**INTEGRATED EMISSIONS CONTROL SYSTEM
FOR RESIDENTIAL CWS FURNACE**

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TABLE OF CONTENTS

1. INTRODUCTION	1
2. PROGRAM OBJECTIVE	2
3. SYSTEM DEFINITION	3
3.1 COAL COMBUSTION REVIEW	3
3.1.1 Coal Chemistry	3
3.1.2 Devolatilization	4
3.1.3 Char Oxidation	7
3.1.4 Volatile Matter Oxidation	11
3.1.5 Soot Oxidation	11
3.2 SLURRY HANDLING REVIEW	11
3.2.1 Pumping	11
3.2.2 Piping	13
3.3 NO _x REDUCTION REVIEW	13
3.4 SULFUR DIOXIDE REMOVAL REVIEW	15
3.5 SYSTEM DESIGN APPROACH	19
4. PROTOTYPE COMPONENT ANALYSIS AND DESIGN	21
4.1 SO ₂ REACTOR ANALYTICAL MODELING	21
4.1.1 Effect of Sorbent Particle Diameter on Reactor Performance	28
4.1.2 Effect of Ca/S Mole Ratio on Reactor Performance	31
4.1.3 Effect of Reactor Height on Reactor Performance	31
4.1.4 Effect of Input Rate on Reactor Performance	34
4.1.5 Effect of SO ₂ Concentration on Reactor Performance	34
4.2 PROTOTYPE COMPONENT DESIGN	38
4.2.1 NO _x Reduction Test Facility	38
4.2.2 SO ₂ Reduction Test Facility	41
5. COMPONENT TESTING AND DEVELOPMENT	46
5.1 NO _x REDUCTION TESTING	46
5.1.1 Unstaged Operation	46
5.1.2 Staged Operation	50
5.1.3 SO ₂ Reactor Testing	61
5.2 INTEGRATED SYSTEM TESTING	88
5.2.1 Integrated Emissions Control System Design	88
5.2.2 Coal Water Slurry (CWS) Fuel Properties	92
5.2.3 NO _x Emissions	99
5.2.4 SO ₂ Emissions	107

TABLE OF CONTENTS (Continued)

6. MANUFACTURING AND OPERATING COST EVALUATION	127
6.1 CWS FURNACE SYSTEM HIGH-VOLUME PRODUCTION COST ESTIMATE	127
6.2 ECONOMIC EVALUATION: TOTAL LIFE-CYCLE COST	128
6.3 ECONOMIC EVALUATION: PAYBACK PERIOD	139
7. CONCLUSIONS AND RECOMMENDATIONS	141
 REFERENCES	142

LIST OF FIGURES

3.1	Peristaltic Pump with Dual Pump Heads	14
3.2	Nitrogen Pathway	16
4.1	Normalized Static Pressure Distributions Inside Axial Recirculation Emissions Control Reactor	22
4.2	External and Internal Circulation in the Emissions Control Reactor	23
4.3	Emissions Control Reactor Model (High Load) Intensive External and Internal Circulation	24
4.4	Effect of Particle Diameter on Reactor Performance	30
4.5	Effect of Ca/S Mole Ratio on Reactor Performance	32
4.6	Effect of Reactor Height on Performance	33
4.7	Effect of Input Rate on Reactor Performance	35
4.8	Effect of SO ₂ Concentration on Reactor Performance	36
4.9	Effect of Inlet SO ₂ Concentration on Reactor Outlet Concentration	37
4.10	NO _x Reduction Test Facility	39
4.11	Cutaway Drawing of CWS-Fired Residential Combustor	40
4.12	Atomizer Operating with Oil	42
4.13	Photograph of NO _x Test Facility	43
4.14	SO ₂ Removal Test Facility	44
4.15	SO ₂ Test Facility	45
5.1	NO _x as a Function of Stoichiometric Ratio for Unstaged Operation	48
5.2	Weight Percent of Total NO _x in the Form of NO ₂ as a Function of Stoichiometric Ratio	49
5.3	Thermochemical Equilibrium NO _x Concentration as a Function of Temperature and Stoichiometric Ratio for the Combustion of Coal Water Slurry	51
5.4	Combustor Configurations Used During Staged Operation	54
5.5	Effect of Excess Air on NO _x Emissions for Typical Test Runs (Configurations 2 and 2A)	55

LIST OF FIGURES (Continued)

5.6	Effect of Stoichiometric Ratio on NO_x for Various Combustor Configurations Burning CWS Number 2	57
5.7	Effect of Primary/Stoichiometric Ratio on NO_x for Various Combustor Configurations	58
5.8	Effect of Primary Chamber Residence Time on NO_x Emissions for CWS Numbers 1 and 2	60
5.9	Effect of Staged Combustion Operation on Combustion Efficiency for CWS Number 2	62
5.10	Comparison of Particle Size Distributions for Sorbents 1 and 1-A .	63
5.11	6-Inch SO_2 Reactor Performance for Various Operating Conditions	64
5.12	SO_2 Concentration as a Function of Time for a Typical Test	67
5.13	SO_2 Reduction for Various Operating Conditions (6-Inch Reactor) .	68
5.14	6-Inch SO_2 Reactor Performance With and Without Axial Recirculation - Sorbent 1-A	69
5.15	6-Inch SO_2 Reactor Performance With and Without Axial Recirculation - Sorbent 1-A, Weight Basis	71
5.16	Comparison of SO_2 Reactor Performance at a $\text{Ca/S} = 3:1$	72
5.17	Particle Size Distribution of Injected Sorbent and Spent Sorbent ..	73
5.18	Plastic Flow Visualization Model	74
5.19	Effect of Axial Recirculation on Particle Size Distribution	76
5.20	Effect of Axial Recirculation on Particle Size Distribution with 60° Taper Cone	77
5.21	Particle Size Distribution for Various Sorbents Tested in the 8-Inch Diameter Reactor	80
5.22	Effect of Ca/S Mole Ratio on 8-Inch Diameter Reactor Performance	81
5.23	Effect of Sorbent Loading (Weight Basis) on the Performance of the 8-Inch Reactor	83
5.24	Effect of Ca/S Mole Ratio on 8-Inch Diameter Reactor Performance	84
5.25	Performance Comparison Between 6- and 8-Inch Diameter Reactor Operating with Sorbent 1-A	85

LIST OF FIGURES (Continued)

5.26	Comparison of Model Prediction and Reactor Data for the 8-Inch Diameter Reactor Operating with Sorbent 3	87
5.27	Collected Material Breakdown at Varying Sorbent Utilizations in Order to Meet the Emissions Goal	89
5.28	Schematic Diagram of Integrated Emission Control System	90
5.29	Photograph of Integrated Emissions Control System	91
5.30	Assembly Drawing of Integrated Reactor/Heat Exchanger	93
5.31	Viscosity vs. Shear Rate for IL6-248-MCW-G	95
5.32	Particle Size Distributions for Illinois No. 6 and Kentucky Hazard Coal Water Slurries	97
5.33	Rheological Properties of Tecogen Coal Water Slurry Made from Kentucky Hazard Coal	98
5.34	CO ₂ , O ₂ , and CO as a Function of Time During Operation of the Integrated System	100
5.35	NO _x , SO ₂ , and CO as a Function of Time During Operation of the Integrated System	101
5.36	Combustor Wall and Reactor Inlet Temperature for Operation of the Integrated System	102
5.37	NO _x vs. Stoichiometric Ratio for Operation of the Integrated System for Kentucky Hazard CWS	104
5.38	NO _x vs. Stoichiometric Ratio for Operation of the Integrated System for Kentucky Hazard CWS	105
5.39	Average NO Emissions as a Function of Excess Air for Kentucky Hazard and Illinois No. 6 CWS Combustion	106
5.40	NO _x Emissions During SO ₂ Reactor Operation at Different Input Rates	108
5.41	Effect of Input Rate on NO _x Emissions for Kentucky Hazard CWS	109
5.42	Integrated Emissions Control Reactor Performance (Kentucky Hazard CWS, Sorbent 2)	111
5.43	Integrated Emissions Control System SO ₂ Reactor Performance with Sorbent 2	113

LIST OF FIGURES (Continued)

5.44	SO ₂ Reactor Performance for Different Sorbents (Kentucky Hazard CWS)	114
5.45	SO ₂ Reactor Performance for Different Sorbents (Kentucky Hazard CWS)	116
5.46a	Partition Configurations	118
5.46b	Partition Configurations (Continued)	119
5.47	Photograph of Reactor with Partition Plate	120
5.48	Partition Configurations 1, 2, 3, and 4	121
5.49	Effect of Partitions on Reactor Pressure Drop	122
5.50	Cross Section of the SO ₂ Reactor Vacuum Bottle	123
5.51	Cross Section of SO ₂ Reactor with Vacuum Bottle	124
5.52	Photograph of SO ₂ Reactor with Vacuum Cylinder Installed	126
6.1	Economic Comparison of CWS Furnace System with Standard Oil System	133
6.2	Economic Comparison of CWS Furnace System with Standard Gas System	134
6.3	Total Life Cycle Cost Break-Even Comparison of CWS Furnace System with a Standard Oil System	136
6.4	Effect of Heating Load Size on CWS Furnace System Economics for Oil Costing \$7/MMBtu	138
6.5	Effect of Life Cycle Duration on CWS Furnace System Economics .	140

LIST OF TABLES

3.1	Carbon-Oxygen Reaction Rate Constants, k_p	10
3.2	Chemical Reactions Involving Fuel NO_x Formation and Destruction	17
4.1	Modeling Parameters	29
5.1	Fuel Properties	47
5.2	Properties of Coal Water Fuels Tested	52
5.3	Test Conditions	65
5.4	Sorbents Used in Testing the 8-Inch Diameter SO_2 Reactor	79
5.5	Comparison of Illinois No. 6 and Kentucky Hazard Coals	96
5.6	Sorbents Used in Testing the Integrated Emissions Control System	110
6.1	Warm Air Heating Systems Used in Economic Analysis	129
6.2	Assumptions Used in Economic Analysis	131
6.3	Degree Days for Various U.S. Cities	137

1. INTRODUCTION

One of the major obstacles to the successful development and commercialization of a coal-fired residential furnace is the need for a reliable, cost-effective emission control system. Ash, sulfur, and fuel-bound nitrogen make the control of NO_x , SO_x , and particulates much more difficult with coal-based fuels than with either natural gas or oil. While techniques for controlling these emissions in large utility power plants are well advanced, they generally would be too complex and costly to be applied directly to residential applications.

To meet the emission goals set by the Pittsburgh Energy Technology Center (PETC), Tecogen is developing a novel, integrated emission control system to control NO_x , SO_2 , and particulate emissions. At the heart of this system is a unique emissions control reactor for the control of SO_2 . This reactor provides high sorbent particle residence time within the reactor while doing so in a very compact geometry. Final cleanup of any fine particulates exiting the reactor, including respirable-sized particulates, is completed with the use of high efficiency bag filters.

With SO_2 and particulate emissions being dealt with by an emissions control reactor and bag filters, the control of NO_x emissions needed to be addressed. Under a previous contract with PETC (Contract No. DE-AC22-87PC79650), Tecogen developed a residential-scale Coal Water Slurry (CWS) combustor. This combustor makes use of centrifugal forces, set up by a predominantly tangential flow field, to separate and confine larger unburned coal particles in the furnace upper chamber. Various partitions are used to retard the axial, downward flow of these particles, and thus maximize their residence time in the hottest section of the combustor. By operating this combustor under staged conditions, the local stoichiometry in the primary zone can be controlled in such a manner as to minimize NO_x emissions.

The development program for this project was divided into three facets, each having specific objectives with clear goals that were met prior to proceeding to the next step. First, all system and component analyses was completed, and the initial prototype component hardware was tested and evaluated. Preliminary manufacturing and operating cost estimates were also made. Second, final prototype hardware was built and tested, and cost estimates updated. Finally, the entire furnace system was tested to demonstrate that the emissions goals were met, and the high-volume production cost estimates were finalized.

2. PROGRAM OBJECTIVE

The objective of this program is to develop a combined particulate, SO₂, and NO_x integrated control system for flue gas cleanup of residential furnaces fueled by coal water fuel mixtures. Specific objectives for this program are:

- To reduce flue gas emissions from the furnace to the following levels:

SO₂: 0.4 lb/MMBtu

NO_x: 0.2 lb/MMBtu

Particulates: 0.02 lb/MMBtu
 (99.9% capture of particles 0.5 to 10 microns)
- To achieve a high reliability and a service life of 20 years or more by utilizing a simple design concept with no critical moving parts.
- To keep scheduled maintenance to twice a year.
- To achieve efficient sorbent utilization in reducing SO₂ so that the volume of ash and spent sorbent collected before removal is acceptable.
- To use a simple design approach to keep manufacturing costs as low as possible.

To achieve these objectives, this program was divided up into 8 individual tasks. These tasks, along with their various subtasks, provide the framework for this project and establish the various goals and milestones. The 8 main tasks for this program are:

- Task 1: Project Work Plan
- Task 2: System Definition
- Task 3: Prototype Component Analysis and Design
- Task 4: Prototype Component Fabrication and Assembly
- Task 5: Testing and Development
- Task 6: Integrated System Operation
- Task 7: Economic Evaluation
- Task 8: Reporting and Management

An additional task was added to the program, Task 9, to conduct additional parametric testing of integrated emissions control system. This task concentrated on testing enhancements to the SO₂ emissions control reactor.

3. SYSTEM DEFINITION

To help define the emissions control system, a literature review was conducted to determine the level of understanding of the factors affecting emissions from small-scale coal-fired systems. This information provided the basis for the development of an analytical model used in the design of the SO₂ emissions control reactor.

3.1 COAL COMBUSTION REVIEW

A review of the existing literature on the various aspects of coal and coal-water mixture combustion has been conducted and is provided below. The main topics reviewed are coal make-up, devolatilization, char oxidation, volatile oxidation, soot oxidation, and coal-water mixture oxidation.

Coal is a complex, solid, heterogeneous material of carbon base, capable of supplying energy for heat and processing. The chemical and physical make-up of coal has been studied extensively and continues to be studied. Coal studies are usually divided into two areas - coal chemistry (chemical and physical make-up) and coal utilization (combustion, gasification, and liquefaction).

3.1.1 Coal Chemistry

Coal is classified by rank and grade.¹ Coal rank is a method of expressing the progressive metamorphism of coal from lignite (low) to metaanthracite (high). There are four ranks of coal: (1) lignite, (2) subbituminous, (3) bituminous, and (4) anthracite. Each rank has also been subdivided, based on ash-free heating values for low rank coals and moisture-ash-free fixed carbon content for high rank coals. The results of the subdivisions - lowest to highest rank - are: lignite B, lignite A, subbituminous C, subbituminous B, subbituminous A, high volatile bituminous C, high volatile bituminous B, high volatile bituminous A, medium volatile bituminous, low volatile bituminous, semianthracite, anthracite, and metaanthracite. The heating value increases from 13,900 kJ/kg for lignite B to 34,900 kJ/kg for low volatile bituminous A and then decreases to 30,100 kJ/kg for metaanthracite. The fixed carbon content increases from 25% for lignite B to 90% for metaanthracite.

Coal is also classified by grade. The three factors that cause coal to have a low grade are: high ash content, low ash fusion temperature, and high sulfur content. Sulfur is the primary constituent that lowers the grade and thus has been studied extensively. Sulfur in coal lowers the quality of iron and steel, causes corrosion and deposits, and promotes air pollution. Sulfur ranges between

0.2 to 7.0% by weight, with the average between 1 and 2%. Sulfur occurs as inorganic pyrite and marcasite (40 to 80%) with the balance contained in the organic structure. The highest sulfur coals are the bituminous coals of the Pennsylvanian age in the Appalachian, Illinois, and Western Interior coal basins. The lowest sulfur coals are the subbituminous coals and lignites in the Rocky Mountain and Northern Great Plains Regions. These coals contain less than 1% sulfur. The molecular structure of coal is not completely known. Investigators (Solomon, Wisner, and Heredy and Wender)² have gathered information from infrared measurements, nuclear magnetic resonances, ultimate and proximate analyses, and pyrolysis data to obtain models for the chemical structure of the organic backbone of coal. This organic backbone of coal is constructed of aromatic and hydro-aromatic units. Some of these units have various functional groups that cross-link the units together. The planar nature of the aromatics gives the coal a layered consistency. The layers may be twisted in such a way that a perfect laminate is not obtained. The imperfection in the laminate creates pores that may house ash or other impurities. Low rank coals have about 90% of the carbon associated with 5-unit layers, while high rank coals have about 90% of the carbon associated in 30-unit layers.^{1,2,3,4}

3.1.2 Devolatilization

Coal devolatilization has been studied widely and many models have been developed.¹ The models reviewed are limited to coal particles less than or equal to 100 μm . These models are categorized as: (1) single reaction, (2) multiple parallel reactions, (3) multiple competing reactions, (4) complex schemes, and (5) schemes involving secondary char formation. These models vary from the very simple to the very complex. The details of these models are discussed below.

