Sulfur in Inlet Gas.

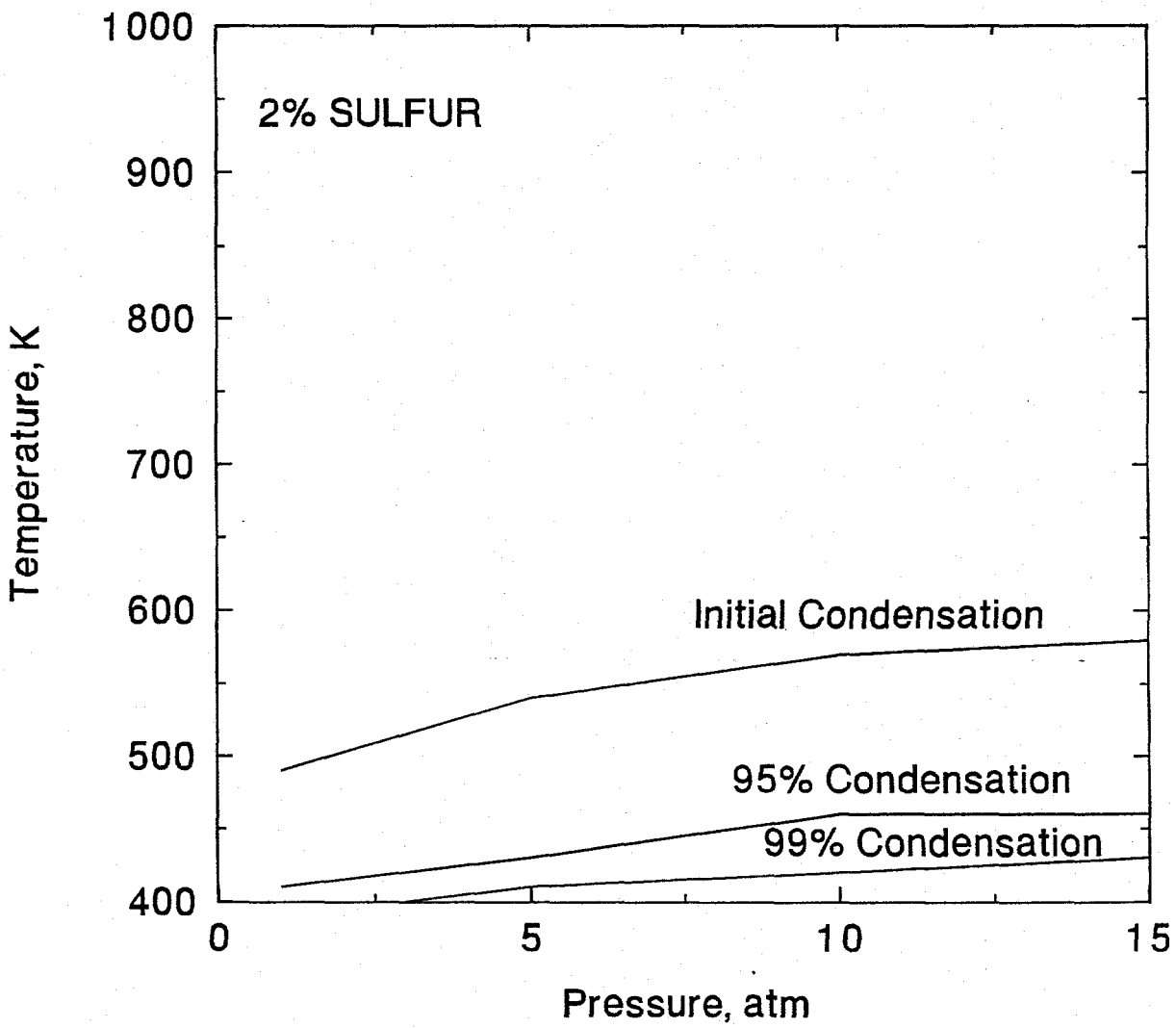


Figure 6. Elemental Sulfur Condensation Temperature as a Function of Pressure: 2% Sulfur in Inlet Gas.

means that the temperature of the lines leading to the condenser would have to be maintained above 760K while essentially complete sulfur condensation should be possible if the condenser is operated slightly below 520K.

If the sulfur content of the regenerator product gas is lower, say 5% elemental sulfur, we see from Figure 5 that the temperatures corresponding to initial, 95%, and 99% sulfur condensation are 630K (357°C, 675°F), 490K (217°C, 423°F), and 450K (177°C, 350°F), respectively. A lower operating pressure of, say, 5 atm would reduce the temperatures to 580K (307°C, 585°F) for initial condensation and to 430K (157°C, 315°F) for 99% condensation.

In the typical regenerator product gas, steam would be the next component to condense after elemental sulfur. Simultaneous condensation of steam and sulfur could occur at low sulfur contents if the remainder of the regeneration product gas is primarily steam. For example, the minimum overall temperature of 390K (117°C, 243°F) is shown in Figure 6 at 2% sulfur and 1 atm. The vapor pressure of steam at this temperature is about 1.8 atm which is near the operating pressure. At 15 atm operating pressure, 99% condensation should occur at 430°C (157°C, 315°F). The vapor pressure of steam at this temperature is 5.7 atm which is below the operating pressure.

Task 3 - PROJECT PLAN

The attached project plan for the initial experimental phase was submitted with the October monthly report, and is repeated in the following. Minor changes have been made in the wording and in certain figures for purposes of improved clarity.

