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GASIFICATION IN PULVERIZED COAL FLAMES

First Quarterly Progress Report

July-September 1975

N. M. Laurendeau

The Combustion Laboratory
School of Mechanical Engineering
Purdue University
West Lafayette, Indiana 47907

Report No. PURDU-CL-75-06

October 1975

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16. Abstracts

The objective of this program is to investigate the feasibility of using currently available pulverized coal burners to produce power or synthesis gas from coal. Two configurations will be considered: (1) the annular confined jet with secondary swirl, and (2) the vortex tube with tangential entry. The first burner is characterized by a single axial injector of high primary velocity; secondary swirl is used to control mixing and residence times. The second burner is modeled after the cyclone combustor; large residence times and slagging operation should lead to high carbon efficiencies.

Species concentrations and temperature are measured both within and downstream of the gasifier chambers. These profiles are used to assess the influence of process variables such as pressure, solid/gas feed rates, swirl intensity, inlet temperature and geometrical injection pattern on both the rate and extent of coal conversion. Simple models governing entrained flow systems will be developed to further interpret the experimental data.

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ABSTRACT

This project is concerned with the production of power and synthesis gases from pulverized coal via suspension gasification. Specifically, the concentric jet and vortex gasifiers, with separation of oxidation and reduction zones, will be investigated. Gasifier performance will be correlated with internally measured temperature and concentration profiles. A suction pyrometer will be used to simultaneously measure temperature and gas concentrations. Rapid species analysis will be provided by a UTI Q-30C mass spectrometer system.

To date, we have developed a coal handling facility, initiated test cell design, and made preliminary literature searches. A coal crusher, pulverizer, feeder and sieve shaker are on order. Preliminary consultations are underway concerning the mass spectrometer system.

I. OBJECTIVE AND SCOPE OF WORK

The objective of this program is to investigate the feasibility of using currently available pulverized coal burners to produce power or synthesis gas from coal. Two configurations will be considered: (1) the annular confined jet with secondary swirl, and (2) the vortex tube with tangential entry. The first burner is characterized by a single axial injector of high primary velocity; secondary swirl is used to control mixing and residence times. The second burner is modeled after the cyclone combustor; large residence times and slagging operation should lead to high carbon efficiencies.

In both burners, coal combustion with air and/or air-oxygen mixtures supports the endothermic gasification reactions. The burners are designed such that axially injected, preheated steam is surrounded by swirling air, thus promoting efficient gasification. Pulverized coal is carried by primary steam in the confined jet system, and by secondary air in the vortex gasifier.

Species concentrations and temperature are measured both within and downstream of the gasifier chambers. These profiles are used to assess the influence of process variables such as pressure, solid/gas feed rates, swirl intensity, inlet temperature and geometrical injection pattern on both the rate and extent of coal conversion. Simple models governing entrained flow systems will be developed to further interpret the experimental data.

II. SUMMARY OF PROGRESS

By surveying modern gasification processes (Appendix A), we have reached the following conclusions:

(1) Suspension gasification has many advantages but has received little consideration since the late 1950's.

(2) Power and synthesis gas production can be optimized by separating oxidizing and reducing zones in the gasifier.

(3) The concentric jet and vortex tube gasifiers with

separate coal/steam injection appear to fulfill the above criterion.

(4) Gasification and carbon efficiency are best correlated with (a) coal throughput, (b) input oxidizer/coal and steam/coal ratios and (c) internal temperature and composition profiles. The latter provides impetus for our main concern: measurements internal to the gasifier.

In designing the test cell, we will develop a common control panel and flow system for the two pulverized coal gasifiers. The system will operate at 1-30 atms. A suction pyrometer will provide simultaneous measurement of temperature and gas concentrations. Preliminary discussions concerning the UTI Q-30C mass spectrometer system have been initiated; a pulverized coal feeder for atmospheric operation has been ordered. Test cell design is now underway.

To avoid long delays in fuel acquisition, we have designed a separate coal handling facility. A crusher, pulverizer, and sieve shaker (with appropriate sieve equipment) has been ordered. We believe that auxiliary equipment, such as a laboratory hood, balance and tables are available from Purdue University.

III. TECHNICAL PROGRESS

To date, we have developed a coal handling facility, initiated test cell design, and made preliminary literature searches. A coal crusher, pulverizer, feeder and sieve shaker are on order. Consultations are underway concerning the mass spectrometer system. Preliminary design criteria have been established for the two pulverized coal gasifiers (Appendix A).

Experimental Equipment and Procedures

Both the confined jet and vortex gasifiers will be externally heated to compensate for the large heat losses characteristic of laboratory size equipment. In this way, internal temperatures can be regulated to maintain reactor stability and simulate large-scale operation. System shakedown will proceed in the following order: (1) coal/air combustion at atmospheric conditions, (2) coal/air/oxygen/

steam gasification at atmospheric conditions and (3) coal/air/oxygen/ steam gasification at higher pressures (1-20 atm).

Pulverized bituminous coal (Indiana/Illinois) will be fed at 5-50 lb/hr. Successful operation with a bituminous coal usually indicates that lignites or subbituminous coals will work, since gasification efficiency increases as coal rank decreases. Comparable steam flows (5-50 lb/hr) will be provided by a small package boiler. Castable ceramic will be used for the gasifier walls; RA 330 or stainless steel may be suitable for the confined jet system. Probe openings and quartz windows will require careful design procedures.

1. Coal Pulverization and Feeding

Separate facilities have been cleared for coal handling, crushing and pulverizing. A dust-free coal crusher (100 lb/hr) and pulverizer (30 lb/hr) have been ordered from Holmes, Inc., Danville, Illinois. The Gilson sieve shaker and associated sieve equipment is also on order; a chemical hood will be necessary to avoid excessive dust during particle size analyses and sieve cleanup. Respirators will be required for protection during crushing, pulverizing and sizing operations.

Feeding pulverized coal at steady rates is a difficult problem, especially at high pressures. For atmospheric operation, Vibra-screw Feeders guarantee 2% accuracy in feed rate; one such feeder is on order. J. Mima of ERDA-Pittsburgh is forwarding a recent design for a high pressure laboratory feeder, based on earlier BCR work. He claims that this feeder is significantly better than any previous design considered by the Bureau of Mines.

2. Temperature and Composition Measurements

Internal measurements of temperature and composition will not be straightforward, due to the presence of particulates. We hope to measure gaseous species and temperature simultaneously, by using a specially constructed suction (sonic) pyrometer. The probe size will be rather large (.5-1.0 cm) to avoid clogging; reverse flow of inert gas will minimize particulate flow through the probe. The probe will be of the triple-walled type; water cooling is anticipated though steam cooling may be necessary to avoid water condensation. A small cyclone and line filters will be used to separate solids and gases as quickly as possible to avoid catalytic effects. The probe will be inserted through a water-cooled elbow. Exhaust measurements will be made initially; internal probing must be done carefully to minimize particulate effects.

We have made contact with UTI concerning their Q-30C mass spectrometer system. They are now in the process of responding to a series of questions that will allow us to assess the adaptability and accuracy of their system for coal gasification studies. Of particular interest are the (1) accuracy of hydrogen measurements, (2) problems with steam measurements and (3) precautions required to avoid particulate flow within the mass spectrometer. A multiple least-squares analysis program will be used to calculate CH_4, CO, CO_2, H_2, O_2, N_2 and H_2O concentrations. If possible, pollutants (NH_3, NO, H_2S, SO_2) will also be measured.