For the single reaction, coal $\rightarrow$ solid + volatile, the reaction rate models are simple and quite limited. Both an Arrhenius expression:

$$\frac{dV}{dt} = k (V_{\infty} - V)^n \quad (3.1)$$

and a non-Arrhenius expression

$$\frac{dV}{dt} = BV_{\infty}/t \quad (3.2)$$

have been developed. The order n in the Arrhenius expression has been estimated to be between 1 and 8, depending on the investigator. The non-Arrhenius expression neglects the observed temperature dependence on devolatilization.

The second category consists of multiple parallel reactions that are first-order Arrhenius expressions. This category has been subdivided by Ubhayakar¹ into two sections, namely, two first-order Arrhenius models and multiple first-order reactions with a statistical distribution of the activation energy. The devolatilization process is modeled by the first of these techniques by the following set of equations:

$$\begin{aligned} C_1 &\xrightarrow{k_1} S_1 + V_1 \\ C_2 &\xrightarrow{k_2} S_2 + V_2 \end{aligned} \quad (3.3)$$

where:

$$\frac{d C_{1,2}}{dt} = - k_{1,2} C_{1,2} \quad (3.4)$$

The second technique is described by the equations below:

$$\text{Coal} = \sum_{i=1}^n C_i \xrightarrow{k_i} \sum_{i=1}^n (S_i + V_i) \rightarrow S + V \quad (3.5)$$

where:

$$\frac{dV_i}{dt} = k_i (V_{i\infty} - V_i) \quad (3.6)$$

$$k_i = A \exp (-E_i/RT) \quad (3.7)$$

$$\int_0^{\infty} f(E) dE = 1 \quad (3.8)$$

Equation (3.8) gives a complete statistical distribution of activation energy.

Multiple competing reaction models, the third category, are also used to express the devolatilization process. In this category, the devolatilization reaction is expressed:

with two rate equations

$$\frac{d(\text{coal})}{dt} = - \left(\sum_{i=1}^n k_i \right) \text{Coal} \quad (3.10)$$

and

$$\frac{dV}{dt} = \left(\sum_{i=1}^n Y_i V_i \right) \text{Coal} \quad (3.11)$$

Kobayashi^{1,2} used this model with $n = 2$. Smith and Smoot² applied this model in their pulverized coal combustion and gasification models with good success.

The fourth category, complex schemes, includes the multiple consecutive parallel first-order reaction:

and the parallel competing first order reaction:

These two mechanisms are complicated, producing several intermediate and final products. A coal combustion model needs a simple, straightforward model of the devolatilization process. These two models do not provide these criteria.

The final category, schemes involving secondary char-forming reactions, contains two models: (1) consecutive competing char-forming reactions and (2) parallel competing char-forming reactions. These models are too complex to be included in a combustor model. The first has seven reactions, producing seven product types, while the second has $n + 2$ reactions and a statical distribution of activation energies.

The models presented above indicate that the fraction of volatile matter evolved is a function of both the temperature and the length of time for which devolatilization occurs. For bituminous coals, Kobayashi et al.² report that at a temperature of 1260 K and for a time of 200 milliseconds, 30% of the coal is volatilized.

3.1.3 Char Oxidation

The devolatilization process produces a solid residue, char. The char consists of the fixed carbon and ash. In reality, the char is a composite material containing carbon, hydrogen, sulfur, oxygen, nitrogen, and ash. An ultimate analysis of the char shows that the fractions of each of these elements in general are in approximately the same ratios as in the parent coal. Char oxidation is a heterogeneous chemical reaction. These reactions have been studied extensively,⁵ and the reaction mechanism developed in those studies is discussed below.

Heterogeneous Char Reactions

Char consists of three major elements: carbon, hydrogen, and sulfur, which are oxidized. The general heterogeneous chemical reaction is expressed as:

Written in this manner, the reactions for carbon, hydrogen, and sulfur become:

This group of reactions is a simplification of possible reactions that have been researched and shown to exist in minor amounts.¹

Smith has reviewed the question as to which product is favored – CO or CO₂ – and suggests that CO is the most likely.²

Kinetics of Heterogeneous Char Oxidation

Heterogeneous chemical reactions have also been studied extensively. Levenspiel⁵ has proposed a mechanism for gas-solid reactions that has withstood numerous investigations and applications. Heterogeneous reactions are envisioned to proceed by the following steps:

- (1) Diffusion of reactant gas from the bulk fluid through the gas film surrounding the particle to the solid surface.
- (2) Penetration and diffusion of reactant gas through a blanket of ash (solid product) to the unreacted core.
- (3) Chemical reaction of reactant gas with solid at the reaction surface.
- (4) Diffusion of the product gas through the gas to the exterior surface of the particle.
- (5) Diffusion of the product gas through the gas film surrounding the particle to the bulk fluid.

One or all of these steps may be involved in the chemical reaction sequence. The steps that offer the greatest resistance to the propagation of the reaction are called the rate controlling steps. For reactions such as char oxidation in which a stationary ash layer is not formed, Steps 2 and 4 have negligible resistances and can be neglected. The driving force for the product gas between the surface and the bulk fluid is in general quite large since the bulk contains little or no product gas. Thus, Step 5 offers negligible resistance and is neglected.

The rate of chemical reaction for the general reaction with Steps 1 and 3 controlling is given by:

$$R = \frac{dm_B}{dt} = \frac{A b M_B C_A}{\frac{1}{k_c} + \frac{1}{k_d}} \quad (3.19)$$

The two terms, $1/k_c$ and $1/k_d$, are the chemical reaction resistance and the mass transfer resistance, respectively. The chemical reaction rate constant, k_c , has been studied extensively for the carbon-oxygen reaction.^{1,2} However, reaction rate constants for the hydrogen-oxygen and sulfur-oxygen reactions are not available in published literature. The mass transfer coefficient, k_d , has been studied extensively for a single particle in a fluid.⁵

The reaction rate coefficient, k_c , for the carbon-oxygen reaction has been well studied by investigators² using the conventional Arrhenius expression $k = A \exp(-E/RT)$ to model the effect of reaction temperature. Table 3.1 summarizes the values for the frequency factor A, and activation energy E, along with the type of coal char and particle size used in the experiment. As can be seen in Table 3.1, the type of coal char greatly influences the values of the frequency factor and activation energy.

The mass transfer coefficient is expressed:

$$k_d = \frac{Sh D}{d_p} \quad (3.20)$$

The Sherwood Number, Sh, is modeled by:

$$Sh = 2 + 0.6 Re^{1/2} Sc^{1/3} \quad (3.21)$$

The Sherwood Number or the mass transfer coefficient modeled by these equations shows good agreement with the experimental data and is an accepted approach to modeling the mass transfer resistance.

TABLE 3.1
CARBON-OXYGEN REACTION RATE CONSTANTS, k_p

$$\text{where } k_p = \frac{k_c M_{O_2}}{T_g R} = A \exp (-E/RT)$$

Type of Coal Char	Particle Size	Frequency Factor	Activation Energy
(μm)	$\left(\frac{g}{cm^2 s atm O_2} \right)$		$\left(\frac{k_{cal}}{kgmol} \right)$
Bituminous	28	92	21,900
38			
82			
105			
Non-Swelling	27	45.7	17,263
Bituminous	38	80.3	21,787
Swelling	31	94.3	23,463
Bituminous	72	1000	35,194
Anthracites	23	20.4	19,000
and			
Semi-Anthracites	78		
Flash Pyrolysis Char	85	16.3	17,597
Petroleum Coke	--	15	19,000

3.1.4 Volatile Matter Oxidation

Experimental information on the volatile oxidation rate has not been found in the literature. Experimental research in this area is needed to verify the theoretical predictions.^{1,2} Volatile product combustion times are estimated by considering only mass transfer resistances and chemical reaction rate resistances.³ The burning time, assuming the volatiles have a molecular weight of 100 and the coal has a volatile fraction of 0.5, is 7 milliseconds for a 50- μm coal particle at 1273 K. If the mass transfer resistance is negligible, such that only the chemical reaction is considered and assuming the reaction rate to be that for CO going to CO₂ (the slowest step), the combustion time can be estimated. These assumptions provide only an order of magnitude value. The burning time for a 50- μm particle at 1273 K is 3.2 milliseconds. These two burning time values are approximately the same, such that the volatile oxidation process is neither only diffusion controlled nor reaction rate controlled.

3.1.5 Soot Oxidation

The oxidation time for soot has been estimated.³ For a soot particle with a 500Å diameter the combustion time is 0.173s at a temperature of 1600 K.³ Lee, Thring, and Beer³ obtained the following expression for the soot combustion rate:

$$r = 1.085 \times 10^4 P_{O_2} T^{1/2} \exp(-39300/RT) [=] \frac{\text{g}}{\text{cm}^2 \text{ s}} \quad (3.22)$$

for 400Å soot particles.

3.2 SLURRY HANDLING REVIEW

The in-house handling of CWF will typically consist of pumping CWF to the atomizer, piping, and cleaning systems.

3.2.1 Pumping

The pumping of the CWF is of critical concern since the CWF fuels are abrasive and exhibit complex rheological behavior. Depending on the shear rates, the CWF can have pseudoplastic, dilatant, or Newtonian shear stress/shear rate behavior. The low solids CWF (50 to 60%) generally exhibit Newtonian behavior for shear rates magnitudes comparable to handling systems and do not require the same concern for fuel handling.^{6,7} In general, pumping system design has to avoid high shear rates and must match the pumping velocities to the type of fuel used.

The rheology of CWF depends strongly on coal type, extent and method of beneficiation, solids content, particle size distribution, and additives used in the formulation, as well as time. Initially, the CWFs show a reduction in viscosity,⁷ the magnitude of which is based on the additive levels and particle size distribution.

After the initial reduction, especially for heavily loaded slurries, the viscosity remains constant at a particular shear rate until the accumulated amount of shear increases to the design point. Once this level of shear is reached, the viscosity of the fuel gradually increases and a yield point forms. This viscosity continues to increase with accumulated shear until the fluid is no longer pumpable. If a sample of this CWF were tested, it would exhibit a completely different shear stress/shear rate behavior from its original rheogram. There are many causes for this behavior: comminution, deagglomeration, additive evaporation, or changes in the zeta potential.⁷

Three types of pumps have been used for CWF: progressing cavity pump, diaphragm pump, and centrifugal pump. Most of the CWF industry has centered on the use of progressing cavity pumps since they develop low shear rate, have minimal or no wear problems, and have a wide selection of flow rate and discharge pressures. Materials compatible with CWFs include modular iron, carbon steel, stainless steel, Buna N, and various polymer compounds.

In general, progressing cavity pumps are highly suitable for handling CWF.⁸ They have predictable performance and pulsationless discharge. Erosion is kept to a minimum by the use of elastomers for the outer stationary gear. Also, the compression fit between the elastomeric stator and rotor makes the pump capable of pumping high viscosity and abrasive CWFs. Progressing cavity pumps have been used extensively and successfully in government research facilities, industrial trial combustion tests, and in commercial slurry producers' plants, both in the United States and abroad. Most of the pumping of the highly loaded CWF, however, has not been continuous so far.

Centrifugal pumps have high internal shear rates and, thus, accelerate the alteration of CWF properties.⁸ CWF behavior is affected more dramatically in cases of highly loaded CWF, as compared to pumping moderately loaded CWF. Thus, the CWF formulation (with respect to the combination of particle size distribution, interaction of chemical dispersant, and stabilizers) greatly affects the performance of the centrifugal pumps. In view of these limitations, centrifugal pumps are not suitable for residential furnace applications.

Another type of pump that has been used for pumping CWF in residential furnace applications is the peristaltic pump. The peristaltic pump offers several advantages over the progressing cavity and centrifugal pumps because of the way it operates. First, none of the mechanical parts of the pump come in contact with the CWF. This eliminates any wear problems associated with the pump as a result of the abrasive nature of CWF. Second, because of the positive displacement nature of the pump, it can be calibrated and used as a metering pump to measure the flow rate of the CWF being burned. These pumps are much less expensive than progressing cavity pumps, and the only maintenance they require is the replacement of pump head tubing after about 1,000 to 1,500 hours of operation. Finally, the peristaltic pumps are available in very small capacities as required for residential applications; therefore, there is no need for recirculation of the excess CWF back to the storage tank.⁷

Should the CWF characteristics require recirculation for keeping the coal in suspension, a peristaltic pump with multiple heads can be installed on the same drive shaft, as shown in Figure 3.1. In this arrangement, the pumps can be sized for slurry recirculation and the amount of CWF needed for atomization.

3.2.2 Piping

By designing the piping for laminar flow, the shear rate in the piping is kept low. At higher velocities, the differential pressures are too great and may also lead to erosion problems. The design of valve pressure drops should also be kept low to minimize erosion. For pipe sizing, Newtonian assumptions are used with standard design equations.

To minimize settling problems, all of the piping is pitched, piping dead-legs are avoided, and rod-out ports are used at branch points. The piping system is provided with a water flushing or cleaning system for removal of residual fuel in the lines. Although the inherent stability of the CWF will allow the slurry to be pumped to the atomizer without the need for flushing during routine operation, during extended outages the water flushing system is operated to avoid buildup of CWF in the piping system.

3.3 NO_x REDUCTION REVIEW

Slurry-fueled systems have a number of advantages over dry pulverized coal systems in controlling NO_x emissions (NO and NO₂). Lower operating temperatures significantly reduce the amount of NO_x produced by oxidation of the nitrogen in the combustion air (often referred to as "thermal NO_x").

3-688

Figure 3.1 Peristaltic Pump with Dual Pump Heads

However, in coal-fired systems as much as three-quarters of the NO_x results from the oxidation of fuel-bound nitrogen (or "fuel- NO_x "). Fuel- NO_x formation appears to depend on the local availability of oxygen during combustion, the complex evolution of intermediate nitrogenous radical species, and coal properties such as volatility. Meeting future stringent NO_x emissions limits without relying on post-combustion controls will require effective control on chemistry of fuel-bound nitrogen.

Figure 3.2 simplistically demonstrates the pathway in which fuel nitrogen converts to NO and NO_2 . Pyrolysis produces volatile nitrogen species, primarily in the amine and cyanide groups. These then undergo oxidation and thermal degradation, producing NO and NH_i ($i = 1, 2, 3$) species. The char nitrogen is oxidized to NO. The NO is reduced to N_2 by reaction with CO, CaSO_3 , C, or NH_3 . These reactions are given in Table 3.2.^{9,10,11}

The first four reactions shown in Table 3.2 are the nitrogen oxide generating reactions. Oxidation of the fuel nitrogen is an essential step in the mechanism transforming fuel-bound nitrogen to N_2 molecule. Careful control of the stoichiometry in this initial region where pyrolysis occurs and NO production begins will provide an abundance of chemical species able to reduce the NO to N_2 by consuming oxygen.

Staged combustion provides a chemical environment favorable to NO reduction by supplying a relatively high concentration of unoxidized carbon in the char, carbon monoxide, ammonia, and calcium sulfite (where calcium oxide is used to control SO_2 emissions). It should be pointed out for clarity, however, that NO is produced throughout the combustor from oxidation of the nitrogen in the char.

Researchers have shown that the reduction of NO by oxidizing the other reactant in reactions 5 through 8 progresses at rates approximately 2/3 as fast as oxidation with O_2 .^{9,10} Reaction 9 shows the conversion of NO to NO_2 , producing a mixture of NO and NO_2 referred to as NO_x . This mechanism has been used to predict NO_x formation from an industrial steam generator¹² with good results.

3.4 SULFUR DIOXIDE REMOVAL REVIEW

The introduction of the Clean Air Act has nurtured the development of sulfur dioxide removal processes. Both wet and dry techniques have been developed to date, with wet processes dominating the market. These processes are expensive to manufacture and operate. Dry processes such as limestone injection systems and fluidized bed coal combustion in a limestone bed are both being researched.