The Direct Production of Elemental Sulfur During

Sorbent Regeneration

Task 3 Project Plan

Introduction

Three possible concepts for the direct production of elemental sulfur during the regeneration of high temperature desulfurization sorbents have been identified. The concepts are based upon partial oxidation in an O_2-starved environment, reaction with SO_2, and reaction with H_2O. A detailed description of these concepts and the results of a thermodynamic analysis to evaluate the feasibility of each concept using a number of known desulfurization sorbents have been presented in a recent topical report (Lopez et al. 1994).

The thermodynamic analysis identified three sorbent systems based upon the oxides of iron, tin, and cerium which show promise for elemental sulfur production. We propose to study Fe_2O_3 and CeO_2 sorbents in the experimental program. Although the desulfurization and regeneration characteristics of SnO_2 and CeO_2 are thermodynamically similar, we recommend that the experimental work utilize CeO_2 for a number of reasons. First, desulfurization and regeneration studies using SnO_2 are being carried out in other laboratories (Copeland, 1993). In contrast, CeO_2 has received relatively little attention. Importantly, the allowable operating window appears to be wider for CeO_2 than SnO_2. In the desulfurization cycle, there is a danger of reducing $SnO_2(s)$ to $Sn(\ell)$ at severe conditions. Tin sulfide, SnS(s), has an appreciable vapor pressure which imposes further limits on operating conditions. Neither of these problems exist in the cerium system. $CeO_2(s)$ is more stable than $SnO_2(s)$, and the sulfided product, $Ce_2O_2S(s)$, is not volatile.

The thermodynamics of sulfur production using an iron-based sorbent are less favorable, but past experimental results using the partial oxidation concept have shown that significant amounts of elemental sulfur can be produced. Experimental effort on the iron system will be limited to the partial oxidation regeneration concept while we propose to examine all three regeneration concepts using cerium sorbents.

Chemical Reactions

Although the primary objective of the research is the production of elemental sulfur during the sorbent regeneration phase, it will be necessary in some tests to carry out experiments for the complete cycle consisting of reduction/sulfidation as well as regeneration.

The thermodynamic analysis indicates that Fe_2O_3 will be reduced to Fe_3O_4 at most desulfurization conditions of interest, and that Fe_3O_4 will react with H_2S and H_2 to form FeS. These reactions may be written as

$$3Fe_2O_3 + H_2 \rightarrow 2Fe_3O_4 + H_2O, \text{ and} \quad (1)$$

$$Fe_3O_4 + 3H_2S + H_2 \rightarrow 3FeS + 4H_2O. \quad (2)$$

CeO_2 is stable in the typical coal gas atmosphere and the probable desulfurization reaction is

$$2CeO_2 + H_2S + H_2 \rightarrow Ce_2O_2S + 2H_2O. \quad (3)$$

Therefore, the sulfided forms of the sorbents are likely to be FeS and Ce_2O_2S.

We propose to study the regeneration of FeS using the partial oxidation concept in which FeS is reacted with a mixture of O_2 and H_2O operated in an O_2-starved manner. The ideal reaction at these conditions is

$$2FeS + \frac{3}{2}O_2 \rightarrow Fe_2O_3 + S_2. \quad (4)$$

However, it is likely that a number of reactions including those listed in Table I will occur simultaneously. The basic approach will be to identify reaction conditions which maximize the rates of those reactions which produce elemental sulfur relative to those reactions which produce SO_2.

The approach to be used to study the partial oxidation of Ce_2O_2S will be similar. The ideal reaction is

$$Ce_2O_2S + O_2 \rightarrow 2CeO_2 + \frac{1}{2}S_2 \quad (5)$$

although a number of simultaneous gas-solid and gas phase reactions analogous to those listed in Table I will likely occur.

The stoichiometry of the regeneration of Ce_2O_2S with SO_2 should be cleaner, and should proceed in the following manner

$$Ce_2O_2S + SO_2 \rightarrow 2CeO_2 + S_2. \quad (6)$$

Similarly, it should be possible to reverse the desulfurization reaction (3) and liberate substantial concentrations of H_2S by reacting Ce_2O_2S with H_2O

Table I. Simultaneous Reactions Involved in the Partial Oxidation of FeS to Produce Elemental Sulfur

Gas - Solid Reactions

1. $2FeS(s) + \frac{3}{2}O_2(g) \rightarrow Fe_2O_3(s) + S_2(g)$

2. $2FeS(s) + \frac{7}{2}O_2(g) \rightarrow Fe_2O_3(s) + 2SO_2(g)$

3. $3FeS(s) + 4H_2O(g) \rightarrow Fe_3O_4(s) + 3H_2S(g) + H_2(g)$

4. $3FeS(s) + 2SO_2(g) \rightarrow Fe_3O_4(s) + \frac{5}{2}S_2(g)$

5. $2Fe_3O_4(s) + \frac{1}{2}O_2(g) \rightarrow 3Fe_2O_3(s)$

Gas-Phase Reactions

6. $SO_2(g) + 2H_2S(g) \rightarrow 2H_2O(g) + \frac{3}{2}S_2(g)$

7. $H_2S(g) \rightarrow H_2(g) + \frac{1}{2}S_2(g)$

8. $SO_2(g) + 2H_2(g) \rightarrow 2H_2O(g) + \frac{1}{2}S_2(g)$

9. $S_2(g) + 2O_2(g) \rightarrow 2SO_2(g)$

$$Ce_2O_2S + 2H_2O \rightarrow 2CeO_2 + H_2S + H_2. \quad (7)$$

Although H_2S and not elemental sulfur is produced in this reaction, it is of interest since existing processes are available to convert high concentrations of H_2S into elemental sulfur. The objective in studying reactions (6) and (7) will be to identify reaction conditions which maximize the concentrations of elemental sulfur or H_2S in the product gas.

