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FACTORS AFFECTING CLEANUP OF EXHAUST GASES FROM A PRESSURIZED, FLUIDIZED-BED COAL COMBUSTOR

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FLUIDIZED-BED COAL COMBUSTOR

by R. James Rollbuhler and John A. Kobak

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SUMMARY

A 200-kilowatt-thermal, pressurized, fluidized-bed (PFB) research reactor test facility was designed, constructed, and operated by the NASA Lewis Research Center. This facility was constructed as part of a NASAfunded project to assess and evaluate the effects of the PFB hot gas effluent on aerospace alloys used in the hot gas path of gas turbine engines. This facility has to have the capability of operating over a wide range of conditions in order to provide meaningful results. The Lewis PFB reactor differed from others in that it was conical so that the combustiongas velocity would be lower at the top of the bed than at the bottom. The lower gas velocity reduced the carryover of bed particles and allowed a wider range of testing conditions than would be possible in a cylindrical PFB reactor.

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Testing conditions included the type and feed rate of the coal and the sulfur sorbent, the coal-sorbent ratio, the coal - combustion air ratio, the depth of the reactor fluidizing bed, and the technique used to physically remove fly ash from the reactor effluent gases.

Testing revealed that the particulate loading matter in the effluent gases was a function not only of the reactor-bed surface gas velocity, but also of the type of coal being burnt and the time the bed was operating. At least 95 percent of the fly-ash particles in the effluent gas were removed by using a unique gas-solids separator under controlled operating conditions. Gaseous pollutants in the effluent (nitrogen and sulfur oxides) were held within the proposed Federal limits by controlling the reactor operating conditions and the type and quantity of sorbent material.

INTRODUCTION

Electric power demands in the United States are almost doubling every decade (ref. 1). Solar and hydroelectric power output is limited in the near term. Therefore a large part of the demand must be filled by fossilfuel-burning powerplants. A major problem with these powerplants is the concern for environmental pollution from their output gases, waste heat, and solid discharges (ref. 2). Existing techniques to diminish such pollution decrease the overall power output efficiency of present-day powerplant concepts.

A technique that promises to improve overall powerplant efficiency (i.e., coal pile to bus bar output) and that will be available in the near future is the fluidized-bed combustion of coal. A concept study (ref. 3) shows an efficiency increase of 10 to 15 percent for a proposed fluidized-

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bed-reactor powerplant over that for a conventional coal-burning plant with flue gas desulfurization. The same study indicates that the cost of generating an equivalent amount of electricity can be reduced in a fluidized-bed powerplant and that expensive gas scrubbers would not be needed to reduce the vent-gas pollutants to acceptable levels. Such combustion would result in a combustion gas containing less than the Federal pollution limits of 1.2 pounds of sulfur oxides per million Btu and 0.7 pound of nitrogen oxides per million Btu (ref. 4).

Present power-generating plants use the energy obtained from burning coal to turn water into high-pressure steam that is used to turn powerproducing turbine-generators. The efficiency can be improved by combining fluidized-bed combustion with a pressurized combustion furnace or vessel and using the pressurized combustion gases to drive a gas turbine directly (ref. 5). This is a form of combined-cycle power generation because steam is still being obtained from water tubes within the combustor and hot exhaust gases from the combustor are being used to power electricity-generating turbines. This scheme appears attractive enough to warrant plans being made to build a full-scale, 170-megawatt, combined-cycle power-generating plant in Ohio (ref. 6).

Fluidized-bed combustion is not a proven technique, and there are still uncertainities as to the mechanics of introducing the combustion materials into the combustor, what the ideal combustion mixture is, and how to minimize the combustion particle carryover in the flue gases. The latter problem is a concern not only in regard to atmospheric particle pollution but also in regard to particle erosion, corrosion, and deposition in a hot gas turbine through which the gases must pass. Current thinking is that the allowable particle limits must be less than the pollution limits (0.05 grain/std ft³ of gas) in order to minimize turbine-blade erosion. (0.005 to 0.040 Grain/std ft³ are said to destroy the usefulness of blades within 2500 hr (ref. 7).) In addition, the particles must be less than 10 micrometers in diameter, and the chemical composition of the particles can influence blade corrosion (ref. 8).

Because of the research and development experience in jet engine turbine materials at the NASA Lewis Research Center, a small, experimental, pressurized, fluidized-bed combustor (PFBC) was designed, built, and tested with various turbine components exposed to the flue gases. Materials tests of the various turbine components were performed and evaluated by the Materials and Structures Division at Lewis (ref. 9). The PFBC was designed and operated by the Power Generation and Storage Division, who also performed the combustion studies. This report deals with the efforts to provide clean, high-temperature PFBC exhaust gases to the turbine test components.