Figure 3.2 Nitrogen Pathway

TABLE 3.2
CHEMICAL REACTIONS INVOLVING FUEL NO_x
FORMATION AND DESTRUCTION

Reaction Number	Reaction
1	Coal + heat $\rightarrow$ volatile N + char N
2	Volatile N + O ₂ $\rightarrow$ NO + NH ₃
3	Char N + O ₂ $\rightarrow$ NO
4	NH ₃ + 5/4 O ₂ $\rightarrow$ NO + 3/2 H ₂ O
5	NO + C $\rightarrow$ CO + 1/2 N ₂
6	NO + CO $\rightarrow$ CO ₂ + 1/2 N ₂
7	NO + 2NH ₃ + O ₂ $\rightarrow$ 3H ₂ O + 3/2 N ₂
8	NO + CaSO ₃ $\rightarrow$ CaSO ₄ + 1/2 N ₂
9	NO + 1/2 O ₂ $\rightarrow$ NO ₂

Dry sulfur dioxide removal processes utilize the following chemical overall reaction.

The reaction proceeds forward at temperatures up to 1500 K for sulfur dioxide concentrations of approximately 3000 ppm.¹³ The calcium oxide is generated by the calcination of limestone (CaCO_3) and dolomite ($\text{CaCO}_3/\text{MgCO}_3$). The calcium oxide and magnesium oxide obtained by calcination are both capable of sulfinating. Sulfination of magnesium sulfate occurs at temperatures below 1110 K. However, experimental evidence shows the reaction to occur only slightly in the 700 to 970 K temperature range.¹³

The sulfination reaction rate has been found to decay with time. This phenomenon is explained by the observation that pores are created in the stone structure by the calcination process. The actual decay in rate is attributed to the substitution of the much larger sulfur trioxide molecule for the carbon dioxide molecule. Researchers have taken two approaches in modeling this phenomenon. Some have used simple exponential decay models^{13,14,15} while others have used complicated grain models.^{16,17} The grain models are more theoretical in nature and probably describe the actual decay phenomenon as it truly is. However, the grain models' complexities limit their use in a combustion process model.

Borgwardt¹³ has conducted extensive experimental studies on stone sulfination. He modeled the reaction rate time delay phenomenon by modifying the conventional Arrhenius frequency factor. Borgwardt presented the following model:

$$R = \frac{\eta}{\rho_s} A_0 e^{-\beta_1 \tau} e^{-E/RT} C_{\text{SO}_2}^n \quad (3.24)$$

He presents a method to obtain β , the exponential decay coefficient.

Lee^{14,15} has developed a simple model, based on two experimental parameters, which predicts the time decay quite well. This model is discussed below. The rate of reaction is expressed by the product of the initial rate and an exponential decay term.

$$R = R_0 e^{-\beta \tau} \quad (3.25)$$

The exponential decay term contains the pore plugging time constant, τ_p , which is taken as one third the pore plugging time.

$$\tau_p = \frac{t_p}{3} \quad (3.26)$$

The pore plugging time has been shown to be inversely proportional to the sulfur dioxide concentration, thus τ_p can be expressed by:

$$\tau_p = \frac{P^*}{C_{SO_2}} \quad (3.27)$$

where P^* is the pure plugging constant. The initial rate, R_0 is expressed by:

$$R_0 = \frac{\pi d_p^3}{6} \rho_s k S \eta C_{SO_2} \quad (3.28)$$

The group $\rho_s k S \eta$ is combined into one constant, τ_{SF} , which has been provided for a number of limestone and dolomite stones. Upon substituting, Lee obtained the following equation:

$$R = \frac{\pi d_p^3}{6} \frac{C_{SO_2}}{\tau_{SF}} e^{(-C_{SO_2}/P^*)} \quad (3.29)$$

The only parameters needed from experimental data are P^* and τ_{SF} . Lee provided these data¹⁴ and conducted all experimental work using 1000 μm particles.

The performance predicted by these models, particularly Borgwardt's model, agrees nicely with experimental data.^{18,19} Only a 3.5% deviation was obtained between predicted and experimental results for an industrial scale steam generation unit. This shows that the Borgwardt reaction model is usable in an engineering model of a desulfurization reactor.

3.5 SYSTEM DESIGN APPROACH

Residential scale systems that utilize coal are subject to different constraints than their industrial and utility counterparts. They must have a low first cost, be easy to operate, and require minimal maintenance. These constraints dictated the approach to be used to meet the emissions goals for NO_x , SO_2 , and particulates.

For NO_x emissions control, a staged combustion approach was adopted. This approach was chosen due to its simplicity and low cost.

For SO_2 emissions control, a high temperature, totally dry sorbent injection approach was chosen for use in the reactor downstream of the combustor. This approach satisfied all of the system criteria and also yielded a dry, non-leaching byproduct, CaSO_4 . Also, by injecting the sorbent at furnace temperature, inexpensive limestone, CaCO_3 , can be used compared to more expensive lime; CaO .

A problem with high temperature, dry sorbent injection is that sorbent utilization is lower than for wet scrubbing approaches, such as for wet scrubbers and spray dryers. On the residential scale, the problem of low sorbent utilization is further complicated by the large surface area to volume ratios found in smaller scale equipment. These larger surface area to volume ratios are conducive to significant heat transfer within the SO_2 reactor, even though the system is well insulated. As the temperature decreases through the reactor, the SO_2 capture process becomes severely kinetics limited. Though sorbent utilization is lower for this approach, it best meets the criteria for residential scale systems.

For particulate emissions control, a high efficiency bag filter system was chosen. This approach was chosen for its simplicity and low floor space requirement.

4. PROTOTYPE COMPONENT ANALYSIS AND DESIGN

In designing and analyzing the system components for individual testing, the SO_2 reactor required considerable attention. The SO_2 reduction reactor was designed on the principle of axial recirculation. In a vortical flow field, the pressure at the periphery is greater than that at the centerline. This is shown in Figure 4.1 where normalized curves of pressure difference as a function of radial position are presented. By connecting one end of a tube tangentially from the periphery at one axial location, and the other end at the centerline, an external axial recirculation flow path is established that operates only on the natural pressure difference present in the vortical flow field without the need of external assistance.

When the principle of axial recirculation is used in a gas/solids system, the residence time of the solids within the reactor can be significantly increased relative to that of the gas. A schematic diagram of both the internal and external particle recirculation paths is shown in Figure 4.2, while a photograph of the external particle recirculation is shown in Figure 4.3 for a plastic flow visualization model. Due to the very complex flow field within the reactor, typical analytical techniques become difficult to apply with any certainty.

4.1 SO_2 REACTOR ANALYTICAL MODELING

In developing an analytical model of the SO_2 reactor, several complexities are encountered due to the coupling of the flow field in the main body of the reactor to that of the axial recirculation loop. These two highly coupled flow fields result in the need to solve a three-dimensional, highly nonisotropic turbulent flow field with a dispersed second phase of particles that undergo chemical reactions.

One possible approach to modeling the reactor is to use available computational fluid dynamics (CFD) computer codes. Several codes are commercially available; however, each has limitations when applied to modeling the SO_2 reactor. The turbulent flow field within the reactor is nonisotropic and as such, the standard two-equation $k - \epsilon$ model used in most codes is not the best representation of the turbulence within the reactor. To more accurately model the flow within the reactor, an algebraic stress model (ASM) should be used. A survey of codes that are commercially available shows that codes that have ASM turbulence models are not capable of accurately modeling the reactor geometry due to the recirculation line. The codes that are available that can handle the reactor geometry are typically limited to using the two-equation $k - \epsilon$ model, which is not the best choice for this problem.

Figure 4.1 Normalized Static Pressure Distributions Inside Axial Recirculation Emissions Control Reactor

Figure 4.2 External and Internal Circulation in the Emissions Control Reactor

**Figure 4.3 Emissions Control Reactor Model (High Load)
Intensive External and Internal Circulation**

In addition to the problems of geometry and turbulence models, the use of CFD codes to model the process inside the reactor can require a considerable computational effort. For example, it was estimated by a software vendor that to resolve the flow within a residential-sized SO₂ reduction reactor, approximately 72,000 nodes would be required. To obtain a solution, the code must solve as a function of position within the reactor the three velocity components, pressure, a minimum of two turbulence parameters, particle trajectories, and chemical reactions with the gas and the particles. This can result in the need to solve more than 500,000 simultaneous, nonlinear equations, which is typically beyond the capabilities of engineering work stations for reasonable turnaround times.

As an alternative to the use of CFD codes, a simplified model of the reactor was developed. The purpose of this model was to determine the dominant parameters that govern the reduction of SO₂ within the reactor, as well as to provide performance estimates under various operating conditions.

In developing the analytical model, one of the prime areas of concern is for given gas-flow conditions, the ability of the reactor to detain particles of a given size. Though the flow within the reactor is quite complex, if the recirculation line effect is neglected, then the reactor is quite similar to a cyclone separator. The ability of cyclones to collect particles of a given size has been well documented, for example, by Zenz.²⁰ For a given geometry and gas flow conditions, the theoretically smallest-size particle size that a cyclone can collect, typically called the cut point diameter, can be found by:

$$Dp_{\text{cut}} = \left[\frac{9 \mu_g L_w}{\pi N_s V_{\text{in}} (\rho_p - \rho_g)} \right]^{\frac{1}{2}} \quad (4.1)$$

where:

μ_g is the gas viscosity

V_{in} is the inlet velocity

ρ_p, ρ_g are the particle and gas density, respectively

N_s is the number of spiral turns, which is found from experiment

L_w is the cyclone inlet width

For application of this equation to the axial recirculation reactor, L_w is defined as the radial distance between the reactor wall and the outside radius of the recirculation tube.

Once the cut point diameter is determined, the separation efficiency of any other size particle can be determined from normalized separation efficiency data, as shown by Zenz.²⁰ With a knowledge of the separation efficiency of a given-size particle, its residence time in the reactor relative to the gas can be determined. Under steady-state conditions and assuming that the reactor is capable of fluidizing all the particles within it, as one particle is added to the reactor, one particle must leave. However, the particle that is added to the system will be recirculated N times before it leaves. The number of times a particle of a specific diameter within the reactor circulates, N , is dependent on the separation efficiency for this size particle. This relationship can be expressed as:

$$N = 1 + \eta + \eta^2 + \eta^3 + \dots \quad (4.2)$$

where η is the separation efficiency. This relation can be expressed in closed form as:

$$N = \frac{1}{1 - \eta} \quad (4.3)$$

Once N is determined, the amount of solid material in the reactor that can participate in the reduction of SO_2 can be found since it is equal to N times the inlet solids flow rate.

With a knowledge of the amount of solids present in the reactor, one-dimensional species-continuity equations for the concentration of SO_2 in the gas phase and CaO in the solid phase can be written as:

$$\frac{d [\text{SO}_2]}{dZ} = \frac{-6 (1 - \epsilon) [\text{SO}_2]}{\epsilon D_p V_s \left(\frac{1}{k_c} + \frac{1}{k_d} \right)} \quad (4.4)$$

$$\frac{d [\text{CaO}]}{dZ} = \frac{-6 [\text{SO}_2]}{D_p V_s \left(\frac{1}{k_c} + \frac{1}{k_d} \right)} \quad (4.5)$$

where:

$[SO_2]$ is the concentration of SO_2 in the gas phase

$[CaO]$ is the concentration of CaO in the solid phase

V_g, V_s are the gas and solid phase velocities, respectively

ϵ is the void fraction (adjusted to account for particle recirculation)

k_c, k_d are the chemical kinetics and diffusion resistances, respectively

D_p is the particle diameter

The diffusion resistance, k_d , can be found from the relationship:

$$Sh = 2 + 0.6 Re^{\frac{1}{2}} Sc^{\frac{1}{3}} \quad (4.6)$$

where:

Sh is the Sherwood number defined by:

$$Sh = k_d \frac{D_p}{D} \quad (4.7)$$

D is the diffusivity

Re is the Reynolds number based on the slip velocity

Sc is the Schmidt number

The chemical kinetics resistance is determined by using Borgwardt's model, which takes into account the decrease in the rate of reaction due to pore plugging:

$$K_c = A_o \exp \left(- \beta n' \frac{w}{p} \right) \exp \left(- \frac{E}{RT} \right) \quad (4.8)$$

where:

A_o is the frequency factor

n' is the sulfate in the stone

W is the weight of the solid sample

R is the universal gas constant

T is the absolute temperature

E is the activation energy

β is a stone-dependent constant determined by experiment

This set of coupled equations is solved numerically as a function of length to determine reactor performance.

The computer model was used as part of a parametric study to determine the effect of such parameters as reactor diameter, Ca/S, and particle diameter on SO₂ capture. For this analysis, the reactor model was operated with the combustion products of the CWS (IL6-248-MCW-G) specified for this program and under conditions that the SO₂ reactor would operate under in the integrated system. The parent coal in this CWS is an Illinois No. 6 coal, which contains 2.63% sulfur and has a higher heating value of 13,579 Btu/lbm. This provides the potential of 3.87 lbm/MMBtu of SO₂. When this CWS is burned with 20% excess air, this SO₂ potential translates into a concentration of 1,950 ppm on a dry basis.

In Borgwardt's model, the reaction rate time delay phenomenon caused by pore plugging is treated by modifying the conventional Arrhenius frequency factor with an exponential decay coefficient. This decay factor is characterized by a stone dependent, pore plugging constant, β . Due to this additional feature, three empirically determined constants must be supplied to the model to characterize the kinetics of the SO₂ capture process. The kinetics constants used in this analysis were obtained from Borgwardt's paper. The parameters used in this analysis are shown in Table 4.1.

4.1.1 Effect of Sorbent Particle Diameter on Reactor Performance

The effect of sorbent particle diameter on reactor performance is shown in Figure 4.4 for reactor diameters of 4, 6, and 8 in., operating with a Ca/S mole ratio of 3:1. The interesting feature of Figure 4.4 is that for each reactor diameter, there is an optimum particle diameter for SO₂ capture. This optimum diameter is based on the reactor's ability to capture particles of a given size. Particles smaller than the optimum diameter cannot be detained in the reactor and pass through without participating in the SO₂ capture process. Particles that are larger than the optimum diameter are able to be detained in the reactor; however, since the sorbent particle surface area to volume ratio decreases as the diameter increases, there is less available surface area in the reactor to participate in the SO₂ capture process compared to the optimum.

Another interesting feature of Figure 4.4 is that the ability of the reactor to capture SO₂ increases as the reactor diameter is increased. This is due to the lower axial gas velocity associated with the larger diameter reactor. As the axial velocity decreases, the gas phase residence time increases inversely, providing more time for the SO₂ capture process.

TABLE 4.1
MODELING PARAMETERS

Fuel:	IL6-248-MCW-G (Specified fuel for the program)
Coal Content (weight %)	53.22
Higher Heating Value (Btu/lbm)	7,227
Sulfur Content (coal)	2.63
Input Rate (Btu/hr):	120,000 (nominal)
Excess Air (%)	20
SO ₂ (ppm dry basis)	1,950
Reactor Operating Temperature (F)	1,600
Reactor Diameter (in.)	4, 6, 8
Reactor Height (in.)	45 (nominal)
Sorbent Properties:	
Activation Energy (cal/g-mole)	10,000
Frequency Factor (1/s)	335,000
Beta (g/g-mole)	333.3
Density (lbm/ft ³)	150

Figure 4.4 Effect of Particle Diameter on Reactor Performance
(Operating with a Ca/S Ratio of 3:1)

4.1.2 Effect of Ca/S Mole Ratio on Reactor Performance

The effect of Ca/S mole ratio on reactor performance is shown in Figure 4.5 for reactor diameters of 4, 6, and 8 in. For each of these cases, the model used the optimum particle diameter for these operating conditions, as shown in Figure 4.4. For the sorbent properties used in this analysis, the required 90% reduction in SO₂ emissions is achieved at a Ca/S mole ratio of about 4.5 for the 8-in. diameter reactor.

The effect of gas phase residence time, i.e., reactor diameter on performance, is also shown in Figure 4.5. Reactor performance strongly decreases as the reactor diameter is decreased. For example, at a Ca/S mole ratio of 5:1, the 8-in. reactor achieves a 93% reduction in SO₂, but the 6-in. diameter and 4-in. diameter reactors achieve reductions of 77 and 50%, respectively.

4.1.3 Effect of Reactor Height on Reactor Performance

For given operating conditions, reactor height is directly proportional to gas phase residence time, and as such, as the reactor height is increased, reactor performance also increases. This is shown in Figure 4.6 for reactor diameters of 4, 6, and 8 in., each operating with the optimum particle diameter for these conditions (as shown in Figure 4.4), and with the reactor operating at a Ca/S mole ratio of 3:1. Figure 4.6 clearly shows the negative exponential dependence of reactor performance on height, with reactor performance increasing with reactor height and diameter.