3. Control Cell and Gasifier Designs

A test cell with explosion-proof walls has been selected to house both the confined jet and vortex tube gasifiers. Common furnace and flow controls will be used; hence, simultaneous gasifier operation will not be possible. Flow control panels and piping charts are in the initial design phase. Literature searches are underway to provide the necessary information for gasifier design.

IV. WORK FORECAST

For the entire term of this contract, we expect to meet the following schedule:

(1) Preliminary design of coal handling facilities, control panels, piping scheme and gasifiers. Purchase crusher, pulverizer, sieve shaker and attachments, flow equipment and mass spectrometer. Initiate literature search. (6 months)

(2) Final design and construction of test cell, including control panel, flow and furnace facilities, low pressure feeder and steam generator. Purchase feeder, valves and gauges, package boiler and furnaces. Complete literature survey and gasifier design. Preliminary construction of gasifiers. (6 months).

(3) Complete gasifier construction. Build probes and integrate with thermocouple gauges and mass spectrometer. Complete purchasing of major equipment. Test cell and gasifier shakedown. Initiate coal/air combustion at 1 atm. (6 months)

(4) Finish measurements and interpretation of results for coal combustion at 1 atm. Initiate survey of entrained flow models. Initiate coal gasification experiments at 1 atm. (6 months)

(5) Complete measurements and interpretation of results for coal gasification at 1 atm. Finish entrained flow models survey and apply if possible to coal combustion and gasification measurements. Construct high pressure coal feeder. (6 months)

(6) Check feasibility of coal combustion and gasification at higher pressures. (6 months)

During the next quarter, we will order the mass spectrometer and flow equipment. We will complete the cell design and initiate construction of the control panel and piping scheme. We will continue our literature search, stressing the following: (1) flame shapes, (2) experimental results, (3) controlling parameters, (4) new designs, (5) theoretical models and (6) predictive procedures. Upon receiving the coal handling equipment, we will design and construct the appropriate facilities for crushing, pulverizing and sizing bituminous coal.

V. RESULTS AND CONCLUSIONS

A survey of modern gasification processes (Appendix A) leads to the following conclusions:

1. Coal gasification is a versatile, environmentally acceptable solution to our short term problems of energy adequacy. However, if practical stack or <u>in situ</u> sulfur removal methods are developed,

fuel economy may favor direct coal combustion.

2. Compared to other gasification products, power gas synthesis represents the most efficient use of our coal resources. Optimization of its heating value demands that we minimize CO_2 and H_2O, yet maximize temperature, CH_4, and CO/H_2.

3. Due to the high nitrogen content of coal, gasification processes using high temperature sulfur removal may produce excessive levels of NH_3; combustion of producer gases will then violate NO_x standards.

4. We lack a fundamental understanding of many chemical and physical processes controlling producer gas formation. The following are high priority research items: (1) pyrolysis/devolatilization kinetics, (2) turbulent gas-solid interactions, (3) gasification kinetics, (4) pore diffusion/kinetics models. Only basic experimental investigation can determine the limitations, performance, and pollutant characteristics of various gasification schemes.

5. High temperature (1400-1600°K) fluidized bed processes are now possible, though requiring perfection of Godel's ash-agglomerating method and Squires' so-called fast fluidized bed. Development of high pressure, in situ sulfur removal techniques will make fluidized bed operation even more attractive. Caking coals and solid disposal still present difficult design problems.

6. Entrained flow methods must be developed, as they show significant advantages compared to fixed and fluidized bed techniques. In particular, pulverized coal operation promotes high reaction rates per unit volume; hence, high temperatures and thus slagging conditions. Caking coals and slurries are acceptable; clinker and tar formation are negligible.

7. Economic production of a power gas of high energy content will require (1) high pressures (20 atm) and temperatures (1400-1800°K), (2) high carbon efficiency, (3) separation of oxidizing and reducing zones, (4) short gasifier residence times and (5) single stage processes (possibly). Pulverized coal burners (concentric jet or cyclone) with coal-steam separation appear to fulfill these criteria.

8. The most useful independent process variables are (1) coal throughput, (2) input oxidizer/coal and steam/coal ratios and (3) internal temperature and composition profiles. Gasifier performance can increase, decrease or peak, depending on the gasifier design and the range of independent process variables. No internal measurements during suspension gasification have been attempted previously.

In developing preliminary experimental procedures, we have reached the following conclusions:

1. The confined jet and vortex tube designs are best suited for pulverized coal gasification with separation of oxidation and reduction zones. Extrapolation of our results to large-scale systems appears feasible.

2. Coal handling facilities are required to avoid long delays in fuel acquisition.

3. A common control panel is advantageous; experimental delays are not anticipated.

4. A suction pyrometer should be developed since simultaneous measurement of temperature and gas concentrations is possible. High particulate loadings may pose severe problems.

5. At present, the UTI Q-30C mass spectrometer system has not been challenged by other methods for ease of operation, versatility and rapidity.

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VII. APPENDIX A: CONCEPTS GOVERNING POWER GAS PRODUCTION IN ENTRAINED FLOWS

Introduction

The energy crisis demands greater use of our vast coal resources (Hottel and Howard, 1971). An environmentally acceptable method is coal gasification; in particular, gasification to synthesis and power gases (Squires, 1974; Osborn, 1974). Synthesis gas is a major feedstock for the production of methane, methanol, hydrogen, ammonia and liquid hydrocarbons. Clean power gas may be used as an industrial fuel, or more importantly, in combined cycles, boilers and MHD devices to generate electricity.

In general, coal consumption is hampered by four major problems: (1) mine safety, (2) transportation costs, (3) water requirements and (4) sulfur/nitrogen removal. Fortunately, the last three drawbacks can be largely eliminated by gasification-slurry pipeline systems (Laurendeau, 1975). Conversion of coal to power gas will minimize SO_x and NO_x, since H_2S and NH_3 are favored under gasification conditions. Stack gas cleanup will remove the latter compounds; ammonia can be sold to the fertilizer industry (Laurendeau, 1975).

Coal Gasification

This investigation is primarily concerned with the formation of a clean power or synthesis gas from coal. As indicated in Table I, synthesis gas is predominantly composed of carbon monoxide and hydrogen. Power gas is essentially a synthesis gas diluted with nitrogen. Its heating value is about one-half that of synthesis gas and one-sixth that of natural gas. Mixtures such as synthesis gas or power gas are sometimes called producer gas.

Producer gases are primarily generated by the endothermic steam-carbon reaction,

$$C(s) + H_2O\ (v) \rightarrow CO + H_2 \qquad \Delta H^o_{298} = 31.38\ \text{kcal/gmole},$$

which is favored at high temperatures ($T > 1200^oK$) and moderate pressures ($P < 20$ atm). Further gasification occurs via

$$C + CO_2 \rightarrow 2CO \qquad \Delta H^o_{298} = 41.21\ \text{kcal/gmole}.$$

Initial producer gas formation is, of course, a result of rapid coal pyrolysis. Final product composition is often determined by water-gas equilibrium:

$$CO + H_2O \rightleftarrows CO_2 + H_2 \qquad \Delta H^o_{298} = -9.83\ \text{kcal/gmole}.$$

As suggested by Table I, significant increases in heating value result if even small levels of methane are produced. Most CH_4 formation occurs via devolatilization; high pressure char hydrogasification ($C + 2H_2 \rightarrow CH_4$) can also contribute.