Over a 2 year period the PFBC operated more than 2500 hours; this included over 600 hours of turbine testing. Besides learning how to operate the PFBC in a steady-state manner, there was the question of what type and how much sorbent (limestone or dolomite) should be added to the coal to absorb the sulfur oxides. The combustion temperature was maintained at less than 2000° F to minimize the nitrogen oxide concentration in the exhaust gas and to prevent fusing of the ash. Testing was done at different bed depths and superficial gas velocities to maintain a low degree of bed material elutriation (ref. 10).

Different mechanical techniques were used to clean the fly ash and other particles from the hot flue gases. This report deals with the efforts involved in obtaining acceptable, relatively particle-free gas for flow through a hot gas turbine while at the same time meeting the Federal guidelines for acceptable atmospheric pollution.

EQUIPMENT AND INSTRUMENTATION

Combustor

A simplified cross-sectional view of the combustor used in this program is shown in figure 1. The combustor was built conical in order to decrease the superficial velocity near the top of the bed and thereby reduce particle carryover. The combustor had a design working pressure of 175 psig. Because of the limits of the existing supply air system the maximum operating pressure was 100 psia. The bed level was maintained by means of a solids-removal auger that could be located in any one of six ports on the side of the combustor. The flow rates of fuel and air into the combustor bed were controlled independently of each other: the coal flow rate by a feed screw rotation rate, and the airflow rate by a control valve. The bed temperature was controlled by adjustments in the fuel - combustion air ratio. The combustor was lined with H.P. Green low-abrade, refractory mixed with stainless-steel needles. It was insulated with Johns-Mansville Superex 2000 block insulation 2 1/2 inches thick at the top and 1 inch thick at the bottom. The main combustion air entered at the bottom of the bed through an air distribution plate that contained nine bubble caps. This air could be preheated by using exhaust-gas waste heat; most of the testing was done with 100° F air.

To start, the bed was filled to a desired level with sorbent (new or used limestone or dolomite) and preheated to 1400° F with a natural-gas burner. The fuel (coal mized with sorbent) was then injected into the bed, where it burned on contact with the hot bed material. When the bed reached operating temperature and became incandescent, the top of the bed was viewed by using a color television camera and monitor.

System

A simplified schematic of the system is presented in figure 2. The coal and sorbent stone were double screened and stored in 55-gallon drums. They were transferred to their respective hoppers through a pneumatic conveyor system. They were blended to a prearranged sorbent-coal ratio (0.06 to 0.30) and loaded into the fuel screw hopper through a lock-hopper system. The fuel was then metered and injected, with air, into the bottom of the fluidized bed.

The exhaust-gas cleanup upstream of the turbine consisted of a two-stage cyclone separator that was designed to theoretically remove 95 percent of the solid particles from the gases. After the exhaust gas passed through the turbine, it was cooled and then routed through a preheater that was used to preheat the inlet air to the combustor. The exhaust was then cleaned and sampled by another stage of cyclones. Downstream of the combustion gas-solids separator, a known portion of the combustion gas was diverted to a 1-micrometer filtering system. The quantity of solids collected in a known time was judged to give a gas solids loading that was representative of the flow through the test turbine or cascade unit. The bed pressure was controlled by a pressure control valve in the exhaust stack.

Instrumentation

The facility was heavily instrumented with thermocouple and pressure transducers, along with weight-measuring load cells on the hoppers. Turbine flowmeters were used in the water cooling systems, and venturis were used for gas flow-rate measurements. A combustion-gas analyzer was used to measure exhaust-gas emissions.

The 300 data parameters were connected electrically to a high-speed data logger and alarm system (Escort). The data parameters were scanned at 5000 words per second, one scan every 4 seconds. The Escort system used a PDP-11 minicomputer that converted all the measurements into engineering units and performed flow-rate calculations. It displayed the data on CRT displays and LED readouts. (On each data scan each parameter was compared with predetermined limits; and if any parameter went beyond its limit, a signal was given to a programmable controller that took corrective action.) The Escort system was also tied to a data collection system that was programmed to automatically take one scan of data every 30 minutes. These data were then processed by an IBM 360 computer, which did all the calculations required.