Due to the nature of the species continuity equations, which are first order, reactor performance can be described by an exponentially decaying solution, provided the coupling between the equations for CaO and SO₂ is relatively weak (i.e., low value of beta). For a system that exhibits exponentially decaying behavior, it is desirable to characterize the response of the system in terms of a time constant. Since reactor height is directly related to residence time, the characteristic time constant for the reactor operating under specific conditions can be treated as a characteristic length. This is shown in Figure 4.6, in which the 8-in. diameter reactor, under these operating conditions, has a time constant equivalent to a reactor length of 23 in.

The idea of equating a reactor length scale with an equivalent time constant provides a convenient way of expressing reactor length with a desired level of performance. By normalizing reactor height with respect to the length equivalent time constant, the required number of time constants to achieve a given reduction in SO₂ can be determined by definition since one time constant represents a

Figure 4.5 Effect of Ca/S Mole Ratio on Reactor Performance

Figure 4.6 Effect of Reactor Height on Performance
(Operating at a Ca/S Ratio of 3:1)

63.2% reduction, 2 time constants represent an 86.5% reduction, 3 time constants represent a 95% reduction, 4 time constants represent a 98.2% reduction, and so forth.

4.1.4 Effect of Input Rate on Reactor Performance

The effect of input rate on reactor performance is shown in Figure 4.7 as a function of particle diameter. For this case, only an 8-in. diameter reactor is considered, since at the higher flow rates, the pressure drop becomes excessive in the smaller units. Figure 4.7 shows that as the input rate is increased, reactor performance decreases. This is due to the reduction in gas phase residence time associated with the fixed reactor height.

An interesting feature of Figure 4.7 is that as the input rate is increased, the optimum particle diameter for SO_2 capture decreases. This is due to the fact that at the higher tangential velocities associated with the increased flow rates, the reactor has a greater ability of capturing smaller sized particles. This provides a greater surface area concentration within the reactor.

Figure 4.7 indicates that if operation at higher input rates is required and comparable performance with lower input rate operation is required, provisions must be made to compensate for the reduction in gas phase residence time. This can be accomplished by increasing either the reactor length or diameter.

4.1.5 Effect of SO_2 Concentration on Reactor Performance

The specified coal for this program, when burned with 20% excess air, has the potential to produce 1,950 ppm of SO_2 on a dry basis. Operation of the reactor with the products of combustion of other slurries yields the potential for different SO_2 inlet concentrations. The performance of the reactor as a function of inlet SO_2 concentration is shown in Figure 4.8 for a Ca/S mole ratio of 3:1 and reactor diameters of 4, 6, and 8 in. Figure 4.8 shows that the percent reduction in SO_2 increases as an exponential function of inlet concentration, approaching a limiting value of close to 100% reduction.

The fact that the percent reduction in SO_2 approaches an exponential limit at near 100% as inlet concentration is increased indicates that in absolute terms, i.e., with respect to the actual outlet concentration (ppm), the reactor would have a lower outlet SO_2 concentration when operated with higher sulfur coals. To illustrate this idea, the percent reduction results shown in Figure 4.8 are plotted in Figure 4.9 in terms of the actual outlet concentration of SO_2 (ppm on a dry basis). Figure 4.9 shows that particularly for the 6-in. diameter and 8-in. diameter

Figure 4.7 Effect of Input Rate on Reactor Performance
(For an 8-in. Diameter Reactor Operating
with a Ca/S Ratio of 3:1)

Figure 4.8 Effect of SO_2 Concentration on Reactor Performance
(Operating at a Ca/S Ratio of 3:1)

Figure 4.9 Effect of Inlet SO₂ Concentration on Reactor Outlet Concentration (Operating with a Ca/S Ratio of 3:1)

reactors, as the inlet concentration is increased, for a fixed Ca/S ratio, there is more sorbent within the reactor and the outlet SO₂ concentration decreases. For example, consider the 8-in. diameter reactor. Using this particular sorbent, an outlet concentration of about 400 ppm would be expected with the reactor operating at the 1,950 ppm expected for this coal. This result is about 200 ppm higher than the level required to meet the 0.4 lbm/MMBtu program goal. However, if the reactor was operated with a coal that produced about 3,500 ppm of SO₂, the outlet concentration would meet the goal of 0.4 lbm/MMBtu (200 ppm).

4.2 PROTOTYPE COMPONENT DESIGN

To simplify NO_x and SO₂ emissions reduction work, two separate initial prototype component test facilities were designed. The purpose of having two separate test facilities was to have as high a degree of flexibility as possible for testing the individual components. For example, while one component in a particular test facility is being modified, testing can continue in the other test facility with no loss of continuity.

4.2.1 NO_x Reduction Test Facility

The first test facility was designed for NO_x reduction work. A schematic diagram of this test facility is shown in Figure 4.10. This test facility consists of the CWS combustor, transition box, heat exchanger, baghouse, and various support equipment and instrumentation. At the heart of the NO_x reduction test facility is the CWS combustor, where NO_x emissions are controlled by operating the unit under staged conditions.

The CWS combustor used in this work is a derivative of the one that was developed by Tecogen under contract No. DE-AC22-87PC79650. A cutaway drawing of the combustor is shown in Figure 4.11. The combustor makes use of centrifugal forces, set up by a predominantly tangential flow field, to separate and confine larger unburned particles in the upper chamber. Various partitions are used to retard the downward, axial flow of these particles and thus maximize the residence time of these particles in the hottest section of the furnace.

As the particles burn and their size decreases, radial drag forces overcome the centrifugal forces, and the particles are able to exit the primary chamber. Combustion of these smaller particles is completed in a secondary chamber. Control of NO_x emissions in this combustor is accomplished by staging the combustion air between upper and lower sections of the primary combustion chamber, such that the unit operates in a substoichiometric manner in the upper section of the primary combustion chamber. The combustor was designed such

Figure 4.10 NO_x Reduction Test Facility

Figure 4.11 Cutaway Drawing of CWS-Fired Residential Combustor

that different inserts could be used to change the height of the upper primary chamber, and thereby change the residence time of the products of combustion in this fuel-rich zone.

The atomizer used in this work is similar in design to the supersonic CWS combustor previously developed at Tecogen. This atomizer was annular in design, with CWS flowing in the central tube while atomizing air flowed in the annular passageway around the CWS tube. The airstream shears the CWS to an unstable ligament sheet which breaks up into droplets external to the atomizer. To aid in the atomization process, the airstream was accelerated to supersonic velocities by means of a converging/diverging section. A photograph of the atomizer operating with oil is shown in Figure 4.12. To accurately determine the primary/stoichiometric ratio in the upper primary chamber of the combustor, the atomizing airflow rate and the combustion inlet airflow rate were measured accurately.

A photograph of the NO_x Test Facility is shown in Figure 4.13.

4.2.2 SO_2 Reduction Test Facility

A schematic diagram of the SO_2 reduction test facility is shown in Figure 4.14. To have more control over the operation of the SO_2 reactor, it was decided to operate the facility with flue gases produced from an oil burner and inject SO_2 as required to obtain desired inlet concentrations. Also, since oil burners can operate over a very wide range of input rates, typically from 68,500 to over 400,000 Btu/hr for residential scale burners, they provide a great deal of flexibility in reactor operation. A photograph of the SO_2 test facility is shown in Figure 4.15.

For the initial prototype reactor components, it was decided to fabricate the reactor from standard stainless-steel pipe to facilitate modifications. Two separate reactors were designed, one 6 inches in diameter, the other 8 inches in diameter. Both reactors were designed to be modular so that various length spool pieces could either be added or removed to vary the gas residence time.

To construct both the NO_x and SO_2 test facilities in a cost-effective manner, it was decided to allow both facilities to share certain components. These included the baghouse and the cooling water/dump radiator loop.

TF54-290

Figure 4.12 Atomizer Operating with Oil

Figure 4.13 Photograph of NO_x Test Facility

Figure 4.14 SO₂ Removal Test Facility

Figure 4.15 SO₂ Test Facility

5. COMPONENT TESTING AND DEVELOPMENT

During this program, testing was conducted on the system components individually and as an integrated system.

5.1 NO_x REDUCTION TESTING

The CWS combustor was tested under both unstaged and staged conditions. Under both operating conditions, the combustor geometry, specifically the upper primary chamber, was modified to determine the effect of residence time on NO_x emissions. The CWS used during this testing contained 65.16% coal by weight, with the parent coal having a mean particle diameter of 14.0 microns. All the properties of the CWS are summarized in Table 5.1.

5.1.1 Unstaged Operation

As part of unstaged testing, NO_x emissions were recorded under various input rates ranging from 113,400 to 181,700 Btu/hr, and for two different primary combustion chamber heights of 3 and 7.69 inches. For these tests, the combustor tangential air inlet height equalled the entire length of the primary chamber. These results are shown in Figure 5.1 as a function of stoichiometric ratio. Preliminary results for unstaged operation have shown NO_x emissions to be fairly insensitive to input rate and primary chamber residence time. Peak NO_x emissions are on the order of 0.78 pound of NO_x per million Btu fired and occurred at just under 40% excess air. Operation of the combustor at stoichiometric conditions produced NO_x emissions on the order of 0.285 pound per million Btu fired.

NO_x emissions were recorded on a dry basis in the form of the species that was actually measured. Using this approach, the reported NO_x emission are the sum of NO and NO₂ emissions on a mass basis.

Though primary chamber residence time has little effect on the total amount of NO_x produced during unstaged operation, it was found to have an effect on the amount of NO_x produced in the form of NO₂. This is shown in Figure 5.2 where the percent of NO_x in the form of NO₂ on a weight basis is plotted as a function of stoichiometric ratio. At lower stoichiometric ratios, the amount of NO₂ produced is low for both chamber heights that were tested; however, as the stoichiometric ratio is increased, the longer chamber (which provides a longer gas residence time) has a higher percentage of total NO_x in the form of NO₂.

TABLE 5.1
FUEL PROPERTIES

Identification	Upper Elkhorn Number 3 Seam
Heating Value	15,251 Btu/lbm
Ash	1.32 percent (by weight)
Sulfur	0.66 percent (by weight)
Ash Fusion Temperatures Initial Softening Fluid T-250	2150°F 2300°F 2400°F 2509°F
Ultimate Analysis Carbon Hydrogen Nitrogen	85.40 percent 5.40 percent 1.59 percent
Solids Content	65.16%
Mean Particle Size	14 μm
Viscosity at 100 s₋₁	419 cps

Figure 5.1 NO_x as a Function of Stoichiometric Ratio for Unstaged Operation

Figure 5.2 Weight Percent of Total NO_x in the Form of NO₂ as a Function of Stoichiometric Ratio

The fact that NO_2 is present in the products of combustion of CWS is quite unexpected when thermochemical equilibrium is considered. As an indication of the amount of NO_x predicted from thermochemical equilibrium for the combustion of CWS, Figure 5.3 shows the effect of both temperature and stoichiometric ratio on NO_x production, primarily in the form of NO. Equilibrium analysis predicts that NO_2 is present only in insignificant quantities, i.e., mole fractions of less than 10^{-5} . The fact that NO_2 is present in the products of combustion when equilibrium does not favor its formation is an interesting phenomenon.

One possible explanation for this occurrence is the presence of HO_2 radicals in the flame region. It has been suggested that in the flame region, NO molecules are scavenged by HO_2 radicals to form NO_2 and OH. Due to the water present in the CWS, it is expected that there is a higher probability of HO_2 radicals being present, which would contribute to the formation of NO_2 .

Another reason for the presence of NO_2 in the products of combustion can be attributed to the presence of sodium. Sodium has been shown to aid in the formation of NO_2 and since it is quite possible to have sodium present in the water used in the preparation of the CWS, it may help account for the presence of NO_2 .

5.1.2 Staged Operation

Under staged operating conditions, the combustor was tested using two different fuels. The first CWS tested was made from Upper Elkhorn Number 3 coal that was deeply cleaned and contained 1.32% ash and 0.66% sulfur. This CWS was used extensively in the development of the combustor under a previous program (Contract Number DE-AC22-87PC79650) and during unstaged operation. This fuel contained 65.16% coal by weight and was called UE3-224-MCW-F by the CWS supplier (Energy International Inc.) and for identification purposes in this program was called CWS number 1. The second CWS tested was made from Illinois Number 6 coal and contained 6.72% ash and 2.63% sulfur. This CWS contained 53.22% coal and was identified by the manufacturer (AMAX) as IL6-248-MCW-G, and in this program was called CWS number 2. CWS number 2 was the fuel specified to meet the emissions goals of this program. Properties of CWS number 1 and CWS number 2 are shown in Table 5.2.

In comparing CWS number 1 and CWS number 2 to potential NO_x emissions, two facts are evident from Table 5.2. First, CWS number 2 contains more water than CWS number 1 (46.78% compared to 34.84%). Second, CWS number 2 contains less fuel-bound nitrogen than CWS number 1. On a normalized basis to the heating value of the coal, CWS number 2 has 1.002 pounds of molecular nitrogen contained in the equivalent of 1 million Btu and CWS number 1 has 1.043 lbm/MMBtu of nitrogen in the coal.

Figure 5.3 Thermochemical Equilibrium NO_x Concentration as a Function of Temperature and Stoichiometric Ratio for the Combustion of Coal Water Slurry

TABLE 5.2
PROPERTIES OF COAL WATER FUELS TESTED

	CWS Number 1	CWS Number 2
Coal Identification	Upper Elkhorn Number 3	Illinois Number 6
Heating Value (Btu/lbm)	15,251	13,579
Ash	1.32%	6.72%
Sulfur	0.66%	2.63%
Ultimate Analysis:		
Carbon	85.40%	77.56%
Hydrogen	5.40%	4.69%
Nitrogen	1.59%	1.36%
Solids Content	65.16%	53.22%
Mean Particle Diameter (microns)	14	9
Viscosity (cps @ 100/s)	301	230

5.1.2.1 Combustor Configurations for Staged Combustion Operation

To date, the combustor has operated for prolonged periods under four different staged combustion configurations. A schematic diagram detailing these configurations is shown in Figure 5.4. The first configuration that was tested was configuration 1 and was only used in tests with CWS number 1. In this configuration, the primary air inlet extended the entire length of the primary chamber, that is, 7.69 in. This long air inlet places a limit on the degree to which the primary chamber stoichiometric ratio could be reduced and provide stable combustion.

In configuration 2, the combustor upper primary chamber was extended in length to 12.5 in. in an effort to provide longer residence time. In this configuration, the primary air inlet was only 2.5 in. long. This provided higher air inlet velocities, which allowed for lower primary chamber stoichiometric ratios. This configuration was used with both CWS number 1 and CWS number 2. As a variation on configuration 2, the primary air inlet width was reduced 15% to provide higher tangential velocities. This variation in configuration 2 was called configuration 2A and was used in tests with CWS number 2.

In configuration 4, the combustor upper primary chamber was extended to 18.5 in. in length to provide even longer primary chamber residence time. This configuration had the same air inlet geometry as did configuration 2A; however, the combustion chamber diameter was decreased from 8.5 to 8 in. in the upper section of the primary chamber. This was done to decrease heat loss in this section of the combustor where the CWS is introduced and to therefore aid in evaporation. Since the primary chamber in configuration 4 is lengthened, the surface area available for heat loss is increased, and, coupled with the high water content of CWS number 2, made combustion difficult without the additional insulation. In fact, the original version of this configuration (called configuration 3) did not have the extra 1/2 in. of insulation, and in this configuration it was not possible to initiate or maintain stable CWS combustion with CWS number 2.

5.1.2.2 Effect of Excess Air on NO_x for Staged Combustion of CWS Number 2

Data obtained during testing in the NO_x reduction test facility were recorded every 15 seconds by a computerized data acquisition system. For each configuration, typically three tests were conducted, and these tests lasted for several hours. This resulted in each configuration generating large amounts of data that needed to be reduced. Typical results for two test runs are shown in Figure 5.5 in terms of NO_x as a function of excess air. To reduce the data to a more

Configuration 4

Configuration 2 - 2A (*)

Configuration 1

* Note: The main difference between configurations 2 and 2A is that configuration 2A had a smaller air inlet so as to provide higher tangential velocities.

Figure 5.4 Combustor Configurations Used During Staged Operation

Figure 5.5 Effect of Excess Air on NO_x Emissions for Typical Test Runs (Configurations 2 and 2A)

useful form, a statistical averaging procedure was used. First, the data for each configuration were sorted in ascending order with respect to stoichiometric ratio. The data were then averaged in increments of 0.03 (3% excess air) of stoichiometric ratio.