Reactor Systems

We propose to use three reactor systems to study various aspects of the problem. Two electrobalance reactors, one capable of high pressure operation and the other limited to atmospheric pressure, will be used to study the kinetics of single gas-solid reactions involving iron. A laboratory-scale fixed-bed reactor system will be used for all cerium studies and for iron studies involving multiple simultaneous reactions. The progress of the reaction in an electrobalance is followed by monitoring the solid weight. Unfortunately, the molecular weights of $2CeO_2$ and Ce_2O_2S are identical which means that all cerium studies must use the fixed-bed reactor where product gas analysis is used to follow the progress of the reaction. The basic components of all three reactor systems are currently available although modifications will be required to enable us to include corrosive sulfur compounds in the reacting gases. For example, all high temperature wetted surfaces will be alonized.

A schematic diagram of the fixed-bed reactor system is shown in Figure 1 while details of the reactor are presented in Figure 2. The temperature and pressure limits are approximately 900°C and 15 atm. Permanent gases are obtained from high pressure cylinders and flow is

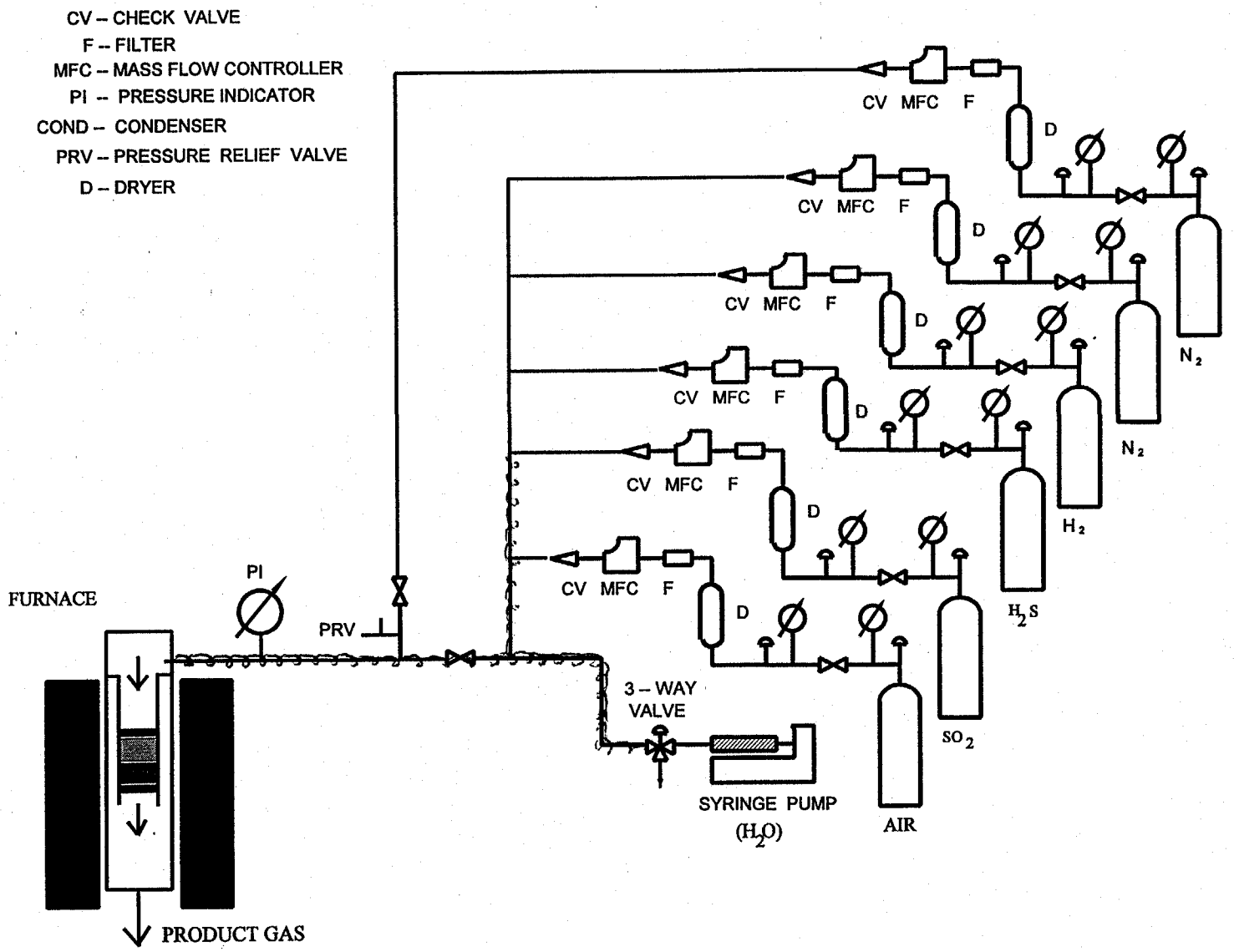


Figure 1. Fixed-Bed Reactor System.