The necessary gasification temperatures are usually obtained by burning a portion of the coal with air or oxygen:

$$C + 1/2\ O_2 \rightarrow CO \qquad \Delta H^o_{298} = -26.42\ \text{kcal/gmole}$$

$$C + O_2 \rightarrow CO_2 \qquad \Delta H^o_{298} = -94.04\ \text{kcal/gmole}.$$

The use of ocygen results in synthesis gas; the use of air results in a low BTU power gas, due to the large quantity of nitrogen in the

TABLE I

PRODUCTS OF COAL GASIFICATION

Product	Major Components	HHV(BTU/SCF)[a]
SNG	CH_4	900-1050
Synthesis gas	CO, H_2	250-400
Power gas	CO, H_2, N_2	100-250

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[a] Higher heating value of the clean, dry product.

final mixture. In either case, maximization of the $CO/H_2/CH_4$ content and minimization of the CO_2/H_2O content of the product will clearly optimize its energy value (Laurendeau, 1975). For power gases, high CO/H_2 ratios are favored since most combustion processes do not allow water condensation (Squires, 1974).

For chemical equilibrium at 1500°K, the gasification process may be represented by

$$2C + H_2O + O_2 + 4N_2 \rightarrow 0.53\ CO_2 + 0.53\ H_2 + 1.47\ CO + 0.47\ H_2O + 4N_2.$$

Air and steam must be preheated to about 550°K to maintain adiabatic conditions. Note that water-gas equilibrium clearly predicts the impossibility of simultaneously reducing the H_2/CO ratio, CO_2 and

H_2O. Thus, practical single stage processes should operate at residence times controlled by chemical kinetics and/or turbulent mixing.

Realistic two-phase gasification models, incorporating both fluid mechanics and chemical kinetics are not available (Laurendeau, 1975). Knowledge of even homogeneous turbulent reactions is minimal; furthermore, simple overall rate data governing the reactions between pulverized char and O_2, H_2O, CO_2 or H_2 are not reliable (Mulcahy and Smith, 1969; von Fredersdorff and Elliott, 1963). Moreover, such data are only applicable over limited ranges of temperature, pressure, stoichiometry and coal type.

Coals generally react in two somewhat separable stages (Howard and Essenhigh, 1967). Initially, heat transfer to fresh coal particles causes pyrolysis and devolatilization; residual chars subsequently undergo heterogeneous combustion or gasification. Gasification is slow compared to combustion and pyrolysis; hence gasification kinetics will control residence time requirements.

The type and rate of volatile evolution depends mostly on local temperature history (Howard and Essenhigh, 1967). Pulverized coal combustion however depends on both particle size and temperature, since these parameters control the relative effects of mass diffusion and chemical kinetics (Sergeant and Smith, 1973; Mulcahy and Smith, 1969). The slow gasification reactions are often controlled solely by chemical kinetics. However, complications persist since most chars react both externally and internally, due to the existence of an extensive pore structure. Moreover, the pore structure itself changes dramatically during reaction.

Coal gasification favors formation of H_2S and NH_3, compared to SO_x and NO_x, due to the availability of molecular hydrogen. Commercial absorption methods remove both H_2S and NH_3 via liquid scrubbing near 400°K. High temperature H_2S cleanup ($T > 1200^\circ K$) via in situ or exhaust scrubber systems will improve gasifier thermal efficiency, but leaves ammonia in the producer gas. Combustion will probably allow conversion of over half of the NH_3 to NO. As a result, NO_x emissions approach the 2 lb/10^6 BTU of coal level, an unacceptable value (Laurendeau, 1975).

Of the three gasification products (Table I), power gas is the least expensive and makes the best use of our coal resources. Production of synthesis gas requires an oxygen separation plant, which represents a 40% increase in capital cost, plus a 10% increase in energy input (Farnsworth et al. 1973). Conversion of synthesis gas to SNG entails a 20% energy loss, plus expensive hardware (Squires, 1974). Moreover, for many purposes, we are really interested in the heating value per volume of product, not per volume of fuel. On this basis, power gas has a heating value only 15% less than that of natural gas (Squires, 1974).

Since synthesis gas is a chemical feedstock, maximization of $CO + H_2$ content, or chemical enthalpy, is important. However, low BTU fuels for combined cycles must be available at high temperatures (1300-1700°K) and pressures (15-30 atm) to optimize overall thermal efficiency. Hence, for power gas, we must maximize chemical plus sensible enthalpy, yet minimize exhaust levels of ammonia and carbon.

Gasification Processes

Three methods are available for the commercial production of a producer gas from coal: (1) stationary fuel beds, (2) fluidized beds, and (3) suspension beds. In general, fixed beds employ lump coal (1-10 cm), fluidized beds crushed coal (10^{-2} - 1 cm) and suspension beds pulverized coal (10^{-4} - 10^{-2} cm). At present, the Lurgi process (a high pressure stationary bed technique) is the principle industrial method for the manufacture of producer gas, and the only method compatible with combined cycle operation. Fluidized bed and entrained flow processes are inherently superior, and are only now receiving warranted attention. The advantages and disadvantages of each method are listed in Table II (Laurendeau, 1975).

The production of power or synthesis gas may be accomplished by either a two-stage (Table III) or single-stage (Table IV) process (Laurendeau, 1975). Single-stage gasification processes are characterized by (1) simplicity of design, (2) high product temperature and

TABLE II

REACTOR TYPES FOR COAL GASIFICATION

Type	Advantages	Disadvantages
Fixed	Developed technology, countercurrent flow, high pressure operation	Non-uniform flow, clinker formation, temperature control, ash removal, tar formation, low capacity, coal fines unacceptable, high H_2O levels
Fluidized	Solids handling, uniform temperature, gas solid contact, countercurrent flow, *in situ* sulfur removal, high gasification rate	Pretreatment of caking coals, multistaged beds, carbon efficiency, ash disposal
Entrained	Highest volumetric reaction rate, slagging conditions, gas-solid contact, coal type irrelevant, high product temperature, compatible with coal slurries and froth flotation	Ash separation, temperature control, co-current flow, coal pulverization

TABLE III

POWER GAS PRODUCTION VIA TWO-STAGE
COAL GASIFICATION PROCESSES

Process	Type	P(atm)	HHV(BTU/SCF)[a]
BI-GAS	entrained	70-100	175
SYNTHANE	fluidized	30-70	200
HYGAS	fluidized	70	235
Westinghouse	fluidized	10-20	140
BCR Multibed	fluidized	16	150

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[a] Higher heating value of the raw gas on a dry basis

(3) low methane content. Two-stage methods promote methane enrichment by separating gasification and pyrolysis steps. As a result, the final product possesses a higher chemical heating value, but its temperature is limited to 1000°K (Laurendeau, 1975).

1. Counter-current Methods

Standard stationary and fluidized methods operate via an upward flow of air and steam through a coal bed. In both cases, the resulting gas composition is largely determined by the bed depth and the flow velocity. In a stationary bed, large coal chunks rest on a fixed or moving grate, while in a fluidized bed, small pieces of coal are held in turbulent suspension by the rapidly moving flow.