The effect of excess air on NO_x emissions for staged combustion operation with CWS number 2, tested in configurations 2, 2A, and 4, is shown in Figure 5.6. These results represent the averaged values of over 1,000 data points. The results shown in Figure 5.6 indicate that NO_x is essentially a linearly increasing function of excess air for each of the configurations tested. The data also indicate that of the three configurations tested, configuration 4 has the lowest NO_x through the operating range. Configuration 2A had the next lowest NO_x levels, followed by configuration 2, which exhibited the highest NO_x levels of the configurations tested.

In comparing the results shown in Figure 5.6, the differences between the three configurations must be considered. Configurations 2 and 2A differ only in the air inlet geometry, with configuration 2A capable of operating at higher tangential velocities which prevent agglomeration of coal particles on the combustor wall. Due to this difference, it was possible to operate configuration 2A at lower primary/stoichiometric ratios than configuration 2. Both configurations 2A and 4 have the same air inlet geometry and therefore provide the same tangential velocity when operated at comparable conditions. However, since configuration 4 was 6 in. longer than configuration 2A it provided longer primary chamber residence time, which further lowered NO_x emissions.

The results shown in Figure 5.6 with the combustor operated in configuration 4 show that emissions levels as low as 0.14 lbm/MMBtu were achieved when the combustor was operated with 5% excess air. The emissions goal for this program was 0.2 lbm/MMBtu, and in order to operate at this level, the combustor can be operated with up to 12% excess air in configuration 4.

5.1.2.3 Effect of Primary/Stoichiometric Ratio on NO_x for Staged Combustion of CWS Number 2

The main difference between configurations 2 and 2A is that the primary air inlet in configuration 2A is smaller than in configuration 2. This difference allowed configuration 2A to operate at lower primary/stoichiometric ratios than configuration 2 while still maintaining tangential velocities high enough to provide stable combustion. The effect of primary/stoichiometric ratio on NO_x emissions is shown in Figure 5.7 for combustor configurations 2, 2A, and 4.

Figure 5.6 Effect of Stoichiometric Ratio on NO_x for Various Combustor Configurations
Burning CWS Number 2

Figure 5.7 Effect of Primary/Stoichiometric Ratio on NO_x for Various Combustor Configurations
(Fuel: IL6-248-MCW-G)

The results shown in Figure 5.7 indicate that for each configuration, NO_x emissions are essentially a linear function of primary/stoichiometric ratio. Figure 5.7 also shows the key difference between configurations 2 and 2A. Configuration 2 could only be operated at a minimum primary/stoichiometric ratio of 0.89 while still maintaining stable combustion and obtained NO_x emission levels of 0.36 lbm/MMBtu. In configuration 2A, the higher tangential velocities allowed the combustor to maintain stable combustion at a minimum primary/stoichiometric ratio of 0.69, and at this ratio achieved NO_x emissions of 0.26 lbm/MMBtu.

Figure 5.7 also shows in an indirect manner the effect of primary chamber residence time on NO_x emissions. Both configurations 2A and 4 were operated over the same range of primary/stoichiometric ratio; however, configuration 4 achieved consistently lower NO_x emissions over this range of operation. This is a result of the fact that configuration 4 has a longer primary chamber than configuration 2A and thereby provides longer residence time for the products of combustion in this fuel-rich zone.

5.1.2.4 Effect of Primary Chamber Residence Time on NO_x Emissions

The results shown in Figures 5.6 and 5.7 present NO_x emissions as a function of excess air and primary/stoichiometric ratio, respectively, and have implicitly shown the effect of primary chamber residence time. To observe this effect directly, Figure 5.8 shows NO_x emissions as a function of primary chamber residence time for operation with CWS number 2 in configurations 2, 2A, and 4 as well as test results obtained while burning CWS number 1 in configurations 1 and 2.

The interesting feature of Figure 5.8 is that if the test results from configuration 2 are ignored, all the results for both slurries operating under different configurations fall essentially on the same curve. This indicates that for this combustor, primary chamber residence time is the dominant parameter in NO_x emissions. This is a significant result since both slurries were made from different parent coals that had vastly different solids loading. The deviation of the results from configuration 2 on the effect of primary chamber residence time is assumed to be due to the fact that configuration 2 could only be operated at higher primary/stoichiometric ratios, and as such, the benefits of deeper air staging could not be realized.

During staged operation of the combustor with CWS number 2 in configuration 2, ash samples were collected for analysis to determine the effect of primary/stoichiometric ratio on combustion efficiency. These results are shown in

Figure 5.8 Effect of Primary Chamber Residence Time on NO_x Emissions for CWS Numbers 1 and 2

Figure 5.9, which shows an increase in combustion efficiency with decreasing primary/stoichiometric ratio. At the lowest primary/stoichiometric ratio tested, the highest combustion efficiency of 97.3% was obtained.

5.1.3 SO₂ Reactor Testing

Individual component testings were conducted in the SO₂ removal facility on both 6 inch and 8 inch nominal diameter reactors both with and without axial recirculation. These tests were conducted with the products of combustion of Number 2 fuel oil and at various SO₂ concentrations obtained by gaseous injection.

5.1.3.1 Six-Inch Diameter Reactor Performance

The first reactor tested in the SO₂ Removal Test Facility was a nominal 6-in. diameter reactor. This reactor had the following geometry:

Inside Diameter:	6.065 in.
Recirculation Line Diameter:	1.049 in.
Active Reactor Height:	45 in.
Inlet Area:	4 in. ²

The calcium-based sorbent used in this testing was a dolomite from northwest Connecticut that is used primarily for residential lawn applications. This dolomite contained about 60% CaCO₃ with the balance being MgCO₃. This sorbent was used in two forms that differed in particle size distribution. The first form was designated sorbent 1, and was the as-received form of the dolomite. This sorbent had a mean particle diameter of 194 microns. The second form of the sorbent was designated sorbent 1-A. This sorbent was obtained by sieving and then selectively mixing sorbent 1 to result in a mixture having a mean particle diameter of 117 microns with a good percentage of fines. A comparison of particle size distributions for sorbents 1 and 1-A is shown in Figure 5.10 in the form of a Rosin and Rammler chart.

Numerous tests were conducted on the 6-in. diameter SO₂ reactor to assess the performance of the reactor for various operating temperatures, Ca/S mole ratios, gas residence times, inlet SO₂ concentration, and sorbent particle diameter. The performance of the 6-in. diameter reactor is shown in Figure 5.11 as the form of percent reduction in SO₂ plotted for various Ca/S mole ratios. Figure 5.11 shows that an asymptotic limit is reached in SO₂ reduction at a Ca/S mole ratio of approximately 4:1. Test conditions for the data shown in Figure 5.11 in terms of average gas temperature and gas residence time are shown in Table 5.3.

Figure 5.9 Effect of Staged Combustion Operation on Combustion Efficiency for CWS Number 2

Figure 5.10 Comparison of Particle Size Distributions for Sorbents 1 and 1-A

Figure 5.11 6-Inch SO₂ Reactor Performance for Various Operating Conditions

TABLE 5.3

TEST CONDITIONS

Test No.	Average Gas Temperature (°F)	Gas Residence Time (ms)	Average Inlet SO ₂ Concentration (ppm)	Average Inlet SO ₂ Concentration (lbm/MMBtu)	Sorbent
SO ₂ _3	1,437	219	2,070	5.29	1
SO ₂ _5	1,579	244	2,280	5.27	1
SO ₂ _6	1,407	333	2,350	5.30	1
SO ₂ _8	1,514	314	1,570	3.22	1
SO ₂ _9	1,534	312	1,170	2.39	1
SO ₂ _10	1,548	312	1,900	3.88	1-A
SO ₂ _11	1,439	337	2,110	4.20	1-A
SO ₂ _12	1,544	329	2,260	4.58	1-A

In obtaining the data shown in Figure 5.11, the following procedure was used. First, the reactor was allowed to reach thermal steady state. At that point, a metered amount of SO_2 was injected well upstream of the reactor. Upon reaching steady state, a metered amount of sorbent was injected at the reactor inlet. Sorbent feed was continued until after the reactor had reached steady-state conditions. The percent reduction in SO_2 was then determined by using the average value of SO_2 from before sorbent was injected and well after injection had stopped. During testing, important data were recorded via a microcomputer-based data acquisition system. A typical plot of SO_2 concentration as a function of time is shown in Figure 5.12.

The data shown in Figure 5.11 are reported as percent reduction in SO_2 concentration on a volume basis. The SO_2 test facility operates with the products of combustion produced by an oil burner. Since the composition of these products differs from that of CWS, it becomes necessary to put these results on a coal equivalent basis, so that weight-based results not volume-based results can be reported. To establish this equivalent basis, it was decided to equate the oxygen content in the products of combustion of heating oil with that of CWS. With an equivalent oxygen level for CWS combustion established, the stoichiometric ratio was determined and the volumetric measurements were converted to a weight basis. These results are shown in Figure 5.13.

In testing the 6-in. SO_2 reactor under conditions similar to the combustion of IL6-248-MCW-G, SO_2 reduction levels approaching 80% have been achieved at Ca/S ratios of 4:1, as shown in Figure 5.13. Though this reduction level does not reach the required 89.7% level, it comes fairly close and is quite good compared to the relatively short gas residence time associated with the 6-in. reactor. The interesting feature about the 80% reduction is that it was obtained with the external axial recirculation line blocked and the reactor operated with the smaller particle diameter sorbent 1-A.

5.1.3.2 Effect of Axial Recirculation on Reactor Performance

Operating the reactor with the external axial recirculation line blocked, yielded high SO_2 reduction rates. To determine the effect of external axial recirculation on reactor performance, a test was conducted in which the reactor was operated under conditions similar to the combustion of IL6-248-MCW-G with the recirculation line both open and blocked. These tests were conducted with the smaller diameter sorbent, sorbent 1-A, and the results are shown in Figure 5.14.

Figure 5.12 SO₂ Concentration as a Function of Time for a Typical Test

Figure 5.13 SO₂ Reduction for Various Operating Conditions (6-Inch Reactor)

Figure 5.14 6-Inch SO₂ Reactor Performance With and Without Axial Recirculation Sorbent 1-A

The data shown in Figure 5.14 indicate that when the reactor is operated without axial recirculation, a higher percent reduction in SO_2 is achieved at lower Ca/S mole ratios than when the reactor is operated with recirculation. As the Ca/S mole ratio is increased, the reduction levels tend to approach one another both with axial recirculation and with the recirculation line blocked. This is also shown in Figure 5.15, in which outlet SO_2 concentration based on lbm/MMBtu is plotted against Ca/S mole ratio.

Another comparison of the effect of particle size and the effect of axial recirculation is shown in Figure 5.16, in which outlet SO_2 concentration is plotted against inlet SO_2 concentration on a lbm/MMBtu basis for various sorbent diameters and operating conditions. The results shown in Figure 5.16 were obtained by interpolating the collected data so that all results could be presented for a common Ca/S mole ratio of 3:1. Figure 5.16 shows the sharp difference with and without axial recirculation where for a moderate Ca/S ratio of 3:1, there is quite a difference in performance. Figure 5.16 also clearly shows the difference in reactor performance when the unit is operated under similar thermal conditions and residence time but with a different sorbent particle diameter. Surprisingly, the larger diameter sorbent 1 exhibited better performance than the smaller diameter sorbent 1-A, which provides a greater surface-to-volume ratio.

Smaller sorbent particle diameters provide the potential for greater SO_2 reduction levels only if they can be detained within the reactor. Since external axial recirculation transports particles from the reactor periphery and introduces them as a jet at the reactor centerline, it is expected that some of the smaller entrained particles might be carried out by the action of this jet and as such decrease their residence time within the reactor. To determine the effect of axial recirculation on particle separation, a test was conducted on the 6-in. diameter reactor to determine the actual particle size distribution within the reactor and compare it to what was injected. For this test, once the reactor reached steady-state conditions in terms of SO_2 reduction while using sorbent 1, the oil burner, the SO_2 and sorbent feed, and the I.D. fan were simultaneously shut off, and the particles within the reactor were removed and analyzed for their size distributions. These results are shown in Figure 5.17 along with the distribution for the injected sorbent. The spent sorbent left in the reactor was found to have a mean diameter of 260 microns compared to the 194-micron mean diameter sorbent particles that were injected.

To quantify the effect of axial recirculation on particle separation, tests were conducted on a plastic flow visualization model, shown in Figure 5.18. To avoid the difficulty of measuring the solids flow rate that exited the model under steady-state conditions, a transient approach was adopted. In this approach, a predetermined amount of particles was injected into the model in a very short period of time.

Figure 5.15 6-Inch SO_2 Reactor Performance With and Without Axial Recirculation
Sorbent 1-A, Weight Basis

Figure 5.16 Comparison of SO₂ Reactor Performance at a Ca/S = 3:1

Figure 5.17 Particle Size Distribution of Injected Sorbent and Spent Sorbent

Figure 5.18 Plastic Flow Visualization Model

When the solid feed was stopped, the inlet to the reactor was immediately blocked and the I.D. fan was turned off. The separation efficiency was then determined by comparing the weight of particles collected within the model with that which was injected. In addition to weighing both the injected and collected particles, a sieve analysis was also performed to determine the particle size distribution.

Three different test conditions were examined with the cold flow test facility. These conditions were with axial recirculation, with the recirculation line blocked at the inlet, and with a 2 3/8-in. diameter, 60° tapered cone situated 3 in. above the recirculation tube outlet. For all these tests, the feed material was the same sorbent used in the majority of the hot flow tests, sorbent 1. For these tests, the highest separation efficiency, 93.8%, was obtained with the recirculation line blocked. With the recirculation line open, a separation efficiency of 87.1% was obtained, whereas with the recirculation line open and the deflector cone in place, a separation efficiency of 93.6% was obtained.

The purpose of external axial recirculation is to provide a more uniform distribution of particles within the reactor and thereby increase the effective particle surface area within the reactor and not to aid in particle separation. Without axial recirculation, flow visualization indicates that there tends to be some particle stratification in the upper part of the reactor. With recirculation, these particles are more evenly distributed axially along the reactor; however, particle separation is decreased. Based on hot testing within the SO₂ reactor, it appears that the benefit of a more uniform distribution of particles provided with axial recirculation is offset by the decrease in separation efficiency. It appears that the high degree of mixing due to turbulence within the reactor offsets the effect of particle stratification, and as such there appears to be a benefit in not using axial recirculation in the configuration that was recently tested.

In addition to particle separation efficiency testing on the flow visualization model, particle size distribution comparisons were made between injected and collected particles. Figures 5.19 and 5.20 show the change in the weight of each sample, for each particle size interval, for the case where the recirculation line was blocked and the tapered cone was used. These figures show that a greater amount of fine particles was collected within the reactor than what was injected. This is due to particle fragmentation within the reactor resulting from high tangential velocities.

Figure 5.19 Effect of Axial Recirculation on Particle Size Distribution

Figure 5.20 Effect of Axial Recirculation on Particle Size Distribution with 60° Taper Cone

5.1.3.3 Eight-Inch Diameter Reactor Test Results

Due to the relatively short residence time of the 6-in. diameter reactor, it was difficult to obtain the 0.4 lbm/MMBtu emissions goal of this program when operating with an inlet concentration of 3.87 lbm/MMBtu. To meet the 0.4 lbm/MMBtu goal, an 8-in. diameter reactor was constructed. For this reactor, external axial recirculation was not used, in an effort to maximize separation efficiency. This reactor had the following geometry:

Inside Diameter:	8.329 in.
Outlet Diameter:	1.5 in.
Active Reactor Height:	43 in.
Inlet Area:	4 in. ²
Recirculation Line Diameter:	External Axial Recirculation Not Used

Four different calcium-based sorbents were tested in the 8-in. diameter reactor, and a summary of these sorbents in terms of specific calcium compound, weight percent, and mean particle diameter is shown in Table 5.4. The particle size distribution for these sorbents is shown in Figure 5.21 in the standard Rosin and Rammler format. The sorbents used in this work represented a cross section of various products found throughout the United States. Sorbent 1-A was a dolomite from northwest Connecticut and was tested extensively in the 6-in. reactor. The as-received version of this sorbent, called sorbent 1, had a mean diameter of 194 microns. To obtain the as-tested size distribution, the as-received dolomite was sieved and then selectively blended. Sorbent 2 was a high-calcium limestone from Pennsylvania that was received in pulverized form. Sorbent 3 was a pulverized lime that was calcined from the same limestone used in sorbent 2 and sorbent 4 was an as-received limestone from Illinois.