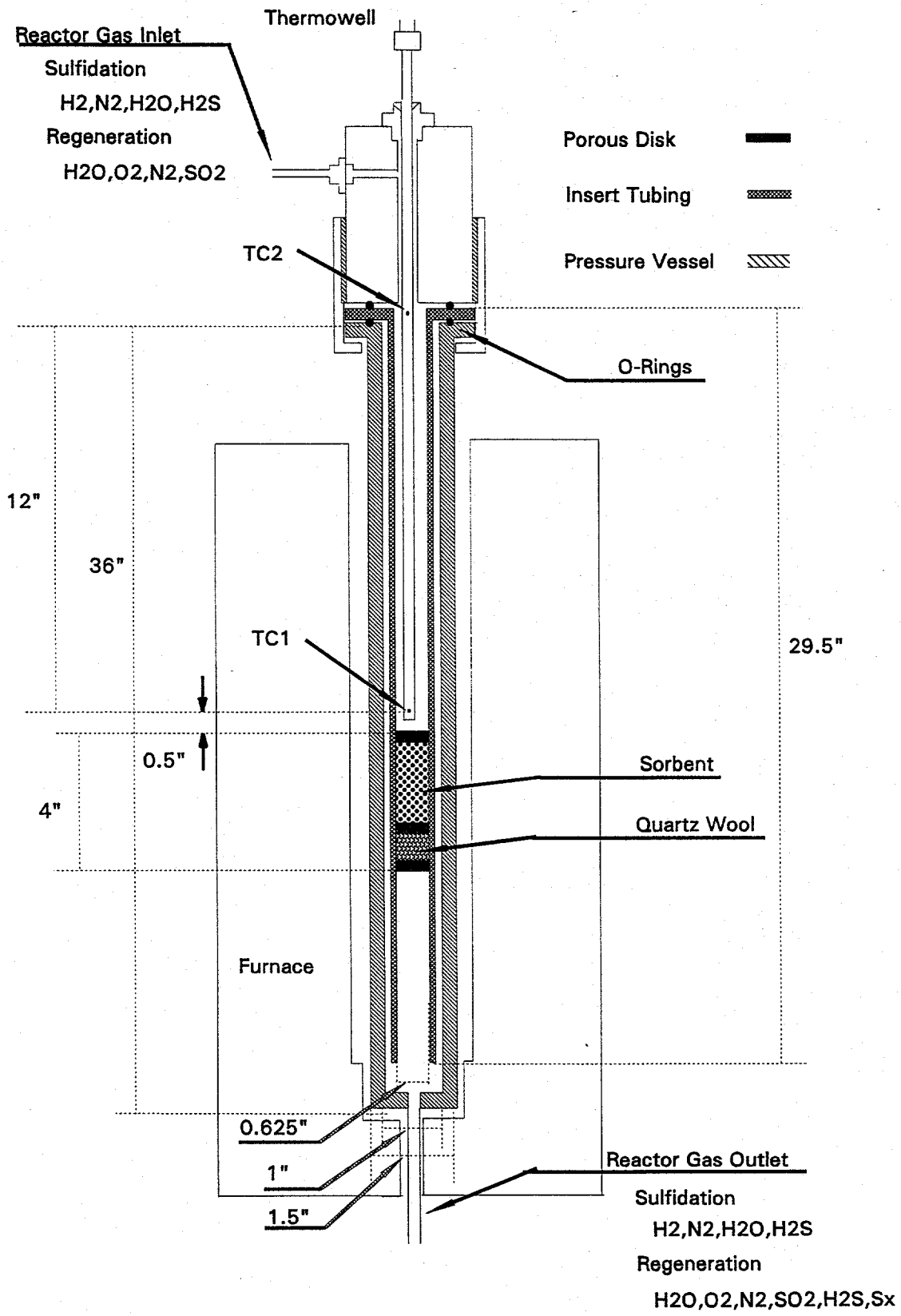


Figure 2. Details of the Fixed-Bed Reactor.

controlled using high pressure mass flow controllers. H_2O is metered as a liquid using a high pressure syringe pump and is vaporized as it mixes with the permanent gases. Feed gases during the sulfidation cycle will consist of H_2, H_2O, H_2S, and N_2. Different feed gases will be used for each regeneration concept. For partial oxidation, the feed gas will contain O_2, N_2, and H_2O. SO_2 and N_2 will be fed to study regeneration with SO_2, and H_2O and N_2 will be fed when studying steam regeneration.

We propose to omit CO and CO_2 from the sulfidation mixture for a number of reasons. Primary emphasis in this study is on the regeneration step, and the experimental procedure will be simplified if the number of sulfidation feed components is reduced. The ultimate sulfur removal capability of the test sorbents as well as the sulfur material balance closure can be determined just as well in the absence of CO and CO_2, and the problem of carbon deposition is avoided.

A schematic of the high pressure electrobalance reactor system is shown in Figure 3. The feed system is effectively identical to that used in the fixed-bed system as are the operating temperature and pressure limits. The operating principle of the low pressure and high pressure electrobalances are the same, but the low pressure system is encased in glass and quartz while the high pressure system is contained within a stainless steel housing and hang-down tube. We plan to use rotameters and needle valves to control feed gas flow rates on the low pressure reactor instead of the more expensive mass flow controllers.