In a fixed bed gasifier, optimization of energy content is promoted by a natural separation of the oxidizing and reducing zones (Laurendeau, 1975). The exothermic carbon-oxygen reaction dominates in the lower portion of the bed, thus supplying heat for the endothermic carbon-steam and carbon-carbon dioxide reactions in the upper portion. Pulverized coal gasifiers should make use of this principle,

TABLE IV

SINGLE-STAGE COAL GASIFICATION PROCESSES

Process[a]	Type	Ash Removal	Product[b]	P(atm)	T(^{o}K)	HHV(BTU/SCF)[c]
Gas Producer (C)	fixed	agglomerates	PG,SG	1	$\sim$1000	140-280
Stirred Producer	fixed	agglomerates	PG	7	$\sim$900	150
Lurgi (C)	fixed	agglomerates	PG	2-30	850	165
			SG	2-40	900-1100	300
Winkler (C)	fluidized	powder	PG	1	1250	120
			SG	1	1100-1400	275
U-GAS	fluidized	agglomerates	PG	24	1300	150
Ignifluid (C)	fluidized	agglomerates	PG	1	1450-1550	140
Koppers-Totzek (C)	entrained	slag	SG	1	2200	300
Combustion Engineering	entrained	slag	PG,SG	1	1400-2000	125-285
Texaco Partial Oxidation	entrained	slag	PG	15	1500-1800	140

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[a] (C) denotes a commercial process

[b] PG - power gas; SG - synthesis gas

[c] Higher heating value of the raw gas on a dry basis

thus increasing gasification efficiency.

Renewed interest in fluidized bed methods has been spurred by recent increases in operational temperatures. Early processes were limited to temperatures (1200-1300^{o}K) somewhat lower than the ash fusion temperature ($\sim$ 1500^{o}K) to avoid clinker formation. However, the development of Godel's ash-agglomerating fluidized bed (Ignifluid) allows temperatures of 1450-1550^{o}K. Here, high fluidization velocities apparently limit clinker size; carbon losses are minimized by recycling coal fines (Squires, 1974). The ash-agglomerating fast-fluidized bed (Squires, 1974) suggests even higher fluidization velocities. The U-GAS and Westinghouse processes are being developed along these lines (Tables III-IV).

Both two-stage and single-stage counter-current methods have been reviewed by Laurendeau (1975) and Bodle and Vyas (1973). Two-stage producers are usually overdesigned, since they were originally intended for SNG production. Exceptions are the Westinghouse and BCR Methods (Table III). Single-stage methods, on the other hand, have been explicitly designed for manufacturing producer gas. Unfortunately, counter-current methods still suffer due to (1) low temperatures, (2) pretreatment/coking coal problems, (3) high H_2O and H_2/CO levels and (4) carbon efficiency problems.

2. Co-current Methods

Entrained flow methods appear to offer the most viable means of using U. S. bituminous coals (Table II). Experience with pulverized coal burners indicates that either caking or non-caking coals are acceptable. In addition, entrained techniques generate high temperatures, a condition required for optimization of thermal efficiency in combined cycles. High temperatures are, of course, a result of the large surface area, and hence high heat release rate per unit volume, generated by small particles, particularly at high pressures and large relative gas-particle velocities. High temperatures also suppress both clinker and tar formation, and allow ash removal under slagging (liquid) conditions (Hottel and Howard, 1971).

Compatibility with caking coals, combined cycles, and coal slurries are significant advantages; however, the entrained gasifier's lack of counter-current flow cannot be dismissed. Co-current flow reduces coal inventory; establishment of endothermic regions becomes difficult, and thus product heating value decreases. Therefore, entrained gasifiers must generate artifical recirculation zones characterized by large residence times and reducing conditions; swirl or vortex designs are necessary.

The benefits of entrained flow have prompted the development of the so-called fast fluidized bed, a fluidized bed using pulverized coal, and hence strongly approaching co-current conditions. The ash-agglomerating fast fluidized bed combines ash agglomeration and fast fluidization, thus allowing reaction with both crushed and pulverized coal (Squires, 1974).

Hottel and Howard (1971) assert that the most desirable future gasifier will use direct gasification of pulverized raw coal with steam and air at elevated pressure under slagging conditions. In the present investigation, we are primarily concerned with this type of gasifier. Unfortunately, industrial entrained flow devices do not allow for efficient separation of oxidizing and reducing regions (as suggested by early gas producers). The significant advantages of pulverized coal systems must be realized; however, new gasifier designs are necessary, particularly for high pressure operation. At present, the Koppers-Totzek, Combustion Engineering, and Texaco Partial Oxidation processes represent single-stage methods (Table III); BI-GAS is the only two-stage method (Table IV). These processes have been reviewed by Laurendeau (1975) and Bodle and Vyas (1973).

In the Koppers-Totzek system (Farnsworth et al., 1973) pulverized coal/oxygen/steam mixtures are delivered through water-cooled burners at high speeds to prevent flashback (Fig. 1). Slag is collected in a water-quench tank at the bottom section of the gasifier. Synthesis gas leaves the upper portion at around 1450°K. Steam is generated along the reactor walls. In most cases, water