Results of 8-in. diameter reactor testing are shown in Figure 5.22 in terms of SO₂ content, lbm/MMBtu, as a function of Ca/S mole ratio along with lines that represent a least squares method fit of the data (exponential fit). Figure 5.22 shows that the SO₂ emissions goal for this program – 0.4 lbm/MMBtu – was reached at a Ca/S mole ratio of about 5:1 for sorbent 1-A.

The data shown in Figure 5.22 bring up an interesting feature about the reactor and the SO₂ capture process itself. Consider the results for sorbents 2 and 3, which have essentially the same mean particle diameter and distribution. Sorbent 3 is made from the calcination of the material used in sorbent 2, and since it is CaO that captures SO₂ directly and not CaCO₃, it was expected that better SO₂ capture would be achieved with sorbent 3 since the calcination step within the reactor was eliminated. This was not the case. Sorbent 2, which undergoes the

TABLE 5.4
SORBENTS USED IN TESTING THE 8-INCH-DIAMETER SO₂ REACTOR

Sorbent	Calcium Compound	Percentage	Mean Diameter (microns)
1-A	CaCO₃	60	117
2	CaCO₃	97.8	88.5
3	CaO	95.4	82.1
4	CaCO₃	98.3	172

Figure 5.21 Particle Size Distribution for Various Sorbents Tested in the 8-Inch Diameter Reactor

Figure 5.22 Effect of Ca/S Mole Ratio on 8-Inch Diameter Reactor Performance

calcination process within the reactor, exhibited a much greater ability to capture SO_2 and at much lower Ca/S mole ratios than sorbent 3. Sorbent 2 achieved an SO_2 emissions level of 0.54 lbm/MMBtu, an 88.3% reduction, at a Ca/S mole ratio of 4.12 compared to 0.73 lbm/MMBtu, an 80% reduction, at a Ca/S mole ratio of 7.53.

In addition to reporting SO_2 reduction as a function of Ca/S mole ratio, it is also desirable to use as an independent variable the sorbent mass loading, i.e., the mass of sorbent injected into the reactor for each unit mass of SO_2 . These results are shown in Figure 5.23. The main difference between Figure 5.23 and Figure 5.22 is with respect to sorbent 1-A, which achieved 0.39 lbm/MMBtu at a Ca/S mole ratio of about 5:1. However, since sorbent 1-A is a dolomite that contains approximately 40% MgCO_3 , it takes considerably more sorbent on a mass basis to achieve a given Ca/S mole ratio compared to any of the other high-calcium-based sorbents. Therefore, to achieve the required reduction in SO_2 , over 13 pounds of sorbent 1-A are required for every pound of SO_2 in the gas stream. For purposes of comparison, consider achieving a reduction in SO_2 to 0.6 lbm/MMBtu. For sorbent 1-A, this would require 12 pounds of sorbent for each pound of SO_2 ; however, for sorbent 2, less than 6 pounds of sorbent are required for each pound of SO_2 .

The results shown in Figures 5.22 and 5.23 are SO_2 content on a lbm/MMBtu basis versus Ca/S and sorbent loading ratio, respectively, which represent absolute values. In some instances, it is desirable to present the data on a relative basis, i.e., percent reduction in SO_2 . This is shown in Figure 5.24, in which percent reduction in SO_2 is plotted against Ca/S mole ratio. This plot shows in a very simple manner how each sorbent performs relative to the others. For example, at a Ca/S mole ratio of 1:1, sorbent 2 exhibits an almost 60% reduction in SO_2 , whereas sorbent 4 at this Ca/S mole ratio only exhibits about a 15% reduction.

5.1.3.4 Performance Comparison Between the 6- and 8-Inch Diameter Reactors

The 8-in. diameter reactor represents an increase in gas residence time of 1.78 times that of the 6-inch diameter reactor previously tested. A comparison of the performance of the 8- and 6-inch diameter reactors is shown in Figure 5.25. For these test results, the reactors were operated under the same operating conditions and both used sorbent 1-A. A comparison of both reactors showed that the 8-inch diameter reactor was capable of meeting the emissions goal of 0.4 lbm/MMBtu, whereas the 6-inch diameter reactor was only capable of achieving an SO_2 emissions level of about 1 lbm/MMBtu.

Figure 5.23 Effect of Sorbent Loading (Weight Basis) on the Performance of the 8-Inch Reactor

Figure 5.24 Effect of Ca/S Mole Ratio on 8-Inch Diameter Reactor Performance

Figure 5.25 Performance Comparison Between 6- and 8-Inch Diameter Reactor Operating with Sorbent 1-A

Along with the data shown in Figure 5.25, solid lines, which represent the least square fit of the data using an exponential fit, are also shown. The purpose of using an exponential fit for the data was based on model predictions for the effect of Ca/S mole ratio on SO₂ reduction. Figure 5.25 shows that the exponential fit is quite good for the 8-inch diameter reactor over the entire range of Ca/S mole ratio tested, and is a good fit for the 6-inch diameter reactor only up to a Ca/S mole ratio of 3:1. Beyond a Ca/S mole ratio of 3:1, SO₂ reduction levels off in the 6-inch diameter reactor, and at higher Ca/S mole ratios, SO₂ emissions levels start to rise. A possible explanation for this occurrence is the higher axial gas velocity present in the 6-inch diameter reactor. During testing, the gas flow rate is held constant as sorbent particles are injected. As the sorbent flow rate is increased and a volume of particles accumulates within the reactor, the actual axial gas phase velocity increases and is capable of carrying more sorbent particles out of the reactor. This effect was not noted in the 8-inch diameter reactor, since due to its larger diameter, the axial gas velocity is 1.78 times lower.

5.1.3.5 SO₂ Reactor Model Results and Comparison with Data

With the amount of data available from individual component testing of the reactor, the question arises as to how well the previously developed computer model fits the data. To describe the reaction kinetics used in this model, three constants need to be determined to characterize the sorbent. These constants are the activation energy, E; the frequency factor, A₀; and the pore plugging parameter, beta. Based on test results obtained with sorbent 3, a high-CaO lime product taken at three temperatures, these constants were determined to be:

Activation Energy:	11,750 cal/gmole
Frequency Factor:	243,100 1/s
beta:	0

which are well within agreement of published data.¹³

With an estimation of the sorbent properties, a computer model comparison was performed for two different cases. These results are shown in Figure 5.26, in which percent reduction in SO₂ is plotted against Ca/S mole ratio. Figure 5.26 shows that a fairly good agreement exists between the computer model and data.

Figure 5.26 Comparison of Model Prediction and Reactor Data for the 8-Inch Diameter Reactor Operating with Sorbent 3

5.2 INTEGRATED SYSTEM TESTING

Upon meeting program emissions goals for NO_x and SO_2 , the individual components were integrated into a complete furnace system. This system was extensively tested for NO_x , SO_2 , and particulate emissions while operating with CWS.

During individual component testing, particulate emissions measurements to access baghouse performance were not made. These tests were not conducted due to the difference between particulate loading and particle size distribution between the NO_x and SO_2 test facilities and what the integrated system must operate under. Consider the amount of both sorbent and flyash that the baghouse must deal with. To perform this analysis, consider a house in the Pittsburgh, Pennsylvania, area (5,931 degree days) that has an overall heat transfer coefficient, surface area product (UA) of 700 Btu/hr-°F, typical of a moderate size home of new construction. Also, assume that the heating system for this home has an annual fuel utilization efficiency (AFUE) of 85%, the system burns CWS number 2 or equivalent, and high-calcium-carbonate limestone is used as the SO_2 sorbent. This house would require 1,690 gallons of slurry to meet its heat requirement of 117.2 MMBtu per year and would emit to the atmosphere each year 23.4 pounds of NO_x , 46.9 pounds of SO_2 and 2.34 pounds of particulates, assuming the system met the required emissions goals.

With respect to particulates, the bag filters must annually remove from the combustion products 86.3 pounds of combustible matter (assuming a 99% combustion efficiency), 574.3 pounds of ash, 865.3 pounds of calcium sulfate, and unutilized sorbent ranging from 41 pounds for a Ca/S mole ratio of 1:1 to 2,028 pounds for a Ca/S mole ratio of 6:1. Results of this analysis are shown in Figure 5.27. This analysis indicates that due to the large amount of collected calcium sulfate and unutilized sorbent relative to flyash and unburned carbon, realistic measurements of both the quantity and size distribution that pass through the bag filter could only be made when the system was totally integrated.

5.2.1 Integrated Emissions Control System Design

The integrated emissions control system comprises a full working furnace system including the combustor, integrated SO_2 emissions control reactor/heat exchanger, baghouse, and air handling unit. A schematic diagram of the integrated emissions control system detailing the system layout and instrumentation is shown in Figure 5.28. A photograph of the integrated emissions control system is shown in Figure 5.29.

Figure 3.27 Collected Material Breakdown at Varying Sorbent Utilizations in Order to Meet the Emissions Goal (For a Typical Home in the Pittsburgh Area)

Figure 5.28 Schematic Diagram of Integrated Emission Control System

Figure 5.29 Photograph of Integrated Emissions Control System

In the design of the integrated emissions control system, attempts were made to integrate individual components wherever possible in an effort to save space and minimize system cost. Some of these integrations include using the SO₂ emissions control reactor as a secondary combustion chamber. To control NO_x emissions, the integrated emission control system employs staged combustion techniques that require a primary, fuel-rich zone to reduce NO_x to N₂ and CO₂ by using CO, and a secondary chamber to complete combustion. During the individual component testing and development part of this program, the primary and secondary combustion chamber were combined to form a single unit that was over 3 feet tall. In the integrated approach, the SO₂ reactor now serves as the secondary combustion chamber, which decreases the height of the combustor to under 2 feet.

In addition to using the SO₂ reactor as the secondary combustion chamber, the reactor is now integrated with the heat exchanger to form a compact assembly. An assembly drawing of the integrated reactor/heat exchanger is shown in Figure 5.30. In the integrated reactor/heat exchanger, the products of combustion, along with dry sorbent, enter tangentially at the base of the reactor and spiral upwards. The gas and entrained particles exit the reactor into a plenum and then pass downward through a tubular heat exchanger where heat is transferred from the gas stream to a circulated water loop. The heat exchanger is connected to a discharge plenum where the gas and entrained particles then proceed on to the baghouse.

In designing the SO₂ reactor/heat exchanger, it was decided not to use external axial recirculation. This was done for two reasons. First, by not having the external recirculation pipe, the reactor and the heat exchanger could be easily integrated together without mechanical interference. Second, no noticeable increase in reactor performance was found at this scale. The reason for external axial recirculation is to reduce particle stratification within the reactor. For large scale systems, this poses a significant problem, however for small scale systems, turbulence seems to provide enough internal recirculation to make external axial recirculation unnecessary.

5.2.2 Coal Water Slurry (CWS) Fuel Properties

Two different fuels were used during testing of the integrated emissions control system. The first CWS was made from Illinois No. 6 coal and was supplied by AMAX Research and Development. The coal used in this slurry contained 2.63% sulfur and 6.72% ash, and had a mean particle diameter of 8 microns. This fuel had a coal loading of 53.2% and was the specified fuel for this program.

The Illinois No. 6 fuel was supplied at the beginning of the program and was used for the two years. This fuel has undergone some significant changes in its rheological properties. The most significant change has been associated with an increase in viscosity that occurred with no noticeable change in solids loading. This increase in viscosity varied from barrel to barrel and has accounted for over a 100 cp increase, as shown in Figure 5.31, above specifications. This higher viscosity made it difficult to obtain stable CWS combustion and required the use of additional A 23 dispersant to bring the fuel properties within specifications and allow stable combustion.

Although adding A 23 allowed for stable combustion, achieving consistent rheological properties from test run to test run was difficult because each time the CWS was remixed, a further increase in viscosity was observed, thus changing the initial conditions before the addition of dispersant. Also, when significant amounts of A 23 were added, a film would tend to form on the surface of the slurry. This film would occasionally be drawn into the pump inlet strainer and this would require testing to be stopped. To overcome these problems, a decision was made to burn a CWS produced from Kentucky Hazard coal. This fuel was produced at Tecogen for the DOE-sponsored program "Development and Testing of Commercial-Scale Coal-Fired Combustion Systems Phase III," Program No. DE-AC22-90PC90156. The coal used in this CWS contained 0.76% sulfur and 3.72% ash. The slurry made with the Kentucky Hazard coal had a solids loading of 58.4%. The two fuels used in the integrated emissions control system are compared in Table 5.5.

In comparing the fuels on the basis of emissions, two features are worth noting. First, the Illinois No. 6 slurry has the potential to produce 3.87 lbm/MMBtu of SO_2 , compared to 1.046 lbm/MMBtu for the Kentucky Hazard CWS. To compensate for this difference, gaseous SO_2 was injected into the combustor to bring the reactor inlet concentration to levels comparable to those obtained during operation with Illinois No. 6 coal. The second key difference between the fuels is that Kentucky Hazard coal contains almost 15% more fuel-bound nitrogen, normalized to the coal heating value, than the Illinois No. 6 coal.

Another difference between the two fuels relates to the particle size that the parent coals were ground to. The Illinois No. 6 coal was ground to a mean particle size of 8 microns, typical of a micronized grind, whereas the Kentucky Hazard coal was ground to a mean particle size of 30 microns, which is more representative of a pulverized grind. The particle size distribution for both of these coals is shown in the Rosin-Rammler format in Figure 5.32.

Based on the particle size distribution, the required solids loading, and the additives package, the low shear rheology for the Kentucky Hazard CWS was measured with a Brookfield viscometer. These results are shown in Figure 5.33 and are fairly close to the original specifications for the Illinois No. 6 CWS.

Figure 5.31 Viscosity vs. Shear Rate for IL6-248-MCW-G

TABLE 5.5
COMPARISON OF ILLINOIS NO. 6 AND KENTUCKY HAZARD COALS

Analysis (Dry)	Illinois No. 6	Kentucky Hazard
Carbon	77.55 (83.15 MAF)	81.09 (84.22 MAF)
Hydrogen	4.69 (5.03 MAF)	5.39 (5.60 MAF)
Nitrogen	1.36 (1.46 MAF)	1.67 (1.73 MAF)
Sulfur	2.63 (2.82 MAF)	0.76 (0.79 MAF)
Oxygen	7.04 (7.54 MAF)	7.37 (7.66 MAF)
Ash	6.72 (- MAF)	3.72 (- MAF)
Heating Value (Btu/lbm)	13,579 (7,227 CWS)	14,535 (8,547 CWS)

Notes:**Illinois No. 6****Kentucky Hazard****SO₂ Potential:****3.87 lbm/MMBtu****1.046 lbm/MMBtu****Nitrogen Content:****1.002 lbm/MMBtu****1.149 lbm/MMBtu**

Figure 5.32 Particle Size Distributions for Illinois No. 6 and Kentucky Hazard Coal Water Slurries

Figure 5.33 Rheological Properties of Tecogen Coal Water Slurry Made from Kentucky Hazard Coal

5.2.3 NO_x Emissions

During operation of the integrated emissions control system, NO_x emissions were recorded under different operating conditions. For these tests, NO_x emissions were recorded as lbm/MMBtu of the measured species, which was predominantly nitric oxide (NO).

5.2.3.1 System Operation

Operation of the integrated emissions control system starts with the system being preheated, which is done by burning No. 2 fuel oil. Once the combustor wall temperature reaches approximately 1,900°F, CWS injection begins and the flow of oil is discontinued. This brief overlap of both fuels being fired simultaneously results in high CO concentrations for a short period. During this time, NO_x emissions are recorded by the computerized data acquisition system; however, these results are not reported in the reduced data due to the artificially low NO_x emissions produced in this highly reducing environment. Once CO emissions reach normal operating conditions, recorded NO_x emissions taken on a volumetric basis are converted to a mass basis.

Typical emissions traces for CO₂, O₂, CO, NO_x, and SO₂ as recorded by the data acquisition system are shown in Figures 5.34 and 5.35. These results start from transition to CWS, to full-load operation on CWS, through sorbent injection. For the results shown in Figures 5.34 and 5.35, CO₂, O₂, CO, and NO_x emissions are recorded on a dry basis; i.e., the water vapor in the products of combustion was condensed out in a cold trap before entering the analyzers. The SO₂ emissions shown in Figures 5.34 and 5.35 were recorded on a wet basis to overcome the problem of SO₂ solubility in water. The wet basis measurements involved heating the sample line above the acid dew point and then filtering and diluting the gas sample to lower the dew point below ambient conditions before it entered the SO₂ analyzer. A Thermo Electron Model 900 Sample Condition Unit was used for this filtering and dilution.