Programmable Controller

The facility was controlled by a Modicon programmable controller (PC) and closed-loop process controllers. The PC had 166 inputs consisting of panel switches; Escort alarm outputs; flow, pressure, temperature, and valve limit switches; and analog signals from the coal and sorbent hopper load cells. It has 134 outputs that controlled solenoid valves, motors, lights, annunciators, and analog outputs to the Escort data system. Using the PC permitted the facility to be fully automated and provided flexibility in making system changes. The system controls are described in more detail in reference 11.

Combustion-Gas Analysis

The combustion gases were monitored on line. Samples could be taken from within the bed, from the top of the combustor, or downstream of the turbine. The sample flowed to the analyzer located in the control room through steam jacketed and electrically heated lines that kept the sample gas temperature at approximately 325° F. Beckman instruments were used to monitor the carbon monoxide, carbon dioxide, oxygen, and total hydrocarbons; a Thermo Electron Corp. instrument to monitor sulfur oxides; and an Air Monitoring, Inc., instrument to monitor nitrogen oxides.

TESTING PROCEDURE

Materials tests were performed with either a gas turbine or cascade test section; combustion tests were performed without any test section. In the combustion tests variations were made in (1) the type and quantity of coal burnt; (2) the type and quantity of sorbent used; (3) the ratio of combustion air to coal; (4) the depth of the combustor bed; (5) the combustion pressure and temperature; and (6) the degree of heat removal from the combustor interior. A given set of test variables were held constant for a number of hours to cstablish steady-state operating conditions. For the materials tests the system had to operate at "ideal" fixed conditions for as long as possible. The longest continuous operating time was 6 days.

During the program two different coals were tested: Pittsburgh Seam No. 8 and Ohio coal. The Pittsburgh coal contained 2.0 percent sulfur, and the Ohio coal contained 2.8 percent. The sorbent used was either limestone from Grove City, Virginia, or dolomite from Ohio. The limestone was almost all calcium carbonate; the dolomite was about half calcium carbonate and half magnesium carbonate.

Before a series of tests was started, the combustor was filled to the desired level with bed material that had been left after previous tests. The removal screw was installed at the desired bed weight. The desired number of combustor internal heat exchangers was also installed.

At the start of a test series the combustor bed was heated to more than 1400° F by using a natural-gas air heater. This usually took a couple of hours, during which time the sorbent in the bed was calcined. During the heatup the exhaust gases were at about 2 atmospheres pressure and were vented through the exhaust-gas bypass cooling system to the atmosphere. After 1400° F had been attained, the coal and sorbent fuel combination was injected into the combustor. The combustion pressure was brought up to its desired value by downstream throttling, and the fuel and air flow rates were adjusted to the desired test condition. The gases were then routed through the test section and the bypass was closed.

During a test the combustion gas was sampled at half-hour interval: and analyzed for sulfur oxides, nitrogen oxides, hydrocarbons, carbon monoxide, carbon dioxide, and oxygen. At least once during a test a metered portion (about 25 percent) of the combustion gas was passed through a micropore filter for a number of hours to determine the solids content in the gas. Other solids were collected from the combustor-bed cleanup system and the combustor-side port discharg(. These samples were later analyzed for chemical composition and particle-size distribution. The chemical and size analysis technique is described in reference 12.

TEST RESULTS

In the 2-year test period 14 series of tests (designated A to N) were carried out in the PFB facility to evaluate PFB combustion parameters. During the first four series (A to D) there were no attempts to evaluate gas-solids separation. Therefore this report does not include those test data. The operating conditions of the tests made after combustion-gas cleanup studies were undertaken are listed in table I. Ten series of tests (E to N) were carried out without turbine components in the test section. In these series of tests there was freedom to make many changes in the operating conditions without being concerned with what might happen in the test section. Four other series of tests (Cl to C4) were conducted with simulated gas turbine blades of various materials and configurations placed in a cascade test section. Five additional series of tests (T3 to T7) were carried out with a small gas turbine in the PFBC test section. During the cascade and turbine tests we attempted to maintain the same operating conditions during an entire test.

The bed depth shown in table I was measured from the combustion-air injection plate up to the solids-removal port in the side of the combustor. During test series A to N the coal-air ratio introduced into the combustor was varied to see the effect. During the cascade and turbine tests, however, it was varied only in attempting to maintain a combustion-bed temperature of 1850° F. The test-bed temperature shown in table I was measured approximately 2 feet above the coal injection inlet. During test series A to N it varied over a 400 degree F span depending on what the coal flow rate was and what coal-air ratio was selected. The temperature ranges shown for the cascade and turbine tests were the extremes measured over the test time shown. During test series A to N the combustion pressure was at times set as low as 40 psia, but for most of the testing it was set at 80 psia. The test time for series A to N was from 4 to 8 hours per individual test. The test times shown for the cascade and turbine tests were cumulative times at constant conditions. The longest continuous test without shutdown was 125 hours, in test series T7.