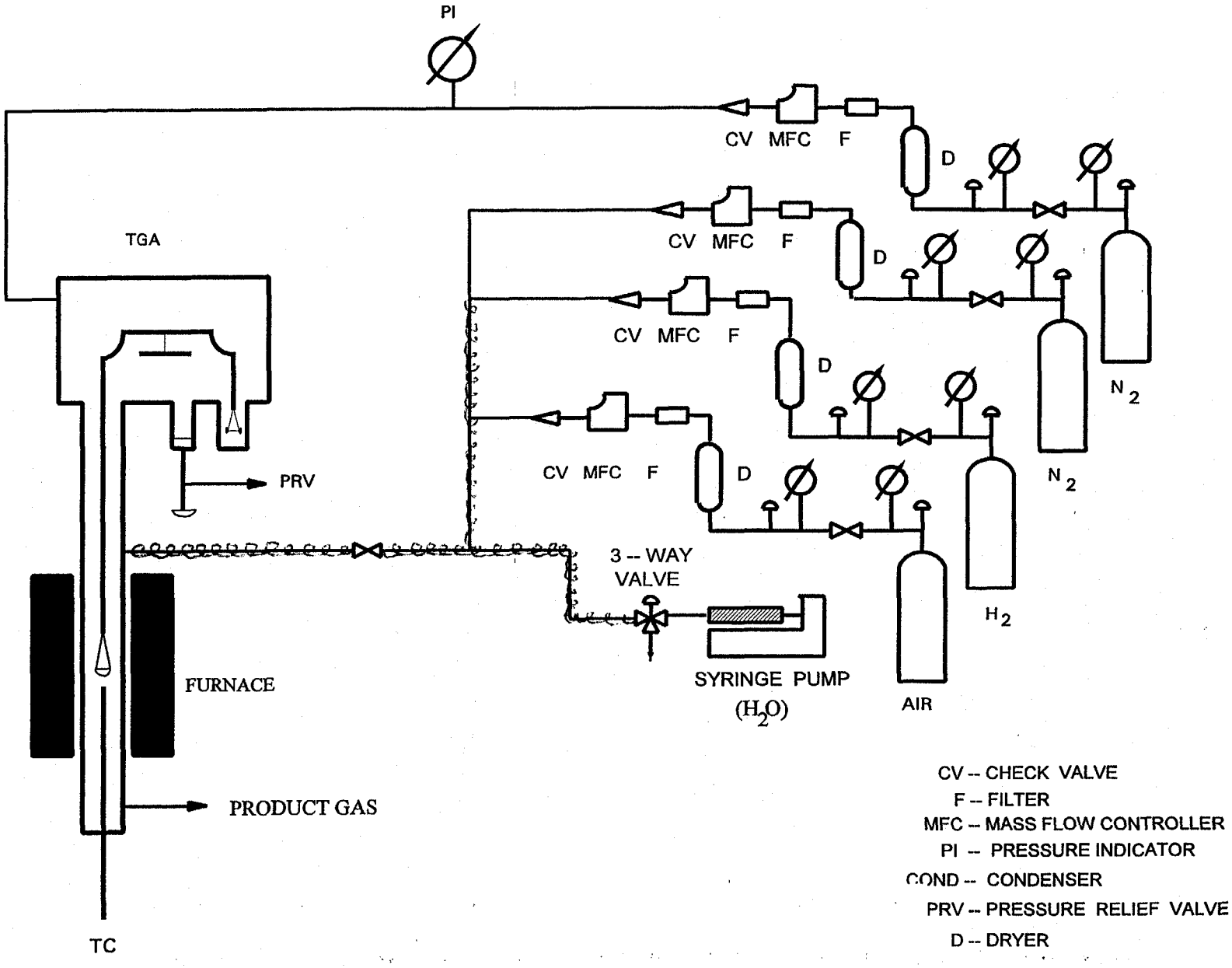


Figure 3. The Electrobalance Reactor System

PRODUCT GAS ANALYSIS

Analysis of the concentrations of sulfur species in the product gas from the fixed-bed reactor is a crucial component of the project. It is important to determine relative concentrations of SO_2, H_2S, and S_x (elemental sulfur) as a function of time. In previous regeneration studies involving the production of elemental sulfur, only time average quantities of elemental sulfur have been determined. Since fixed-bed regeneration is intrinsically an unsteady-state process, this is an important limitation which we propose to overcome.

The analytical method to be used has not been finalized at this time. Our original plans called for IR analysis but the vendors have not been able to assure us that the IR windows would not react with the sulfur compounds. IR is used for analysis of trace sulfur compounds, but we are anticipating much higher concentrations and temperatures than normally encountered.

An alternate scheme currently being considered is shown in Figure 4. Regeneration gas product is shown at typical conditions of 15 atm and 600°C. A slip stream will be directed to an Antek Model 7000S total sulfur analyzer. In this unit all sulfur is oxidized at 1100°C to SO_2 which is then analyzed using pyro-fluorescence. The analytical range is said to be from low ppb to 40% sulfur. Elemental sulfur in the main product stream will be removed in a condenser operating at approximately 15 atm in the temperature range of 150 - 300°C. The noncondensible gases will pass through a sampling valve and samples will be periodically directed to a GC equipped with a chemiluminescent detector for separate determination of H_2S and SO_2.

At the time this is written, we are awaiting a response from Antek Instruments, Inc., concerning the feasibility of this scheme or their suggestion of an alternate approach. Antek is definitely one of the leaders, if not the absolute leader, in instrumentation for sulfur analysis.