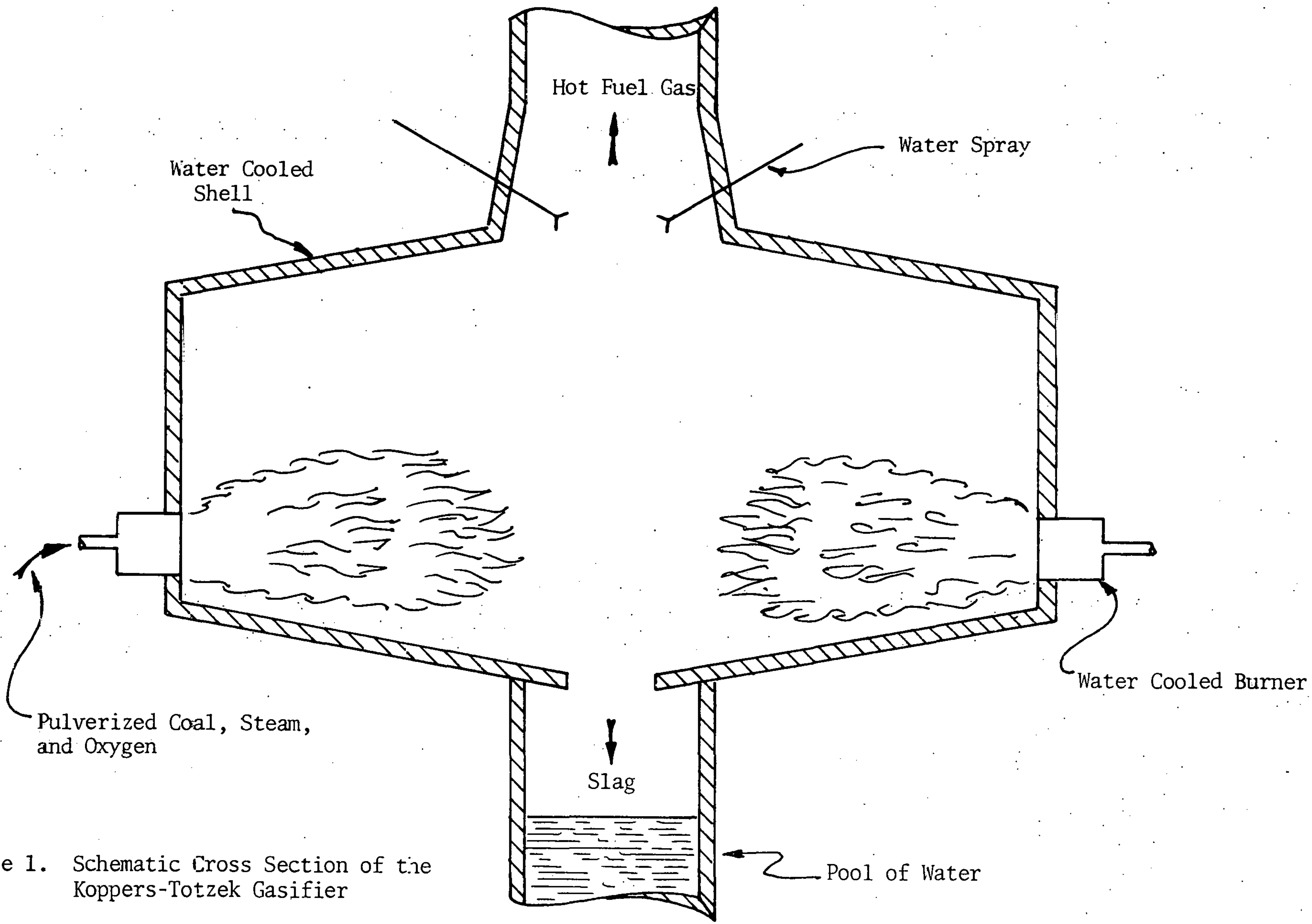

Figure 1. Schematic Cross Section of the Koppers-Totzek Gasifier

must be sprayed into the gas to lower its temperature so that ash matter will not stick to downstream tubes. Less than 0.1% CH_4 and 20% CO_2 + H_2O are present in the final product (Bodle and Vyas, 1973). The excess temperatures created by this method suggest that oxygen might be replaced by air or an air/O_2 mixture, thus forming power gas. Apparently, this relatively simple procedure has never been attempted (Squires, 1974).

In the Combustion Engineering process, the principles of Koppers-Totzek gasification have been used to produce power gas rather than synthesis gas. In this case, endothermic reactions are promoted by arranging burners into four vertical levels. Each level consists of eight individual burners, spaced equally in a circular pattern. The two lower levels burn a coal/air mixture, the upper levels use coal/steam mixtures, thus producing a methane enriched power gas. (Note that the geometric configuration of this process closely resembles a fixed-bed gas producer.)

The Texaco Partial Oxidation process (Hottel and Howard, 1971) is a linear flow (no swirl), atmospheric system. Fuel is fed into the gasifier via a coal-water slurry. On a weight basis, the H_2O/coal ratio is 0.75 and air/coal is 2.6. Most of the ash appears as slag. Ninty-five percent of the entering coal is gasified in about three seconds. Low BTU fuel is generated near $1500^{O}K$ in water-gas equilibrium.

The BI-GAS system (Hottel and Howard, 1971) uses two stages to separate the exothermic and endothermic zones. Coal and steam are fed into the upper reactor by four nozzles; heat is supplied by the hot gases emanating from the lower reactor. Entrained char, separated from the raw gas in a cyclone, feeds the vortex flow of the gasification-combustion stage, which operates on air or air/oxygen mixtures.

Pulverized Coal Gasification

Based on the obvious advantages of pulverized coal operation (Table II), this investigation seeks to perfect entrained flow gasifiers using currently available pulverized coal burners, hence minimizing development time. We seek to maximize the chemical (CO, H_2, CH_4) plus sensible heating value of the producer gas, yet minimize pollutants and carbon inefficiency. We are concerned more with power gas than synthesis gas. Hence air or air/oxygen mixtures will be used rather than pure oxygen. Compatability with combined cycle operation suggests that we consider a single stage process operating at higher pressures (1-20 atm) and temperatures (1400-1800°K).

Present entrained flow gasifiers are limited by the premixed nature of the incoming reactants. In line with fixed bed producers, we will attempt to optimize gasification conditions by separating oxidizing and reducing zones within two types of pulverized coal gasifiers: (1) the annular confined jet with secondary swirl and (2) the vortex tube with tangential entry. In this way the high temperatures generated in the combustion region will support the endothermic gasification reactions. Moreover, residence time adjustments can be used to minimize the CO_2/CO ratio of the final product.

To determine the physical and chemical mechanisms controlling gasification characteristics in entrained flow systems, we measure temperature and concentration (CO, H_2, H_2O, CO_2, CH_4, N_2) profiles (axial and radial), both within and downstream of the flame region. Pollutants (NO_x, NH_3, SO_x, H_2S) will also be considered, especially NH_3 and H_2S. System optimization will be obtained by correlation of these measurements with fundamental burner parameters such as pressure, solid/gas feed rates, swirl intensity, inlet temperature, and burner configuration. Our main purpose is to determine the feasibility of using available pulverized coal burners to produce power or synthesis gas from coal. If this phase is successful, simple models governing entrained flow systems will be developed to further interpret the experimental data.

The Confined Jet Gasifier: Preliminary Design

The double concentric confined jet gasifier is patterned after the traditional pulverized coal burner (Fig. 2). Separation of oxidation and reduction zones is attained by using coal/steam mixtures