Operation of the PFBC system required knowledge of how much of a change the operator could make in the reactor input fuel and/or air flow to achieve a desired change in the combustion temperature. There was a finite time span between when the fuel or air flow changes were made and when a change would occur in the bed temperature. Too much of a fuel or air flow change would result in large temperature excursions that would either cause tars to form or sinter the ash and coal particles.

A typical material balance for the PFB combustor burning coal with limestone is shown in table II. For this case the bed was about 70 inches deep and was filled with bed material from previous tests. The chemical analysis shows typical values.

DISCUSSION OF RESULTS

Combustion-Gas Loadings

The burning of any carbonaceous fuel results in various quantities of fuel-residue particles becoming entrained in the combustion gases. This is a particularly bad problem when the gases flow through a gas turbine. The turbine blades' useful life is a function of the quantity, particle size, particle density, and chemical composition of the solid matter passing by them. The average chemical composition of the solids, or fly ash, in the combustion gases generated in this program is presented in table II. About one-quarter of the solids was lime (calcium oxide), and about three-quarters of the solids was coal ash components (oxides of silica, aluminum, iron, magnesium, and sulfur).

The quantity of the solids in the combustion gases was a function of the type of coal being burnt, the rate at which the coal was burnt, and the quantity of combustion air injected into the reactor. The type of sulfur sorbent used or the reactor-bed composition did not significantly influence the gas particle entrainment.

In this program two coals were used: Pittsburgh Seam No. 8 (with 2.0 percent sulfur), and Ohio coal (with 2.8 percent sulfur). As shown in figure 3, the burning of Ohio coal at any flow rate resulted in a greater solids loading, or particle entrainment, in the combustion gases than did the burning of Pittsburgh coal. The quantity of ash in either coal was similar and within variation from one delivery of coal to the next. The figure 3 data also show a trend of increasing solids loading as the coal flow rate was increased.

Using either limestone or dolomite as the coal sulfur sorbent did not have any major effect on the solids loading in the combustion gases. A plot of solids loading as a function of fuel (coal plus sorbent) flow rate is presented in figure 4. These data are from tests made in a 70-inch-deep fluidized bed with Ohio coal and either limestone or dolomite sorbent. Within the accuracy of the data there was an increase in solids loading with increased fuel flow but no apparent difference in loading with the different sorbents. The ratio of sorbent to coal in the fuel was varied from 0.06 to 0.20 from one test to another, but this variation did not reveal any significant effect on solids loading.

A significant factor in the solids loading was the gas velocity through the fluidized bed. As the quantity of injected combustion air was increased, and the bed gas velocity increased, the gas solids loading value increased. This is shown in figure 5, where the solids loading is plotted as a function of the gas velocity through the fluidized-bed top surface. Figure 5 is based on tests done with Pittsburgh coal and limestone. The gas velocity was determined by assuming a uniform gas flow out of the entire to: surface area of the bed. The deeper the bed, the larger the top surface area in the conical reactor, and the lower the gas velocity. Because the fuel flow rate was also being varied when the airflow rate was changed, there is data scatter due to coal flow-rate effects on the solids loading. In spite of these variations there is a direct relationship between the gas velocity and the solids loading. Turbine test data are also shown in figure 5. They were taken at a gas velocity of approximately 1.4 to 1.6 ft/sec, and most of the solids loadings were from 1.3 to 1.7 grains/std ft³. Exxon has reported (ref. 13) a solids loading of 5.5 to 8.3 grains/std ft³.

A plot similar to figure 5 but based on tests with Ohio coal and limestone is presented as figure 6. The data show the same kind of relationship between the gas velocity and solids loading as for Pittsburgh coal, but the solids loading was higher at any given gas velocity with the Ohio coal. There is more data scatter in figure 6 than in figure 5,

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indicating that the coal flow rate rather than the gas velocity was the major influence in gas solids loading when Ohio coal was burnt. One long-duration turbine test agreed closely with the combustion test data of figure 6.