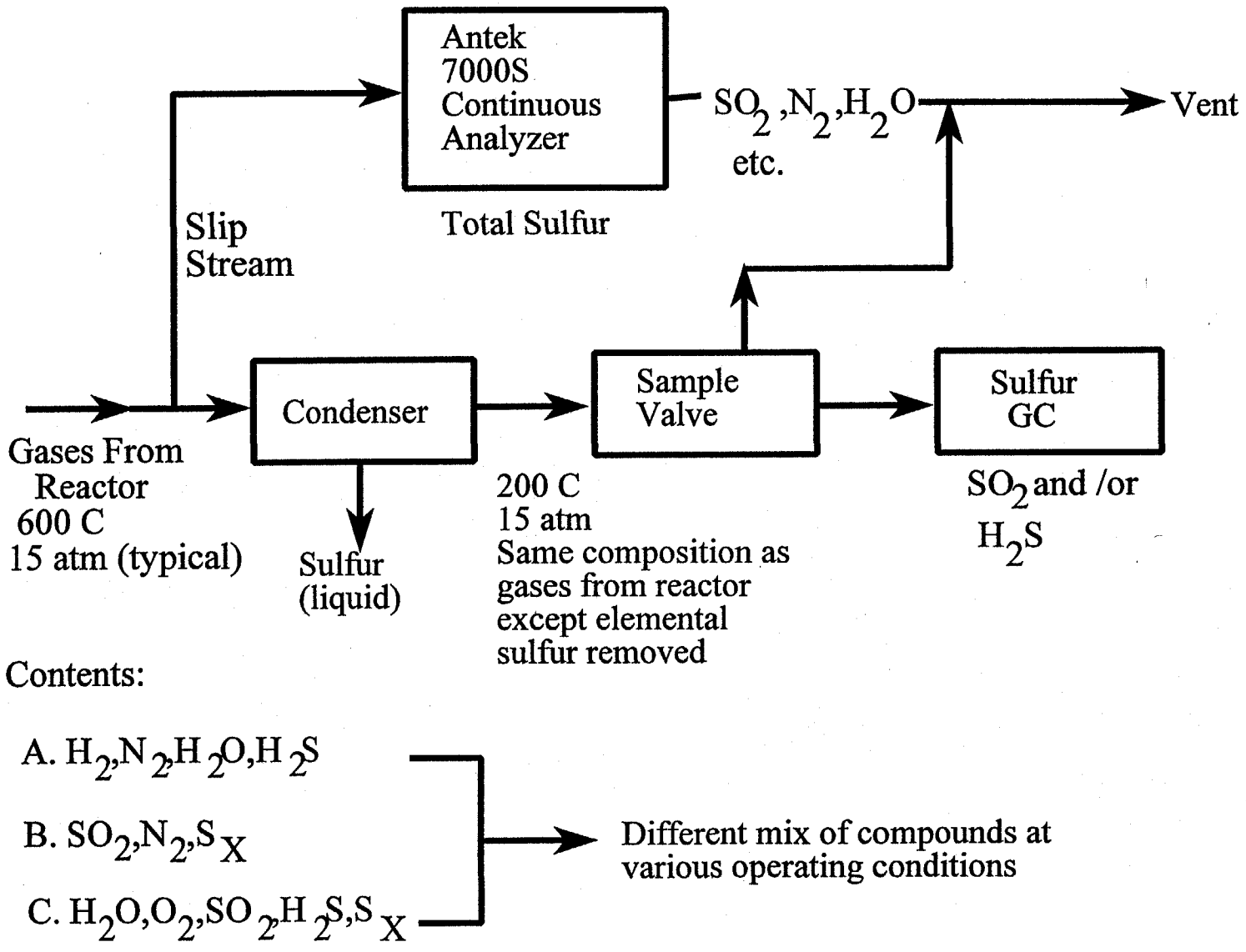


Figure 4. Proposed Sulfur Analysis System

Test Sorbents

As previously discussed, we propose to study iron- and cerium-based sorbents. In task 3.2, we consider that the primary objective is to prove the feasibility of elemental sulfur production. Development and optimization of a commercial sorbent will be addressed at a later time. For this reason we propose to utilize pure chemicals purchased from chemical supply houses in task 3.2. Iron oxide (Fe_2O_3, Fe_3O_4, and/or FeO), iron sulfide (FeS), and cerium oxide (CeO_2) are all available for direct purchase, but, to the best of our knowledge, cerium oxysulfide cannot be purchased directly.

Since the desulfurization ability of iron oxide is generally well-known and accepted, many of the iron tests will be limited to the regeneration phase and will utilize purchased FeS.

Both the sulfidation and regeneration phases of the reactions involving cerium are of interest. Less is known about the desulfurization capability of CeO_2, and there is uncertainty regarding the free energy values of Ce_2O_2S. For these reasons, and because Ce_2O_2S cannot be purchased, all cerium tests will begin with purchased CeO_2 and will involve both the sulfidation and regeneration phases.

Reaction Parameters

The reaction parameters to be studied are temperature, feed gas composition, reactor space velocity, and, to a lesser extent, pressure. Proposed base values and ranges of the parameters to be studied in each phase of the research are presented in Table II. Pressure and space velocity ranges are common to all tests while temperature and gas composition vary depending on the reaction phase being studied.

Table II. Reaction Parameters: Base Values and Ranges to be Studied

Parameter Values Common to all Reactions	Base Conditions	Range of Conditions
Pressure, atm	15	1 - 15
Space Velocity (fixed-bed reactor), hr^{-1} (STP)	2000	1000 - 5000
Sulfidation		
Gas Comp., % H_2S	1	1
H_2	30	20 - 40
H_2O	15	10 - 20
N_2	54	balance
Temperature, °C		
Iron Sorbent	600	500 - 750
Cerium Sorbent	800	700 - 900
Regeneration		
Partial Oxidation (Iron and Cerium Sorbents)		
Gas Comp., % O_2	1	0.25 to 3
H_2O	20	0 to 40
N_2	79	balance
Temperature, °C	600	500 - 800
Regeneration With SO_2 (Cerium Sorbent only)		
Gas Comp., % SO_2	25	5 - 50
N_2	75	balance
Temperature, °C	600	500 - 800
Regeneration With Steam (Cerium Sorbent only)		
Gas Comp., % H_2O	25	5 - 50
N_2	75	balance
Temperature, °C	600	500 - 800

The base pressure is designated as 15 atm in all cases. The thermodynamics of the sulfidation and regeneration reactions are only mildly pressure dependent while the kinetics are favored by high pressure. High pressure operation will be required to integrate the desulfurization steps with the remainder of the IGCC process. Consequently, most sulfidation and regeneration tests will be conducted at 15 atm. Exceptions will be the initial scoping tests using the low pressure electrobalance reactor and a limited number of tests carried out specifically to examine the sensitivity of the reactions to pressure.