In addition to recording emissions, the data acquisition system also recorded key operating temperatures including combustor wall temperature and reactor inlet temperature. To compensate for the effect of thermal radiation, the thermocouple at the SO₂ reactor inlet was aspirated. Typical temperature traces for the combustor upper primary wall temperature and the reactor inlet temperature are shown in Figure 5.36. For the conditions shown in Figure 5.36, the reactor inlet temperature operated at just over 1,500°F, with an average combustor wall temperature of 2,000°F.

Figure 5.34 CO_2 , O_2 , and CO as a Function of Time During Operation of the Integrated System

Figure 5.35 NO_x, SO₂, and CO as a Function of Time During Operation of the Integrated System

Figure 5.36 Combustor Wall and Reactor Inlet Temperature for Operation of the Integrated System

5.2.3.2 Effect of Stoichiometric Ratio and Primary/Stoichiometric Ratio on NO_x Emissions

The effect of stoichiometric ratio on NO_x emissions is shown in Figure 5.37 for the combustion of Kentucky Hazard CWS. The data in Figure 5.37 indicate not only the effect of excess air on NO_x emissions but also the effect of primary/stoichiometric ratio. In Figure 5.37, there are two distinct values of NO_x emissions at a given stoichiometric ratio, one below the 0.2 lbm/MMBtu program goal, and the other above this goal. For the values below the emissions goal, the combustor was operated at a lower primary/stoichiometric ratio than those at the higher NO_x emissions levels.

Plotting the data in Figure 5.37 as a function of primary/stoichiometric ratio eliminates this dual value presentation of the data and the result is shown in Figure 5.38. The interesting feature of Figure 5.38 is that NO_x emissions reach a minimum value at a primary/stoichiometric ratio of just over 0.59.

5.2.3.3 Comparison of Illinois No. 6 and Kentucky Hazard CWS NO_x Emissions

The parent coal in the Kentucky Hazard CWS contains 1.73% fuel-bound nitrogen on a moisture and ash free (MAF) basis compared with 1.46% MAF for the Illinois No. 6 coal. When these values are normalized to the heating value of each fuel, the Kentucky Hazard coal contains almost 15% more nitrogen. Since for CWS combustion the predominant form of NO_x emissions is from fuel-bound nitrogen, an increase in NO_x emissions was expected. This was not the case, however. When average NO_x emissions for both Kentucky and Illinois No. 6 coals are plotted against stoichiometric ratio, no distinct difference can be seen, as shown in Figure 5.39. Though the difference in the coal nitrogen content is not very large, an increase in NO_x emissions was expected. The fact that no increase was observed is worth noting.

5.2.3.4 Effect of Thermal Input Rate on NO_x Emissions

The integrated emissions control system was designed for a nominal thermal input rate of 125,000 Btu/hr; however, at this input rate, the combustor discharge temperature (which is the reactor inlet temperature) is approximately 1,500°F. With regard to SO₂ emissions, this temperature is low for effective calcination of the dry limestone sorbent and subsequent SO₂ capture. For this reason, the thermal input rate needed to be increased to achieve higher sorbent utilization. To achieve acceptable reactor inlet temperatures, a combustor input rate of around 150,000 Btu/hr is required.

Figure 5.37 NO_x vs. Stoichiometric Ratio for
Operation of the Integrated System
For Kentucky Hazard CWS

Figure 5.38 NO_x vs. Stoichiometric Ratio for Operation of the Integrated System for Kentucky Hazard CWS

Kentucky Hazard Illinois No. 6

□

●

Figure 5.39 Average NO Emissions as a Function of Excess Air for Kentucky Hazard and Illinois No. 6 CWS Combustion

Increasing the combustor input rate increases the volumetric heat release rate and increases NO_x emissions. This is shown in Figure 5.40, in which NO_x emissions are plotted as a function of stoichiometric ratio for several different input rates. In addition to the combustor being operated at different input rates, the data shown in Figure 5.40 were obtained during operation of the SO_2 reactor under Ca/S mole ratios ranging from 0 to 4.12. Figure 5.40 shows that as the input rate was increased, NO_x emissions increased; however, the change with increasing stoichiometric ratio appeared constant for the cases where sorbent was injected into the reactor.

The effect of input rate on NO_x emissions is best shown in Figure 5.41, in which these two quantities are plotted against each other at a fixed stoichiometric ratio. Figure 5.41 shows that the increase in NO_x emissions with input rate tends to level off at the higher operating conditions. Operation of the combustor above the 170,000 Btu/hr input rate shown in Figure 5.41 is difficult to achieve with the current configuration due to the size of the air inlets which limits the combustion air flow rate for the given system I.D. fan.

5.2.4 SO_2 Emissions

SO_2 emissions were recorded under different operational parameters. These parameters included reactor inlet temperature, Ca/S mole ratio, and particle separation efficiency. During this testing, two different sorbents were used. These were sorbents 1 and 2, which were tested under the individual component development aspect of this program. Properties of sorbents 1 and 2 are shown in Table 5.6. The key difference between these sorbents, apart from particle size distribution, was that sorbent 1 was a dolomite containing 60% CaCO_3 and sorbent 2 was limestone containing over 98% CaCO_3 .

Operational data from the SO_2 reactor were achieved in the following manner. Once the reactor reached steady state, dry sorbent was injected at the inlet at a preset flow rate by means of a screw feeder. The reactor was then allowed to reach steady-state conditions. Upon reaching steady-state, typically the sorbent flow rate would be changed to vary the Ca/S mole ratio, and again the reactor was allowed to reach steady-state conditions. Emissions samples were taken at the outlet plenum of the reactor before the gases were turned downward and passed through the heat exchanger. A typical plot of SO_2 emissions as a function of time is shown in Figure 5.42. An interesting feature of Figure 5.42 is that once the sorbent feed is discontinued, it takes a considerable amount of time (over 40 minutes) for the reactor to reach the initial operating conditions, due to the large inventory of sorbent particles confined within the reactor.

Figure 5.40 NO_x Emissions During SO₂ Reactor Operation at Different Input Rates

Figure 5.41 Effect of Input Rate on NO_x Emissions for Kentucky Hazard CWS

TABLE 5.6
SORBENTS USED IN TESTING THE
INTEGRATED EMISSIONS CONTROL SYSTEM

Sorbent	Calcium Compound	Percentage	Mean Diameter (microns)	Origin
1	CaCO₃	60	194	Connecticut
2	CaCO₃	98.7	88.5	Pennsylvania

Figure 5.42 Integrated Emissions Control Reactor Performance
(Kentucky Hazard CWS, Sorbent 2)

5.2.4.1 Effect of Inlet Temperature on Reactor Performance

Because of the small scale associated with the SO₂ reactor, it has a relatively large surface-area-to-volume ratio of 6 ft²/ft³. This can result in significant heat losses even though there is a 3-inch layer of ceramic fiber insulation that separates the reactor liner from the inner wall of the heat exchanger. This large surface-area-to-volume ratio can lead to over an 800°F temperature drop across the reactor.

To compensate for the axial temperature gradient across the reactor, it is necessary to operate the reactor at higher inlet temperatures to obtain higher sorbent utilizations. The effect of reactor inlet temperature on performance is shown in Figure 5.43, which shows that increasing the reactor inlet temperature by 100°F can achieve the same reduction in emissions at a significantly lower Ca/S mole ratio.

To increase sorbent utilization, both operational changes and equipment modifications were made to increase the reactor inlet temperature. These changes included additional insulation in the combustor to limit heat transfer to the cooling water loop, and the system was operated at higher thermal input rates than the nominal design conditions of 125,000 Btu/hr. (The effect of these changes on NO_x emissions is discussed in Section 5.2.3.4.) These changes to the system resulted in reactor inlet temperatures of close to 2,000°F.

5.2.4.2 Effect of Particle Separation on Reactor Performance

With the system configured to operate at elevated reactor inlet temperatures, a series of tests were conducted with both sorbents and with an improved vortex finder at the reactor outlet. The new vortex finder provided a tighter seal at the reactor outlet and thereby improved particle separation within the reactor. Results of these tests are shown in Figure 5.44, in which reactor performance in terms of outlet SO₂ concentration in lbm/MMBtu is plotted as a function of Ca/S mole ratio. The results shown in Figure 5.44 were obtained with the reactor operating with an inlet temperature of just over 2,000°F and with the reactor operating with a pressure drop of approximately 2.5 in. w.c.

The key feature of Figure 5.44 is that as the ability to separate and detain sorbent particles was increased by the addition of the new vortex finder, reactor performance increased dramatically. This was shown in tests conducted with sorbent 2. Without the new vortex finder assembly, the required 90% reduction in SO₂ emissions was achieved at a Ca/S mole ratio of around 7:1; however, with the new vortex finder assembly, the required 90% reduction was achieved at a Ca/S mole ratio of 4.5:1. This ability to increase sorbent particle residence time within

Figure 5.43 Integrated Emissions Control System SO₂ Reactor Performance with Sorbent 2

Figure 5.44 SO_2 Reactor Performance for Different Sorbents (Kentucky Hazard CWS)

the reactor allowed for SO₂ emissions of 0.15 lbm/MMBtu, which represents a 95% reduction compared to uncontrolled emissions and is well below the 0.4 lbm/MMBtu program goal.

To verify the results obtained with the new vortex finder, additional tests were conducted. Figure 5.44 shows the results of prolonged testing conducted at an essentially fixed Ca/S mole ratio of 4:1. These results follow the same trend established in the previous testing and thereby verify the improved performance obtained by increasing the reactor's ability to separate and confine sorbent particles.

The data shown in Figure 5.44 represent SO₂ reduction on an absolute basis, lbm/MMBtu. To put these results on a relative basis, they are plotted in terms of percent reduction as a function of Ca/S mole ratio in Figure 5.45. Figure 5.45 shows that by using sorbent 2 with the improved vortex finder assembly, a 95% reduction in SO₂ emissions can be achieved at a Ca/S mole ratio of 5.5:1.

5.2.4.3 SO₂ Reactor Performance Enhancement Testing

The high temperature, dry sorbent injection approach used in this work was chosen to meet the constraints imposed on residential scale systems. These constraints require the system to have a low first cost, be easy to operate, and require minimal maintenance. High temperature, limestone injection meets all of the constraints and yields a dry non-leaching byproduct, CaSO₃.

A problem with high temperature dry sorbent injection is that sorbent utilization is lower than for wet scrubbing approaches. On the residential scale, the problem of low sorbent utilization is further complicated by the large surface area to volume ratios found in smaller scale equipment. These larger surface area to volume ratios are conducive to significant heat transfer within the SO₂ reactor, even though the system is well insulated. This heat transfer from the hot combustion products to the heat exchanger water jacket results in a significant temperature gradient along the reactor. As the temperature decreases through the reactor, the SO₂ capture process becomes severely kinetics limited.

The purpose of the parametric testing enhancement was to examine methods for increasing sorbent utilization. Two approaches were investigated, the first approach involved the use of partition plates within the reactor to increase the residence time of the sorbent particles within the lower section of the reactor which operates at the highest temperature. The second approach involves the use of a louver separator to increase the separation efficiency of fine sorbent particles within the reactor and thereby further increase the solids inventory available for SO₂ captures.

Figure 5.45 SO₂ Reactor Performance for Different Sorbents (Kentucky Hazard CWS)

Four different partition configurations were tested in the SO₂ emissions control reactor. These configurations are shown schematically in Figures 5.46a and 5.46b and represent several different approaches from successive decreases in partition opening along the reactor axis to having the smallest opening even with the reactor inlet. The sorbent used in testing all of the configurations shown in Figure 5.47 was sorbent 2, a high calcium carbonate (98.7% CaCO₃) limestone from Pennsylvania. A photograph of the reactor with partition plate configuration 3 is shown in Figure 5.47 after a series of tests.

Test results for the four partition configurations are shown in Figure 5.48 along with the standard configuration. These results indicate that the addition of partition plates within the reactor have a minimal impact on reactor performance with respect to SO₂ capture. These results would tend to indicate either of two possible explanations. The first explanation is that all of the SO₂ capture is occurring in the lower section of the reactor and the addition of partition plates to keep the sorbent particles in the lower section of the reactor is not required since the capture process is completed in this lower section. The second explanation is that though the particle inventory in the lower section of the reactor is increased via the use of partition plates, the gas residence time in this section is not sufficient to make additional use of the faster reaction kinetics associated with this higher temperature zone. It was for this reason that the second approach of using the louver separator to increase the solids inventory in the reactor was not tested.

Though the use of partition plates within the reactor had minimal effect on SO₂ capture, a noticeable change in reactor pressure drop was observed. These results are shown in Figure 5.49 where pressure drop is plotted as a function of reactor inlet velocity for the standard configuration without partitions and configuration 4 which had one 4 in. diameter partition plate situated 12 inches above the reactor inlet. Figure 5.49 indicates that the use of partition plates can more than double the reactor pressure drop and since no benefit of greater SO₂ capture was observed, the use of partition plates as a method of increasing sorbent utilization does not appear warranted.

A view of a cross section of the vacuum cylinder constructed to raise the temperature of the flue gas is presented in Figure 5.50 and its placement within the SO₂ reactor is shown in Figure 5.51. The cylinder contains two radiation shields held radially by 16 sets of support studs. The studs are offset to reduce conduction heat losses across the cylinder. An expansion bellows was attached to the bottom of the cylinder allowing for uneven thermal expansion of the cylinder's inner and outer walls due to the temperature gradient across the cylinder. It also provides mechanical support for the radiation shields, which are free to expand at the top of the cylinder.

Figure 5.46a Partition Configurations

Partition Configuration 4

Partition Configuration 3

Figure 5.46b Partition Configurations (Continued)

Figure 5.47 Photograph of Reactor with Partition Plate

Figure 5.48 Partition Configurations 1, 2, 3, and 4

Figure 5.49 Effect of Partitions on Reactor Pressure Drop

Figure 5.50 Cross Section of the SO_2 Reactor Vacuum Bottle

Figure 5.51 Cross Section of SO_2 Reactor with Vacuum Bottle

As shown in Figure 5.51, the cylinder was placed outside of the thermal blanket insulation of the SO₂ reactor and immediately inside the water jacketed downcomer tubes. A photograph of the reactor with the vacuum cylinder installed is shown in Figure 5.52. It was taken before the thermal blanket insulation was installed between the vacuum cylinder and the inner gas duct of the vortical flow reactor.

Use of the cylinder successfully raised the average temperature of the flue gas in the reactor by approximately 200°F in tests conducted using Illinois No. 6 coal with heat inputs of 105,000 to 130,000 Btu/hr. Tests conducted on the integrated system with the vacuum cylinder installed yielded the following results:

Heat Input:	107,000 Btu/hr, 105,000 Btu/hr
NO _x Emissions:	0.2 lbm/MMBtu, 0.2 lbm/MMBtu
SO ₂ Emissions:	0.3 lbm/MMBtu, 0.2 lbm/MMBtu
Ca/S Ratio:	5.77.0
Particulate Emissions:	0.008 lbm/MMBtu, 0.009 lbm/MMBtu

These results show that the use of the vacuum cylinder reduced NO_x emissions while maintaining the SO₂ emissions below the specified goal of 0.4 lbm/MMBtu. However, the cylinder did not have the expected effect of reducing the amount of sorbent required to achieve the specified emissions levels.

The particulate levels measured during testing of the integrated system show that the requirement of 0.02 lbm/MMBtu was easily met. Isokinetic sampling of the baghouse effluent was performed to determine the level of particulate emissions.

TF8-393

Figure 5.52 Photograph of SO_2 Reactor with Vacuum Cylinder Installed

6. MANUFACTURING AND OPERATING COST EVALUATION

To determine the economic incentives for a CWS heating system, a detailed comparison must be made with conventional oil- and gas-fired systems. The high-volume manufacturing cost of a CWS furnace system must be determined, so that this comparison can be made.

6.1 CWS FURNACE SYSTEM HIGH-VOLUME PRODUCTION COST ESTIMATE

A manufacturing prototype unit had to be designed to determine the high-volume production cost of the CWS warm air furnace system. This design had to reflect the constraints imposed by various manufacturing procedures on the high-volume production of warm air furnaces. This design effort was conducted in conjunction with Yukon Energy Inc. Located in Minnesota, Yukon Energy is a manufacturer of residential oil, gas, and multifuel solid-fired warm air furnaces.

One modification made to the design of the laboratory furnace was the removal of the intermediate heat transfer water loop that cooled the combustor and recovered heat from the products of combustion. In the modified design, heat is transferred directly from the products of combustion to the warm air delivered to the heating load, which serves to reduce the cost of the unit. This change in the CWS furnace design was accomplished by having the warm air that passed through the furnace not only recover heat from the heat exchangers but also cool the combustor. To aid in heat transfer, the combustor was redesigned with longitudinal fins.