Another consequence of increased gas velocity through the fluidized bed was changes in the reactor mass balance. When the injected combustion air was increased too greatly and the calculated gas velocity became too high, the combustion gases entrained not only all the unburnable incoming fuel (coal ash and sorbent), but also a portion of the bed material. For example, in test series L the bed initially contained 216 pounds of old bed material (same composition as shown in table II). After the test series, which covered a range of different operating parameters, was completed, there was only 112 pounds of material left in the reactor bed, a loss of 104 pounds. During the test series, 211 pounds of unburnable material was part of the injected fuel. Approximately half of this unburnable material, by weight, was coal ash and the other half was calcined sorbent. The net loss of 315 pounds from the reactor can be assumed to be the solids carried out of the reactor in the combustion gases. Based on average solids loading values determined during the test series, a value of 294 pounds of combustion-gas solids was calculated, which is close to the reactor net loss of 315 pounds.

The percentage of reactor input unburnables (coal ash and calcined sorbent) that were entrained, or elutriated, in the combustion gases is plotted in figure 7 as a function of the reactor-bed surface gas velocity. Elutriated solids values greater than 100 percent indicate that not only the incoming unburnable solids, but also material from the fluidized bed, were elutriated. Data from three test series plotted in figure 7 show that quantities elutriated are affected not only by the gas velocity, but also by the coal used and the depth of the fluidized bed. The deeper the bed, the closer its upper surface was to the reactor gas-discharge port (smaller freeboard volume) and the greater the quantity of solids it would lose at a given gas velocity. Turbine tests were usually made with gas flows that gave a surface velocity of about 1.5 ft/sec in a 70-inch-deep bed. This would indicate (fig. 7) that 50 to 60 percent of the incoming unburnables were being elutriated and that, for a constant fluidized-bed height to be maintained, the other 40 to 50 percent of the incoming unburnables had to be removed by the reactor-solids removal screw. This did occur. The figure 7 data indicate that with a shallow bed, such as 46 inches deep, the surface velocity could be increased to 3 ft/sec before any bed material would be lost and that, if the freeboard volume were reduced by increasing the bed depth to 70 inches, the bed material would start elutriating when the surface gas velocity exceeded 2 ft/sec.

Data from other PFB investigations are also presented in figure 7. They show that at some critical surface gas velocity all the input unburnables to the reactor will be elutriated. This critical velocity does not agree with the data of this program because of the question of what constitutes unburnable material and how accurate the gas velocity value is. The gas velocity was based on a uniform flow of gas through the entire fluidized-bed surface. However, in viewing the incandescent bed surface on a television monitor, it could be seen that bubbling was greater in some regions of the surface than in others. Also both the Argonne (ref. 14) and the National Coal Board's Coal Utilization and Research Laboratory (NCB/CURL) (ref. 15) data were obtained in reactors whose maximum bed height was less than 50 percent of the reactor height. The important criterion about figure 7 is that, if the injected combustion airflow rate was too great, too much of the solids would be elutriated and during long tests the bed depth would be reduced.

Operating time had an indirect influence on the elutriation of solids from the reactor because of particle size. Figure 8 shows the weight percentage of reactor-bed particles smaller than a stated size as a function of that stated size. Lewis data are presented for bed particle sizes before and after a given series of tests. During the test series shown in figure 8, the particles decreased in size such that initially 65 percent were larger than 1000 micrometers and afterwards only 30 percent were larger than 1000 micrometers. During the time between the bed particle-size determinations, the gas solids loadings went from 2.69 to 6.81 to 9.12 grains/std ft³, an indication of increasing solids elutriation with decreasing particle size. The change in bed particle size and its effect on elutriation are also mentioned by Babcock & Wilcox Co. in regard to their atmospheric, fluidized-bed reactor (ref. 16). Argonne Laboratory also experienced a change in bed particle size when elutriation increased as finer bed material was used (ref. 14). Plotted along with the Lewis data in figure 8 are data from the fluidized beds operated by Pope, Evans & Robbins, Exxon, NCB/CURL, and Rivesville. Their data are for samples taken from fluidized beds after unknown test durations; however, all the data are essentially similar in size distribution.

Separation of Solids from Combustion Gases

The solid particles in the combustion gases leaving the PFB reactor are too many and too large to flow through a gas turbine. For example, in proposed PFB plant designs by Burns and Roe (ref. 17), the combustion gases going into the gas turbines are to have no more than 0.2 grain/std ft³ of 2-micrometer or smaller particles, no more than 0.003 grain/std ft³ of 2to 10-micrometer particles, and no particles larger than 10 micrometers. Early testing in the Lewis PFB facility of sample blades exposed direct. to the hot gases leaving the reactor resulted in high blade erosion and corrosion for solids loadings over 1 grain/std ft³ (ref. 9).