The base value and range of space velocity proposed to be used in fixed-bed tests are based on recent experience with another reaction system using the same fixed bed reactor. From a practical standpoint, we wish to operate at the largest possible space velocity which is consistent with the global reaction rates being sufficiently rapid to permit equilibrium conditions to be approached.

Proposed sulfidation feed gas compositions omit CO and CO_2 in order to simplify the experimental tests and to avoid the problem of carbon deposition. The reducing power of the base feed gas, which is established by the H_2:H_2O ratio of 2, is reasonably close to the reducing power of a Texaco gas. The proposed range of H_2 and H_2O concentrations will result in a variation of H_2:H_2O ratios between 1 and 4.

Base sulfidation temperatures and temperature ranges are based upon results of the thermodynamic analysis. The proposed range of iron oxide temperatures is lower because of the tendency for iron oxide reduction, and because the H_2S removal ability of iron oxide decreases with increasing temperature. CeO_2, in contrast, is not easily reduced and its H_2S removal capability increases with increasing temperature.

The proposed base regeneration temperature and temperature ranges using both iron and cerium sorbents are the same for all regeneration concepts (iron studies will be limited to the partial oxidation concept). Once again, these proposed temperatures are based upon the thermodynamic analysis.

Gas compositions for the partial oxidation regeneration studies involve low oxygen concentrations and, in most cases, a large ratio of H_2O to O_2. Operation in an "O_2-starved" mode is believed to be important in maximizing the yield of elemental sulfur, while the reaction between the sorbent and H_2O is believed to be important in producing H_2S for reacting with SO_2 and increasing the elemental sulfur yield via the Claus reaction.

Regeneration with SO_2 should, according to thermodynamics, proceed cleanly with elemental sulfur being the only significant gaseous product. The objective, therefore, will be to determine the SO_2 content which maximizes the ratio of S_x:SO_2 in the product gas and is consistent with an acceptable global reaction rate.

Regeneration with steam represents a direct reversal of the sulfidation reaction and the product is H_2S instead of elemental sulfur. The reaction should proceed cleanly with H_2S as the only significant gaseous sulfur product. The objective, therefore, will be equivalent to the objective of the SO_2 regeneration studies. We will seek to determine the H_2O content which maximizes the H_2S:H_2O ratio in the product gas and is consistent with an acceptable global reaction rate.

Sulfur Material Balances

Closing the sulfur material balance is an important component of the study. Sulfur feed rates are known from the rate and composition of the reactor feed. Thus the total amount of sulfur fed during a test is simply the product of the sulfur feed rate and the duration of the test. Sulfur will be present in the feed gas only during sulfidation and regeneration tests involving the SO_2 regeneration concept. The quantity of sulfur present in the reactor at the beginning of a test (if any) can be calculated from the known mass and composition of sorbent. Total sulfur content, as well as sulfur species concentrations, in the reactor product gas will be monitored as a function of time, and the total sulfur leaving in the product gas can be determined by integrating the product gas composition over the duration of the test.

Weighing the total quantity of elemental sulfur collected in the condenser at the conclusion of a test provides an additional check on the sulfur material balance. The accuracy of this measurement will be limited, however, since the mass of sulfur condensed is likely to be small compared to the mass of the condenser.

By using a similar material balance approach in another reaction system, we were typically able to close the total carbon material balance to within ± 10%. We hope to be able to achieve a similar closure for the sulfur material balance, but we recognize that the analytical problems are more difficult.

Sorbent Characterization

Previous experience on a number of related research projects has shown that sorbent characterization tests provide valuable information to supplement the kinetics results. Structural

properties such as specific surface area, pore volume, and pore size distribution are generally different for the sulfided and oxidized sorbent forms. Such properties may also vary with time because of high temperature sintering. X-ray diffraction studies are useful in identifying specific crystalline phases in the sorbent. Scanning electron microscopy may be used to map the spatial distribution of sulfur species within individual particles, as well as to provide visual images of the sorbent particles in oxidized and sulfided forms.

Instruments required to accomplish all of these characterization measurements are available in departmental laboratories, and will be utilized on an as-needed basis.