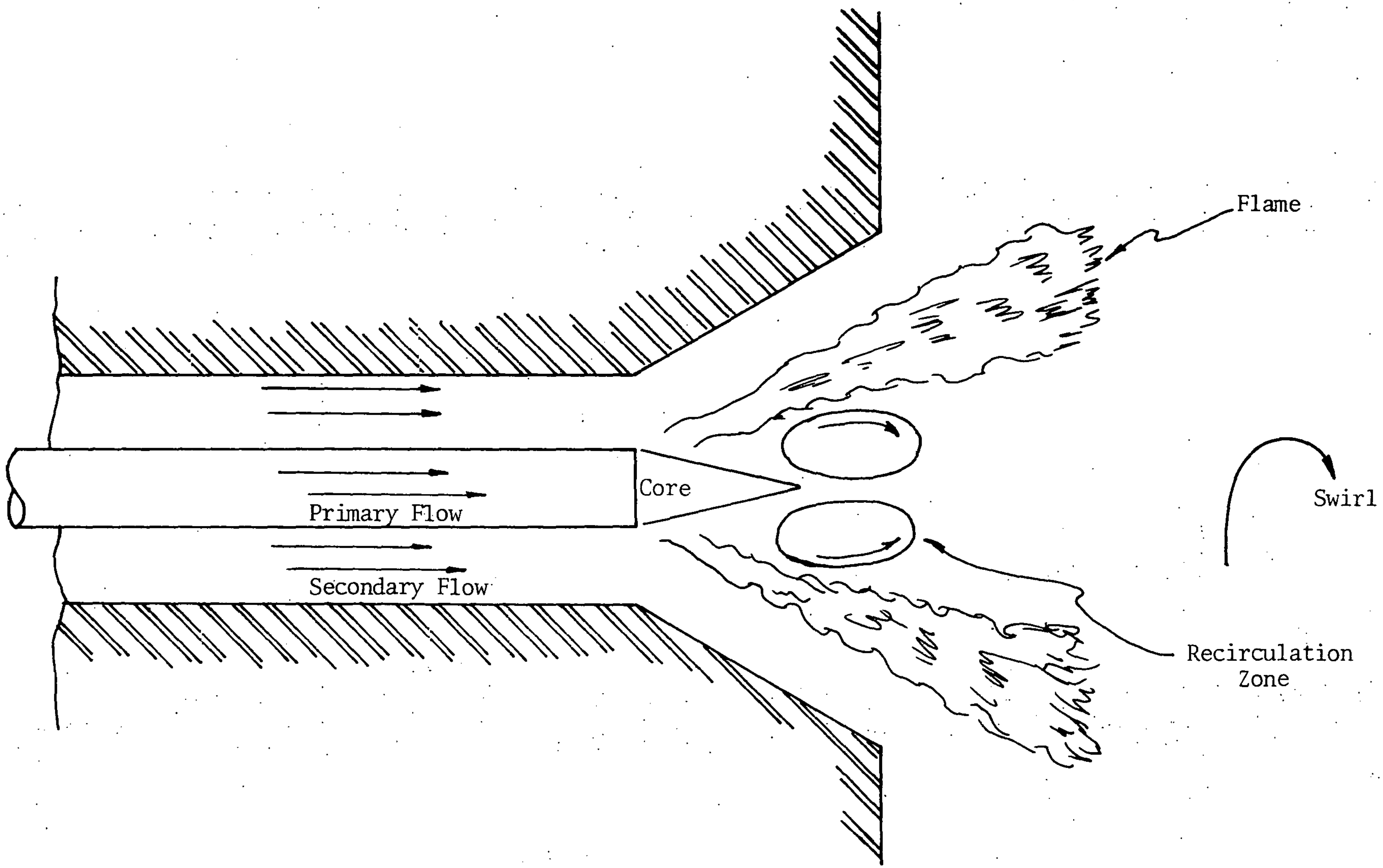

Figure 2. Concentric Jet Gasifier Schematic

in the primary flow and swirling air in the secondary flow. Fundamental experimental data applicable to confined and free jets have been obtained for cold flow (Beer and Chigier, 1972), homogeneous combustion (Beer and Chigier, 1972; Heap et al., 1973) and pulverized coal combustion (Beer and Chigier, 1969; Heap et al., 1973). These results allow preliminary design of our confined jet gasification system.

The combustion process in a pulverized coal burner is dominated by the highly turbulent flow field produced by secondary swirl. At high swirl intensities ($S > 0.6$), an adverse pressure gradient exists along the axis, and thus a torroidal vortex type of recirculation zone becomes stabilized in the flow, hence promoting air/coal entrainment and high carbon efficiency. (The dimensionless swirl number S, where $S = G_\phi / G_x R$, characterizes swirling flows. Here G_ϕ and G_x are the fluxes of angular and axial momentum, and R is the burner radius.) The size of the recirculation zone depends primarily on the swirl number and the shape of the divergent quarl at the burner exit. Increasing swirl generally lengthens the recirculation zone while divergence broadens the zone (Beer and Chigier, 1972).

Gasifier performance will depend quite strongly on the extent of turbulent mixing between the fuel rich recirculation zone and the outer air flow. Entrainment must be sufficient to promote combustion, but not enough to destroy the identity of the reduction zone. Previous work suggests that entrainment is largely controlled by the following burner parameters (Heap et al., 1973):

(1) secondary swirl
(2) primary/secondary composition ratios
(3) design and position of fuel injector
(4) primary velocity
(5) exit quarl divergence.

The first three parameters have the strongest effect, as they control flame shape during pulverized coal combustion. Our model double concentric gasifier will be designed such that all parameters can be easily varied over reasonable ranges. To reduce entrainment and thus preserve the reduction zone, Heap et al. (1973) recommended a single hole, high velocity injector, with enough swirl to stabilize the reaction zone at the injector face. However, in view of carbon efficiency requirements, the appropriate choice of burner parameters is not at all obvious.

The model burner diameter will be limited to 2-3 cm, with the primary jet size near 0.5-1.0 cm. The exit quarl half-angle will be initially designed for maximum performance ($\sim 35^{\circ}$), as suggested by Beer and Chigier (1972). Primary velocity and secondary swirl will be chosen to achieve both penetration (high primary velocity) and divergent (high swirl) flames. All fuel will be fed via the non-swirling primary jet. Secondary swirl will be controlled by a calibrated tangential entry or movable block swirl generator (Beer and Chigier, 1972). The trumpet-shaped quarl must be water-cooled, and approximately 3-6 cm in length.

The concentric jet will be confined by an externally heated tunnel gasifier. If our confined turbulent flame is to model the free jet flame characteristic of large-scale industrial equipment, we must avoid excessive wall recirculation, caused by limitations in entrainment volume. Confined jet analysis indicates that free jet conditions will be approached for Thring-Newby parameters $\theta < 0.1$, or $d/D < 0.05$-0.10, where d is the burner diameter and D the chamber diameter. Chamber length L should be somewhat greater than 3D, the distance required for flame-wall impingement (Beer and Chigier, 1972).

Although $\theta < 0.1$ allows free jet conditions, the entrained gas will probably be deficient in oxygen due to the presence of combustion products. Consequently, reaction length will increase compared to a free turbulent system. However, our confined jet environment is not unrealistic, since multiple burner operation produces similar conditions.

For turbulent jets, comparison between model and prototype is aided by the unique profiles characteristic of all turbulent jets. More generally, however, partial modeling techniques have led to the development of rather simple similarity rules. If the following parameters are maintained equal in model and prototype, reliable extrapolation is assured (Beer and Chigier, 1972):

(1) stoichiometry and initial density
(2) residence time
(3) swirl number
(4) d/D and L/D
(5) $\dot{m}_{primary}/\dot{m}_{secondary}$
(6) $d_{primary}/d_{secondary}$.

The Vortex Gasifier: Preliminary Design

The tangential entry vortex tube gasifier is patterned after the cyclone burner (Fig. 3). Separation of oxidation and reduction zones is attained by tangential flow of coal/air mixtures coupled with axial steam injection. Fundamental experimental data for both homogeneous and heterogeneous (two-phase) cyclone chambers have been obtained for cold flow (Strickland, 1973), homogeneous combustion (Schmidt, 1970; Syred and Beer, 1974) and pulverized coal combustion (Syred and Beer, 1974). These results allow preliminary design of our vortex gasification system.

The vortex gasifier possesses several advantages compared to the concentric jet system (Syred and Beer, 1974): (1) greater turbulent mixing levels, (2) ease of operation at high pressures and (3) larger particle residence times. These three factors promote high temperatures and slagging conditions, and hence good carbon and gasification efficiencies. There are disadvantages, however: (1) chamber wall temperatures are much higher, causing material and heat transfer problems; (2) slagging operation presents difficulties due to heat losses and residue disposal; (3) injection of coal/air vs coal/steam mixtures increases the opportunity for spontaneous combustion at high pressures.