To remove the solids from the gases, a cyclone separator was installed in the combustion gas line of the Lewis PFB facility, between the reactor and the turbine or cascade test section. The objective was to attain the lowest possible solids content in the gases and to minimize any drop in gas temperature or pressure. Rather than a series of cyclone gas separators such as other investigators have used, the Lewis PFB facility made use of a two-in-one cyclone separator. It consisted of a heavily insulated vessel with one separator stage built inside the other, as shown in a simplified cross-sectional view in figure 9. The solids-laden gases entered the unit through a tangent side port and swirled around inside the first-scage cyclone separator. The second-stage separator was mounted inside the first stage, and there was a port in the side of the second stage through which about one-third of the gases entered the second stage. These gases flowed through a swirler in the second stage and then swirled upward to the second-stage exit. The other two-thirds of the total gas flow continued swirling up the first-stage separator to the top, where there was a swirler entrance to the second-stage separator. The gases entered the second stage and swirled downward countercurrent to the one-third gas flow coming up the second stage. The two gas flows met and were mixed in the second stage before flowing out of the separator gas exit. The solids that dropped out of the gas streams in the first and second stages were collected in separate solids-discharge hoppers. The gas temperature drop at steady-state conditions through the separator was about 150 degrees F, and the pressure drop was less than 5 psi.

During testing the as-received separator performed erratically. The efficiency at normal solids loading values would vary from 25 to 90 percent of the solids removed at identical operating conditions. A pseudo gas flow coefficient (having an unknown area factor but proportional to the gas flow rate and inversely proportional to the square root of the unit gas pressure drop and the gas density) was calculated, and the values were plotted in figure 10 as a function of the exhaust-gas solids loading. The coefficient had a high degree of variation. Some of the difficulties with the separator were uneven thermal expansion of portions of the separator, which resulted in internal gas leaks and plugging of various flow passages with gas solids.

After several efforts a final modification was made in the separator unit, as is shown in figure 9. The port between the first and second stages was closed. All the gases had to swirl up the first stage and enter through the top swirler into the second stage. The gas exit in the second stage was extended deeper into the second stage such that the gases swirled around the second stage until they were nearly to the bottom, where they reversed direction and flowed out the exit pipe. Although the pressure drop through the separator was essentially doubled, it was still a minor portion of the overall system pressure drop.

The pseudo gas flow coefficients calculated after the separator was modified are shown in figure 10. The postmodification values are much more consistent over the full range of exhaust-gas solids loading.

The efficiency of the separator in removing solids from the exhaust gases was a direct function of the quantity of solids in the gases entering the separator. This is presented in figure 11, where the percentage of solids removed from the gases is plotted against the solids loading value of the input gas. If the input gas had over 10 grains/std ft³, 5 percent or less of the solids would be removed; if the input gas had 1.5 to 2.0 grains/std ft³, 95 percent or more of the solids would be removed from the gas. As shown in figure 5 the turbine and cascade test operating conditions were usually such that the combustion gases did have about 1.5 grains/std ft³, and therefore the gases passing over the blades had solids loading values of about 0.01 to 0.10 grain/std ft³. Also presented in figure 11 are some solids-removal efficiencies reported by Exxon (ref. 18). They used two or more cyclone separators in series, with the first separator discharing the solids directly back into the PFB reactor.

The solids being removed from the gases in the Lewis facility were collected, weighed, and screened for size. About one-quarter of the total

weight came from the first stage, and three-quarters of the total weight came from the second stage. Screening of the solids indicated that 50 weight percent of the first-stage collected solids were smaller than 120 micrometers and 50 weight percent of the second-stage collected solids were 30 micrometers or smaller. These data, plus those of others who have analyzed PFB fly ash, are presented in figure 12. Argonne data (ref. 14) are similar to the Lewis data, but Exxon's primary separator seemed to collect mostly large particles (ref. 13).

The particle-size distribution of the solids remaining in the combustion gases coming out of the separator and entering the turbine or cascade test section is presented in figure 13. On the average, 50 weight percent of the particles were less than 20 micrometers, and 20 weight percent were less than 7 micrometers. Data from the National Coal Board, Argonne, and Exxon are also presented in figure 13. The NCB and Argonne data are similar to the Lewis data. Exxon used three separators in series, with the result that their first-stage-separator outlet gas had a solids loading close to that of the Lewis data, but the second- and third-stage Exxon units had solids loadings lower than the Lewis results. However, putting the combustion gases through three cyclone separators exacted a gaseous temperature drop penalty.