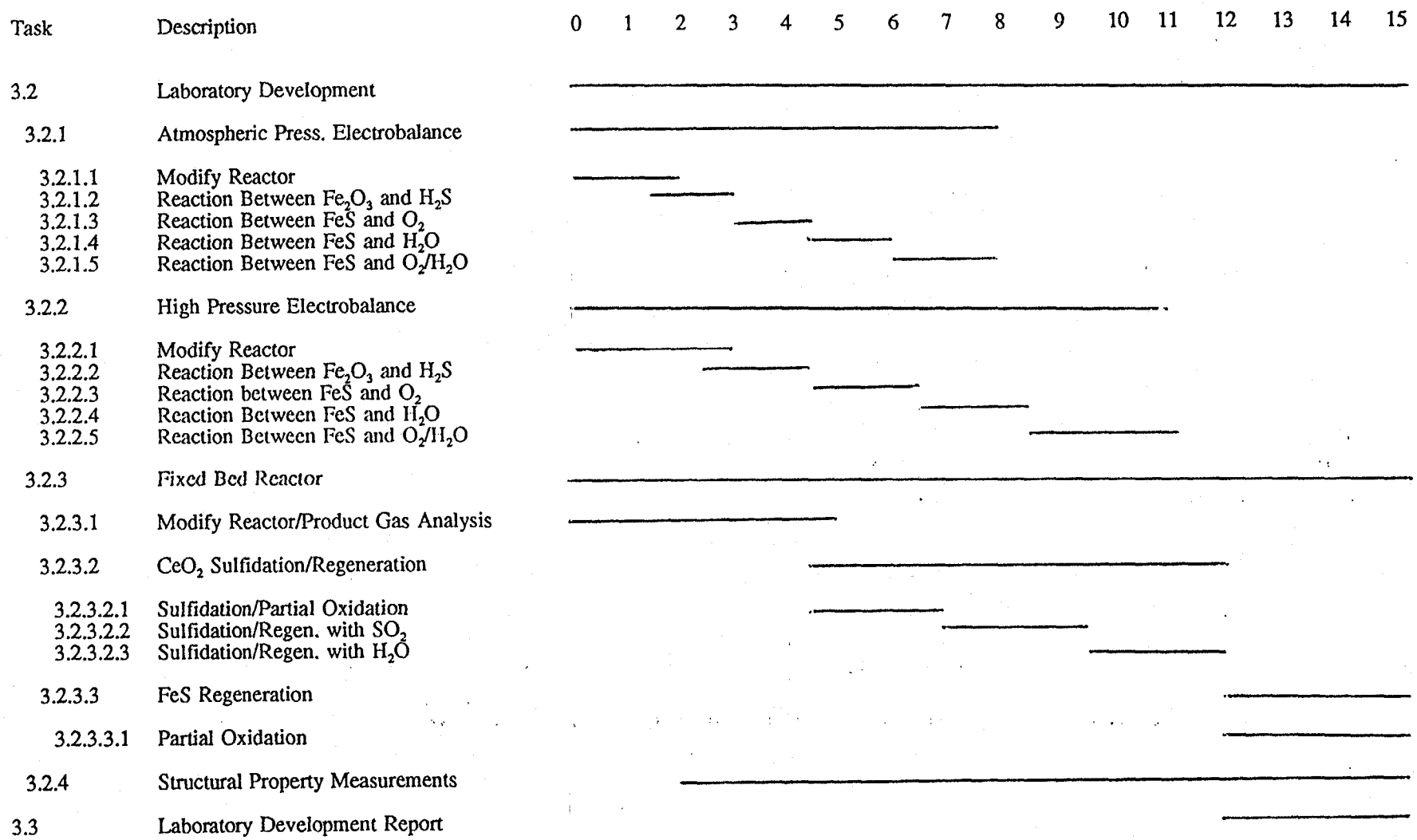
MILESTONE SCHEDULE

A detailed milestone schedule for achieving the objectives of Task 3.2 is presented in Figure 5. We propose a 15 month project period to accomplish the experimental goals and to complete the Laboratory Development Report of Task 3.3. Four major subtasks involving each of the three reactor types as well as structural property measurements are identified.

Work on modifying and commissioning the three reactors will be initiated immediately after the project plan is approved. The estimated time required to accomplish the modifications to the atmospheric electrobalance is 1½ to 2 months. An additional month is allocated for high pressure electrobalance modifications, while the time required for the fixed-bed reactor modifications and commissioning is estimated to be 5 months. Time for perfecting the product gas analytical procedure is included in the 5-month estimate.

Atmospheric pressure electrobalance tests will begin as modifications to the reactor are completed, and approximately 1½ months is allocated to each of the four groups of atmospheric

Figure 5. Milestone Schedule



pressure tests involving the iron sorbent system. Recall that no electrobalance tests using the cerium sorbent system are planned since the primary sulfidation and regeneration reactions produce no change in the solid mass. A primary objective of the atmospheric pressure tests will be to determine if temperature and concentration conditions can be identified where the rates of the FeS-O_2 and FeS-H_2O reactions are approximately equal. We anticipate little, if any, sulfur formation from the FeS-O_2 reaction since it is not feasible to operate the electrobalance in the O_2-starved mode. However, knowledge of the relative rates of the two reactions will assist in determining conditions where SO_2 and H_2S would be formed in the proportions needed for the Claus reaction in a fixed-bed reactor.

The groups of tests proposed for the high pressure and atmospheric pressure electrobalance tests are the same. An additional month is allocated for reactor modifications because of the added complexity of the apparatus, and two months is allocated to complete the tests in each group, also because of the added complexity involved in high pressure operation. The primary objective of the high pressure electrobalance tests is to determine if pressure alters the conclusions reached in the atmospheric pressure test series.

Five months is the estimated time required to modify and commission the fixed-bed reactor system, and to develop the analytical procedures for product gas analysis. 2½ months are then allocated to complete each of the three phases of the cerium sorbent study to be followed by a 3-month study of FeS regeneration using the partial oxidation concept. These are, of course, the key tests in Task 3.2, and the results will play an important role in defining the bench-scale tests to be conducted in Task 4.2.

Structural property measurements will be initiated approximately two months after the task start time and will be carried out on an as-needed basis throughout the duration of Task 3.2.

References

Copeland, R.J., 1993, "High Temperature H_2S Removal," Proceedings of the Symposium on Coal-Fired Power Systems '93 - Advances in IGCC and PFBC," Morgantown, WV, DOE/METC 93/6131.

Lopez, A., et al., 1994, "Advanced Sulfur Control Concepts in Hot-Gas Desulfurization Technology: 1. Feasibility of the Direct Production of Elemental Sulfur During the Regeneration of High Temperature Desulfurization Sorbents," Topical Report, DOE Contract DE-AC21-94MC30012.