Turbulent mixing and particle residence times in vortex tube systems are primarily determined by the tangential swirl velocity (Strickland, 1973). This dominant velocity profile is satisfactorily

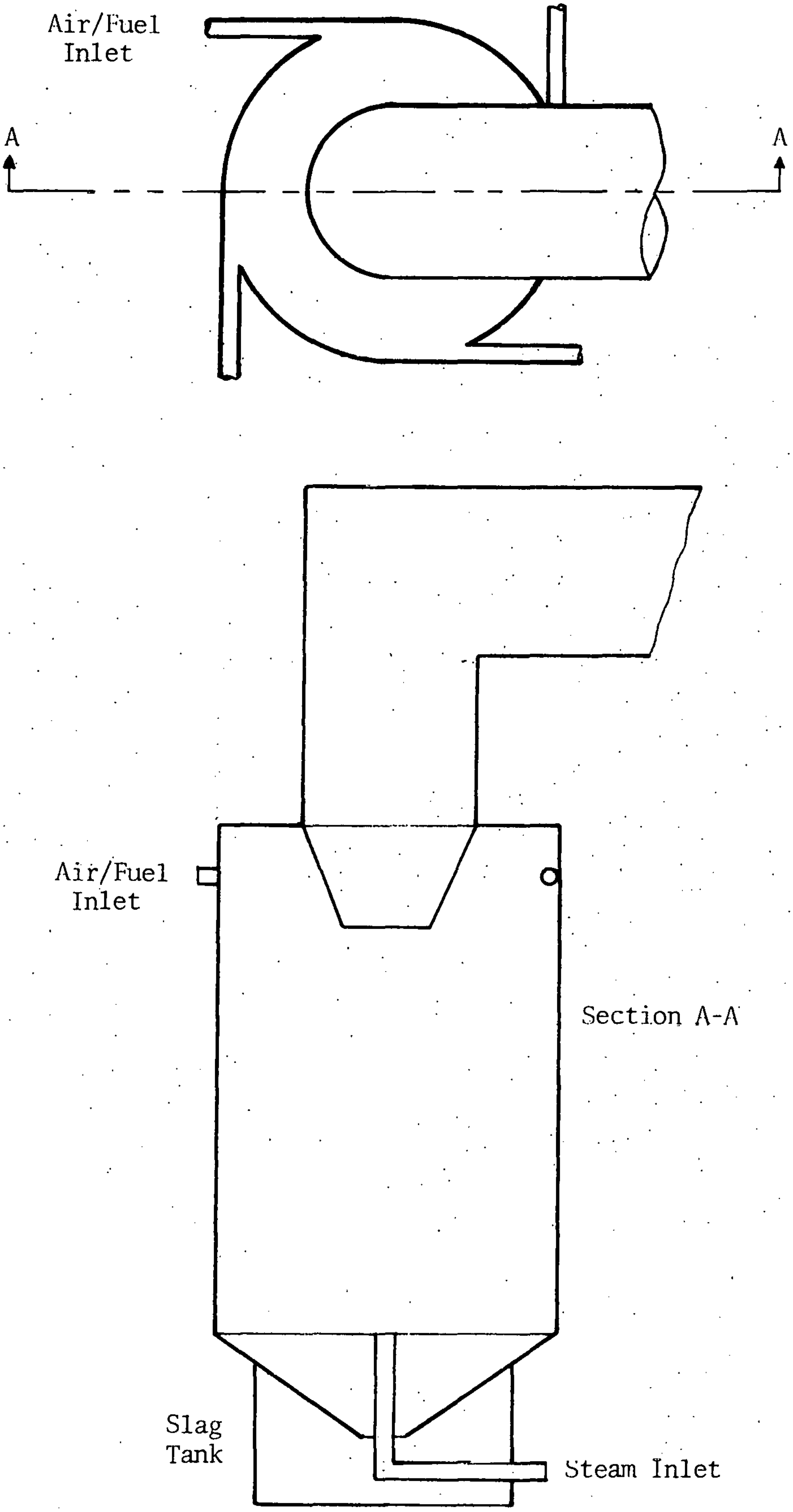

Figure 3. Vortex Gasifier Schematic

described by the Rankine model, i.e., potential or free vortex flow ($W_r \alpha r^{-1}$) near the wall, and rotational or forced vortex flow ($W \alpha r$) at the chamber core. In idealized rotary motion (Schmidt, 1970), the free vortex zone is characterized by downward axial flow (Fig. 3), while the rotational zone is characterized by upward flow. Thus, coal particles move downward via a helical path, followed by upward movement along a helical path of smaller radius. Lighter particles will of course undergo more rapid migration to the center of the chamber.

In Fig. 3, tangential air/coal injection occurs at the top of the cyclone to avoid slag buildup around the injection ports. If higher particle residence times are required, air/coal entry may be attempted at the bottom. In this case, two flow reversals occur - one at the throat annulus and another at the slag port (Schmidt, 1970). The half-angle of the throat quarl will probably determine much of the flow structure, regardless of bottom or top injection. Flow structure is also affected by the chamber endwall boundary layer (Schmidt, 1970); for example, average particle residence times are apparently increased by attaching rectangular fences radially to the throat endwall. Similarly, axial vs. radial steam injection will generate different mixing patterns, and hence affect gasification efficiency (axial injection promotes greater separation of oxidizing and reducing zones). At low steam velocities, mixing will be dominated by vortex entrainment in the core (Strickland, 1973).

Our proposed design is characterized by three important features: (1) entrained vortex flow, (2) coal/steam/air rather than coal/steam/oxygen and (3) separation of oxidation and reduction zones. The third characteristic has not been considered previously; the first two have received limited attention, most notably in the Ruhrgas process (von Fredersdorff and Elliott, 1963).

To our knowledge, the only extensive investigation of pulverized coal gasification in a <u>small-scale</u> cyclone system was conducted by Yagi and Kunii (1957). Production of both power gas and synthesis gas was correlated with carbon efficiency, and hence reduction zone temperature (1300-1650°K). By externally heating the 20 cm diameter chamber, 70-80% of the ash appeared as slag. Their preliminary results are quite encouraging, thus providing impetus for our vortex gasifier design.

The gasifier walls will be constructed from a rough castable ceramic material to maximize radiative heat exchange, and hence reactor stability. The chamber diameter will be 14-20 cm, with L/D $\approx$ 1.5-2.0 to optimize vessel residence time (Syred and Beer, 1974). The throat diameter D_e will be related to the chamber diameter D by $D_e/D \approx 0.4\text{-}0.7$ to optimize residence time and minimize chamber pressure losses (Syred and Beer, 1974). Four tangential nozzles will be used to maintain flow symmetry. Slag will be gathered in a water quench tank, as in the Koppers-Totzek gasifier.

Process Parameters and Experimental Measurements

Fundamental understanding of gasification processes requires (1) minimizing the number of independent process variables and (2) investigation of the controlling phenomena internal to the gasifier. The latter will lead to the former; more importantly however, relationships between dependent and independent process variables remain obscure (von Fredersdorff and Elliott, 1963) until internal probing clarifies the effects of chemical kinetics, fluid mechanics and heat/mass transfer. This observation is the major impetus for the stress upon internal measurements in our investigation.

Von Fredersdorff and Elliott (1963) have reviewed much of the work on suspension gasification. Given the gasifier design and coal properties, the most useful independent process variables are (1) coal throughput ($\dot{m}_c$), (2) input oxidizer/coal (OX/C) and steam/coal (H_2O/C) mass ratios, (3) reactant injection temperatures (T_{in}) and (4) chamber pressure. Presumably, dependent process variables, such as (1) carbon efficiency, (2) producer gas composition and heating value, (3) gas output rate and (4) sensible plus chemical (cold) gas efficiency, can be correlated with these independent variables. The important question to consider, however, is the following: Are we disguising more fundamental and beneficial relationships by using only these classical gasification variables? As an indication of the possible ramifications of this query, consider the following discussion.