Nitrogen Oxide Contamination of PFB Combustion Gases

The burning of fossil fuel produces nitrogen oxides in the combustion gases. The nitrogen oxides are not a significant problem in hot gas turbine operation, but they are an atmospheric pollutant when the gases are vented out of the system downstream of the turbine.

There has been argument as to whether the concentration of nitrogen oxides in the combustion gas is a result of nitrogen in the combustion air beig oxidized or whether the nitrogen oxides are coming from the fuel. Although most PFB reports suggest that the nitrogen oxide concentration is influenced by the incoming quantity of combustion air, studies by Argonne (ref. 19) showed that when combustion air was replaced with mixtures of oxygen and argon, the resultant values of nitrogen oxides did not change. This would indicate that nitrogen oxides are formed from the coal-bound nitrogen compounds reacting with the oxygen in the combustion air, which is the controlling factor.

Exxon reports that nitrogen oxide levels are greatest at maximum combustion efficiency (ref. 13); as efficiency drops and carbon monoxide levels increase, the nitrogen oxide values drop. Argonne has reported (ref. 20, contract E49-18-1780) that the nitrogen oxide concentration is a function of the combustion pressure and goes from 200 ppm at 8 atmospheres to 1600 ppm at 1 atmosphere. Such changes in combustion pressure might be having an adverse effect on the combustion efficiency.

According to Lewis testing, the nitrogen oxide concentration is a function of the excess air going into the combustor. This is shown in figure 14, where the nitrogen oxide concentration, in terms of pounds of nitrogen oxides per million Btu's generated, is plotted against the percentage of excess air. In addition to excess air increasing the nitrogen oxide level, an increase in combustion temperature at a given excess-air value will also increase concentrations. Exxon test data fall right on top of Lewis test data. The results show that within conceivable values of excess air, the nitrogen oxide concentration will remain less than the maximum allowable pollutant concentration of 0.7 lb/million Btu.

Another way of monitoring the nitrogen oxide level is to measure the oxygen concentration in the combustion gases. This is shown for Lewis tests in figure 15. When the oxygen concentration reached 14 percent in the gases, the nitrogen oxide contamination level was about 0.6 to 0.7 lb/million Btu's with the bed temperature between 1800° and 1900° F.

Sulfur Oxide Contamination of Combustion Gases

A more serious problem of atmospheric pollution, and possible system corrosion if moisture is present, is the sulfur oxides in the combustion gases. The Federal standard of a maximum of 1.2 pounds of sulfur oxides per million Btu's is hard to meet with existing fossil fuel reactors unless expensive exhaust-gas scrubbers are used. The big advantage of fluidized-bed combustion is that various sorbents can be mixed with the coal to absorb the sulfur compounds before they can escape as gaseous compounds.

Numerous investigations have been performed with limestone as the sulfur sorbent. Exxon has made a point of showing that the limestone becomes an effective sulfur reactant after it has been calcined (i.e., oxidized). With uncalcined limestone only 20 to 40 percent of the possible sulfur emissions were captured; with calcined limestone 55 to 70 percent of the sulfur emissions were captured (ref. 21). (This is plotted in fig. 18 along with Lewis data.) Exxon states that large pores are created in the limestone particles as they heat up and calcine and the carbonate decomposes into carbon dioxide gas. As the particle structure opens up, more calcium surface is exposed for reacting with the sulfur oxide gases to form calcium sulfate (ref. 22). Argonne research (ref. 23) has found that small quantities of salt also will improve the limestone-sulfur reaction.

When starting up any Lewis PFB test series, the amount of sulfur oxides in the combustion gases differed depending on whether the fluidized bed was composed of used material or was just new calcined material. This was most evident during the long-duration, relatively constant-operating-condition testing. Figure 16 is a plot of the sulfur oxide concentration in the combustion gases as a function of time after test startup. Test C4 was begun with the bed containing 250 pounds of calcined limestone. After over 16 hours of continuous operation the combustion gas still contained only about 5 ppm of sulfur oxides. Soon afterwards the concentration started increasing until at test termination, 40 hours after startup, the sulfur oxide concentration was 150 ppm. Test T6 started with 210 pounds of calcined limestone in the reactor, and the sulfur oxide concentration did not start increasing until after 12 hours into the test. Test T5 started with the bed containing 66 pounds of limestone and 134 pounds of used bed material, and the sulfur oxide concentration in the combustion gases began increasing after 2 hours. The tests that were begun with a bed composed entirely of used material, such as listed in table II, had combustion-gas sulfur oxide concentrations that remained at approximately 300 ppm during

the entire test. All the tests shown in figure 16 were performed at about the same fuel and air flow rates. The fluidized-bed depth was 70 inches.