In an effort to keep the production cost down, the manufacturing prototype furnace was redesigned with as many components as possible that are either directly used in other Yukon furnaces or are slight derivatives. Also, the combustor was designed as a casting to keep costs down even further.

Based on the production of 1,000 units per year, the estimated cost is \$4,000. This cost, which is representative of the price paid by the end user, includes the oil burner needed for startup, the air compressor needed to supply compressed air for the supersonic CWS atomizer and baghouse, and the CWS supply pump as well as the three main components: the combustor, the SO₂ control reactor, and the baghouse. It is likely that with additional value engineering, this cost could still be reduced.

6.2 ECONOMIC EVALUATION: TOTAL LIFE-CYCLE COST

The high-volume cost of the CWS furnace is typically greater than the cost of comparable oil- and gas-fired systems. For this reason, in order for the CWS furnace to be a viable option, over its lifetime its total cost must be less than or equal to the lifetime costs of conventional systems.

For both conventional and CWS fired furnaces, the total lifetime costs must include purchase price, installation, maintenance, and fuel costs, as well as the effects of inflation. The sum of all these factors, translated to present day value, represents the furnace's Total Life-Cycle Cost (TLCC). By determining the system with the lowest TLCC, the system with the best economic merit of rating can be found based on the assumptions of the analysis.

The economic analysis is based primarily on the replacement furnace market. The reason for this is that a large fraction of furnaces are sold for this application. Also, the associated installation costs are much more clearly defined. For example, in new construction, if gas is used, the cost of excavating and installing the gas line can be passed on to the homeowner.

For the purpose of analysis, a comparison was made between the CWS furnace and four other furnace systems, all rated at an input rate of 100,000 Btu/hr, that would be considered for a furnace replacement installation of either an existing oil or gas system. These systems included standard and high-efficiency versions of oil- and gas-fired furnaces. The heating system used in the economic analysis, along with the Annual Fuel Utilization Efficiency (AFUE), purchase price, installation cost, and estimated first year maintenance costs are shown in Table 6.1.

The purchase prices of the commercially available heating systems (shown in Table 6.1) and their associated installation costs were obtained from quotations by various heating contractors. Installation of these systems included any needed duct work to connect the furnace to the existing heating system within the home, electrical wiring, and any gas or oil line plumbing. Also, the installation price included any fireproofing that needed to be done around the furnace. Not included in this price is the cost of the fuel storage tank for either oil or CWS systems.

The standard oil furnace listed in Table 6.1 represents a system that is close to the top of the product line model. This unit had an AFUE of 85.5%, which is very high for a noncondensing furnace. This type of furnace was chosen for analysis since it represents a large fraction of replacement furnace sales. The first-year maintenance costs for this furnace were based on quotations from full-service oil dealers. The price includes an annual cleaning and system tune-up.

TABLE 6.1
WARM AIR HEATING SYSTEMS USED IN ECONOMIC ANALYSIS

System	AFUE (%)	Purchase Price (\$)	Installation Cost (\$)	1st Year Maintenance (\$)
Standard Oil	85.5	1,030.00	1,000.00	75.00
Standard Gas	77.0	532.22	1,000.00	0.00
High Efficiency Oil	91.0	1,758.00	1,000.00	75.00
High Efficiency Gas	92.0	1,097.00	1,000.00	35.00
Coal Water Slurry (CWS)	80.0	4,000.00	1,000.00	100.00

The standard gas furnace listed in Table 6.1 represents a pilot-ignition system that is closer to the bottom of the product line. Though the AFUE for this unit is not as high as for other gas furnaces, the unit is quite low in cost. For this furnace, no first-year maintenance costs were considered since the controls for the furnace are quite simple. Also, since little soot is formed while the unit is burning natural gas, heat exchanger cleaning was assumed to be unnecessary.

The high-efficiency oil furnace listed in Table 6.1 represents a top-of-the-line condensing unit. This unit has an AFUE 5.5 points higher than the standard oil unit; however, it costs \$728 more. The first-year maintenance costs for this unit are the same as those for the standard oil unit, i.e., \$75.

The high-efficiency gas furnace listed in Table 6.1 also represents a top-of-the-line condensing unit. This furnace has an AFUE 15 points higher than the standard gas unit used in this analysis, and costs \$564.78 more. Since this furnace is much more sophisticated than the simple standard gas unit, a first-year maintenance cost of \$35 was assumed. This cost includes the replacement of the condensate acid neutralizing canister that is needed to neutralize the acidity of the furnace condensate before it can be discharged into a sewer or septic system.

For the CWS furnace listed in Table 6.1, various assumptions had to be made about installation cost, AFUE, and maintenance during the first year. For installation, a cost of \$1,000 was assumed. This should be a reasonable assumption since, due to the packaged nature of the furnace, installation should not cost any more than traditional oil and gas furnaces.

The AFUE efficiency for the CWS furnace was chosen to be 80%. In the original specifications for this furnace, a steady-state efficiency of 85% was chosen. Since AFUE is often less than the steady-state efficiency, to be conservative, the steady-state efficiency was penalized by 5 points to obtain the AFUE.

In estimating the maintenance cost, it was assumed that \$100 worth of maintenance would be required after the first heating season. This cost is higher than the cost for the oil-fired systems since a much more thorough cleaning of the heat transfer surfaces is expected. Also, since the CWS furnace requires an oil burner for startup, the burner must be serviced as well as the CWS combustor.

To perform the economic analysis, various assumptions had to be made. These assumptions are summarized in Table 6.2 and are based on heating a home that has an overall heat transfer coefficient surface area product (UA) of 700 Btu/hr-°F. This is representative of a newly constructed home with about 1,850 square feet of living area. The average number of degree days for this home was chosen to be 5000°F-days per year.

TABLE 6.2
ASSUMPTIONS USED IN ECONOMIC ANALYSIS

Building Heating Load (UA Product Btu/hr-°F)	700
Number of Annual Degree Days (°F-Days)	5000
General Interest Rate (%)	8.00
Fuel Limestone and Waste Removal Inflation Rate (%)	6.00
Maintenance Inflation Rate (%)	5.00
Heating System Life (years)	20
Limestone Cost (\$/ton)	100
Waste Removal (\$/ton)	100

To account for variations in the value of money with time, a general interest rate of 8% was assumed to be available to the homeowner. To account for changes in the price of fuel and maintenance, inflation rates of 6% and 5% were assumed, respectively. For this analysis, the economic life of the heating system was chosen to be 20 years.

The goal of the economic analysis was to determine for various costs of oil and gas, the price that CWS needs to be to have a TLCC equal to or less than that of conventional heating systems. This is shown in Figure 6.1 where the ratio of the TLCC of CWS to oil is plotted against the CWS-to-fuel-oil cost ratio, as a function of the price of oil. For the CWS furnace to obtain economic parity with the standard oil system, the ratio of the total life-cycle cost must be equal to one. Any value less than one results in the CWS furnace having a lifetime economic advantage over the oil-fired system.

In referring to Figure 6.1, economic parity is achieved at fuel cost ratios of 0.22, 0.43, 0.54, and 0.64 for oil prices of 5, 7, 9, and 12 dollars per million Btus fired. Based on these results, the price of CWS needs to be 1.1, 3.01, 4.86, and 7.68 dollars per million Btu for oil prices of 5, 7, 9, and 12 dollars per million Btu, respectively. If CWS can be produced for these values or less, the CWS furnace expenditures will be equal to or less than the total expenditures for the oil system over its life.

A comparison of the total life-cycle cost ratio for a CWS furnace with a standard gas furnace is shown in Figure 6.2. Results of this comparison show that in order to obtain economic parity over the life of the system, i.e., having the TLCC ratio equal to one, CWS must cost 79, 58, 47, and 37% less than gas for gas prices of 5, 7, 9, and 12 dollars per million Btu, respectively. This translates into CWS prices being 1.05, 2.94, 4.77, and 7.56 dollars per million Btu for the range of gas prices considered.

The previous comparison between CWS and standard oil- and gas-fired systems was based on the furnaces being located in a region having an average annual degree day heating season of 5,000°F-days. To extend this analysis to various other regions of the country where the number of degree days can vary widely, a comparison was made between CWS and standard oil systems with the dependent variable being the number of degree days required for both the CWS and standard oil system to have the same total life-cycle cost, i.e., to break even.

In this analysis, the CWS furnace is compared only with the standard oil system. The reason for this is that both are liquid fueled systems and any additional costs for converting from oil to CWS should be minimal since there is

Figure 6.1 Economic Comparison of CWS Furnace System with Standard Oil System

Figure 6.2 Economic Comparison of CWS Furnace System with Standard Gas System

already a storage tank and delivery system on site. Results of this comparison are shown in Figure 6.3, which plots the required number of annual degree days to achieve the break-even point against the price of CWS for various oil prices.

The use of the required number of degree days to achieve the break-even point works in the following way. For a given cost of CWS and oil, the number of degree days required for the total life cycle of the CWS furnace to equal that of the oil system can be found. The ratio of this number to the actual number of annual degree days for a given location indicates whether CWS is the better economic choice. If the ratio is equal to one, then both systems have the same total life-cycle cost. However, if it is less than one, then the CWS furnace has a lower total life-cycle cost and is therefore the better economic choice.

To aid in the use of Figure 6.3, Table 6.3 shows the average annual amount of degree days for 50 different U.S. cities. Degree days in Table 6.3 range from 10,364 for Homer, Alaska to 0 in Honolulu, Hawaii.

As an example of how to use Figure 6.3 and Table 6.3, consider CWS costing \$4.25 per million Btu and oil costing \$7 per million Btu. From Figure 6.3, about 4,400 degree days are required for both the CWS and standard oil systems to have the same total life-cycle cost. If the systems were located in Boston, MA, which has 5,618 degree days, then the ratio of heating degree days is 0.78 and the CWS furnace is a better economic choice than the standard oil system. However, if the systems were located in Norfolk, VA, which only has 3,487 degree days, then this ratio is 1.26 and the standard oil system is a better economic choice.

Another feature that can be seen in Figure 6.3 is that as the price of CWS gets closer to the price for oil, the number of required annual degree days approaches infinity. This is due to the fact that since the CWS furnace has a greater first cost, in order to have the same TLCC, CWS must cost less than oil.

The results shown in Figure 6.3 are based on the model home having an overall heat transfer coefficient surface area (UA) product of 700 Btu/hr-°F. To determine the effect of the building's UA product on economics, an analysis was conducted for UA products of 500, 700, 900, and 1,000 Btu/hr-°F. In this analysis, the CWS furnace was compared to the standard oil furnace with an oil price of \$7 per million Btu. The results of this analysis are shown in Figure 6.4, where, as expected, for a given price of CWS, fewer degree days are required for TLCC parity between CWS and oil, for higher UA products due to the greater utilization of the furnace. For this reason, more fuel is required, and by having CWS at a lower cost than oil, the difference in fuel cost has a more significant impact on TLCC.

Figure 6.3 Total Life Cycle Cost Break-Even Comparison of CWS Furnace System with a Standard Oil System

TABLE 6.3
DEGREE DAYS FOR VARIOUS U.S. CITIES

City	State	Degree (F) Days
Homer	AK	10,384
Birmingham	AL	2,840
Little Rock	AR	3,353
Phoenix	AZ	1,552
Los Angeles	CA	1,818
San Francisco	CA	3,046
Denver	CO	6,017
Hartford	CT	6,349
Wilmington	DE	4,941
Miami	FL	203
Atlanta	GA	3,094
Honolulu	HI	0
Des Moines	IA	6,707
Boise	ID	5,837
Chicago	IL	6,129
Indianapolis	IN	5,576
Wichita	KS	4,691
Louisville	KY	4,642
New Orleans	LA	1,463
Boston	MA	5,618
Baltimore	MD	4,719
Portland	ME	7,501
Detroit	MI	6,226
Minneapolis	MN	8,158
St. Louis	MO	4,570
Jackson	MS	2,300
Billings	MT	7,267
Charlotte	NC	3,218
Bismarck	ND	9,043
Scottsbluff	NE	6,775
Concord	NH	7,358
Newark	NJ	5,035
Albuquerque	NM	4,283
Las Vegas	NV	2,601
New York	NY	4,847
Cincinnati	OH	5,069
Tulsa	OK	3,681
Portland	OR	4,792
Pittsburgh	PA	5,931
Charleston	SC	2,147
Rapid City	SD	7,319
Memphis	TN	3,227
Houston	TX	1,433
Salt Lake City	UT	5,983
Norfolk	VA	3,487
Burlington	VT	7,877
Seattle	WA	5,186
Milwaukee	WI	7,443
Huntington	WV	4,624
Casper	WY	7,555

Figure 6.4 Effect of Heating Load Size on CWS Furnace System Economics for Oil Costing \$7/MMBtu

6.3 ECONOMIC EVALUATION: PAYBACK PERIOD

In the previous economic analysis, a 20 year life cycle was considered since this criterion was established for the operational life of the furnace. However, in making a choice to buy a CWS furnace over a conventional oil-fired system, a prime economic consideration is payback. The payback period can best be defined as the amount of time necessary for the accumulated savings to equal the investment. When comparing oil and CWS fired systems, this occurs when the total life-cycle costs for each system are equal.

To determine the effect of the time period being considered on the CWS furnace economics, the analysis was extended to cover 5-, 10- and 15-year life cycles. For this analysis, the assumptions about home size, general interest, and inflation rates were the same as in Table 6.1. Also, the comparison was made between the CWS furnace and the standard oil system only. In comparing the two systems, the annual number of required degree days approach was chosen because it allows the analysis to be extended to various geographic regions.

The effect of time on the economic performance of the CWS furnace is shown in Figure 6.5, in which the number of degree days per year required for break even is plotted against the fuel cost differential between oil and CWS. In referring to Figure 6.5, it can be seen that when the analysis is based on a 20 year life cycle, if the fuel differential cost (oil cost minus CWS cost) is \$3/MMBtu, then the furnace is a good economic choice in any region which has more than 4,000 degree days per year. This corresponds to the vast majority of the United States, and would be representative of CWS costing \$4/MMBtu and oil costing \$7/MMBtu, which is equivalent to \$0.96 per gallon.

When the life cycle period is considered to be 10 years, it can be seen from Figure 6.5 that to obtain the same number of required degree days, the cost differential must increase from \$3/MMBtu for the 20 year case to \$6/MMBtu for the 10 year case. If CWS is assumed to be available at \$4/MMBtu, then oil must cost \$10/MMBtu or \$1.38 per gallon to obtain a 10 year payback.

When the life cycle period is considered to be 5 years, there is a large increase in the number of required degree days needed to break even as compared to the 10 year period. For example, consider Boston, MA, which annually has 5,618 degree days. To obtain a 5 year payback for a CWS system compared to an oil-fired system, from Figure 6.5, oil must cost \$7.25/MMBtu more than CWS. If CWS is assumed to be available at \$4/MMBtu, then oil must cost \$11.25/MMBtu or \$1.58 per gallon. Oil prices this high have occurred in Boston, but were due to supply shortages and remained at that level for only a few weeks.

Figure 6.5 Effect of Life Cycle Duration on CWS Furnace System Economics

7. CONCLUSIONS AND RECOMMENDATIONS

During the program, a system to control the emissions from small scale coal fired combustion systems was developed and demonstrated. The emissions goals were achieved. NO_x emissions were less than or equal to 0.2 lb/MMBtu, SO_2 emissions were less than 0.4 lb/MMBtu and particulates were less than or equal to 0.02 lb/MMBtu. The actual emissions achieved were much better than the program goals. Tests of the integrated system demonstrated that SO_2 emissions could be controlled to levels half of the program goal and particulate emissions could be controlled to levels 60% less than the program goal while maintaining NO_x emissions at the program goal.

The control of these emissions to such low levels is remarkable. It clearly demonstrates the effectiveness of the technology. In particular, for the SO_2 control reactor to perform to this level given the reactor residence time demonstrates the effectiveness of the reactor as a gas solid contacting device. Based upon the performance of this reactor, it is recommended that further work be conducted to scale this technology to the utility scale. Foreign manufacturers are developing low temperature dry SO_2 control reactors based on circulating fluidized bed (CFB) technology. The performance of the Tecogen reactor is superior to the SO_2 control that can be achieved in CFB combustors operating at the same temperature. The CFB units require gas residence times an order of magnitude greater than the Tecogen reactor, 3 seconds compared to 0.3 seconds. Given this comparison, the Tecogen reactor technology should be scaled up to develop a U.S. based highly efficient, low temperature, dry SO_2 control system.

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