In any gasification process, system performance will depend on the three classical chemical kinetic parameters: stoichiometry, temperature and time. Therefore, it should not be surprising that gasification performance depends primarily on OX/C, $\dot{m}_c$, H_2O/C and T_{in}, in that order. Gasification performance can be measured in many ways. The most useful performance (dependent) variables are carbon efficiency (η_c), producer gas heating value (Q), fuel ($CO + H_2$, CH_4) output rate ($\dot{V}_f$, SCF/hr) and cold gas efficiency (η_{cg}). Although many relationships exist among these parameters, usually η_c is primarily determined by OX/C, and Q by H_2O/C (von Fredersdorff and Elliott, 1963).

In studying the gasification literature, we can become quite confused by the variety of perplexing correlations between the major dependent and independent process variables (von Fredersdorff and Elliott, 1963). However, we believe that this confusion results from not considering the proper form of the dependent-independent variable relation. We propose that any dependent variable (η_c, Q, $\dot{V}_f$, η_{cg}) is related to any independent variable (OX/C, $\dot{m}_c$, H_2O, T_{in}) by the general curve depicted in Fig. 4. Therefore, depending on gasifier design, gasification performance can increase, decrease or peak with an increase in one of the independent process variables. This behavior is consistent with experimental observation. For example, producer gas heating value peaks at OX/C = 0.5, and cold gas efficiency is maximized at OX/C = 0.9 (von Fredersdorff and Elliott, 1963).

The behavior depicted in Fig. 4 must of course be explained on physical grounds. Consider first the effect of $\dot{m}_c$. Here, gasification performance (Q, $\dot{V}_f$, η_{cg}) is primarily a function of residence time. Initially, lower residence times prevent oxidation of CO and H_2; eventually, however, the residence time is too short, thus leading

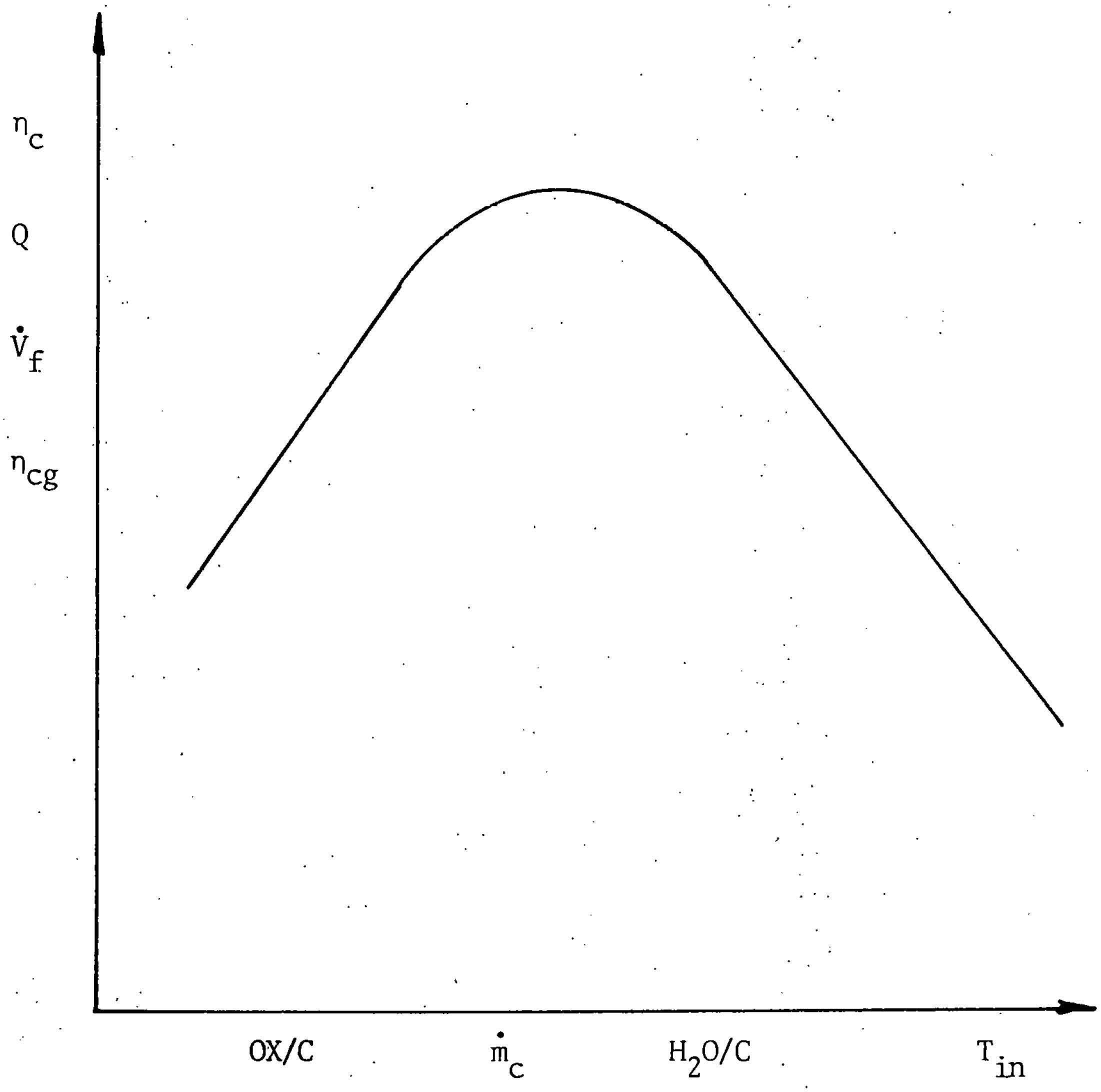

Figure 4. The Correlation Between Dependent and Independent Process Variables in Suspension Gasification

to a drop in gasifier temperature and performance. This temperature decrease also affects η_c; initially, η_c increases with $\dot{m}_c$ since higher values of $\dot{m}_c$ reduce heat losses (von Fredersdorff and Elliott, 1963).

The effect of OX/C and T_{in} are similar. Initially, performance increases due to higher temperatures. However, further increases in OX/C or T_{in} promote oxidation of CO and H_2; moreover, heat losses become more probable. For H_2O/C, initial increases are beneficial since steam promotes formation of CO and H_2. At higher H_2O/C, however, gasifier temperature and hence performance decreases.

The interpretation of Fig. 4 emphasizes again the importance of stoichiometry, residence time and temperature. Stoichiometry and residence time are primarily a function of OX/C, H_2O/C and $\dot{m}_c$. However, local temperature is a complex function of all of the above, plus T_{in} and gasifier design. Gasifier design controls turbulent mixing, and hence local stoichiometry and temperature. Thus, Fig. 4 disguises the effect of a major independent process variable, the internal temperature (and composition) field. Clarification of gasifier performance will only be achieved by internal probing for both temperature and composition. Performance parameters must be correlated with OX/C, H_2O/C and $\dot{m}_c$; more importantly, however, they must be correlated with internal temperature and composition profiles.

We must, of course, recognize that the relative importance of residence time and local stoichiometry/temperature will change depending on gasifier conditions. For example, η_c and η_{cg} are controlled by residence time at low pressures, and by heat losses at high pressures (von Fredersdorff and Elliott, 1963).