Once a fluidized reactor bed reached a composition state, such as described in table II, any change in the sulfur oxide concentration in the combustion gases was a function of the coal and air burning rates and the quantity of sorbent in the input fuel. The Federal limit on vented sulfur oxides is no more than 1.2 lb/million Btu's. Burning 71 pounds of Pitusburgh coal or 78 pounds of Ohio coal produced a million Btu's in the Lewis reactor. For the Pittsburgh coal, this released 1.4 pounds of sulfur that formed 2.8 pounds of sulfur oxides. Fifty-seven percent of these oxides had to be removed to meet the Federal guideline. For the Ohio coal 4.4 pounds of sulfur oxides were formed per million Btu's. And 73 percent of these oxides had to be captured before the combustion gases were vented.

These oxide limits could be met, depending on the coal burning rate and the ratio of input calcium (in the sorbent) to the input sulfur (in the coal). This is shown in figure 17 where the reduction in sulfur oxides, or conversely the percentage of total oxides escaping the system, is plotted against the Lewis coal burning rate for two series of Lewis tests in which Pittsburgh coal was burned with limestone. Three data curves are presented: (1) tests with a calcium-sulfur mole ratio of less than 1;(2) tests with a calcium-sulfur mole ratio between 1 and 2; and (3) tests with a calcium-sulfur mole ratio greater than 2. All three data curves show that with higher coal burning rates more sulfur oxide was vented. Up to 30 lb/hr of Pittsburgh coal could be burned at a calcium-sulfur ratio of less than 1 and not exceed the Federal sulfur oxides emission limits. Likewise, 45 lb/hr could be burned at a calcium-sulfur ratio of 1 to 2, and more than 70 lb/hr at a calcium-sulfur ratio greater than 2.

Theoretically at any calcium-sulfur ratio greater than 1 there should be enough calcium available to capture all the sulfur in the coal. However, in the Lewis tests it was apparent that other factors contribute to the sulfur oxide capture efficiency. This is shown in figure 18, where the percentage of reduction in sulfur oxides is plotted against the calciumsulfur mole ratio. Data for three different bed depths are plotted, and the sulfur oxide reduction increases with increased bed depth. The 99-inch-deep-bed tests were made with a bed containing half new limestone material, and this bed might not have reached an equilibrium condition. Both the 46- and 70-inch-deep-bed tests were made with old or equilibrium-composition bed material. These data suggest that longer contact time between the sorbent particles and the sulfur oxides helps in the capture efficiency. Exxon data curves are also presented in figure 18 and, although obtained in a deeper bed (120 in.) and at a higher gas velocity (7 to 9 ft/sec), the results are similar to the Lewis data.

The efficiency of capturing the sulfur oxides from Ohio coal by using a limestone sorbent is presented in figure 19 as a function of the calciumsulfur mole ratio. As in figure 18, an increase in the mole ratio resulted in a greater capture efficiency. All the data are for one bed depth. The capture efficiency was reduced wih increasing bed operating temperatures. In figure 18 the scatter in the 70-inch-deep-bed data may be due to bed temperature variations, but even at the lowest efficiencies (i.e., the highest bed temperatures), the efficiency was still 20 percentage points better with Pittsburgh coal than with Ohio coal.

The greatest efficiency in capturing or retaining the sulfur oxides was realized when using dolomite as the sorbent. Dolomite retained more sulfur per mole of calcium than did limestone. This is presented in figure 20, where the percentage of reduction in potential sulfur oxide emissions is plotted against the calcium-sulfur ratio. Pittsburgh coal was tested with dolomite in a 46-inch-deep bed; Ohio coal was tested with dolomite in a 70-inch-deep bed. The data from both series follow the same curve. Bed temperature variation did not appear to have a pronouned effect on the results. Comparison of the 46-inch-deep-bed data in figure 18 with those in figure 20 shows an almost doubling of sulfur retention efficiency with dolomite. The same is true when the Ohio coal data of figure 19 are compared with that of figure 20, even at the same sorbent-coal weight ratios. Also presented in figure 20 is a data curve obtained by Exxon (ref. 21).

Besides the percentage of reduction in sulfur oxides as a function of the calcium-sulfur mole ratio, the pounds of sorbent required per 100 pounds of coal for a given calcium-sulfur ratio is given in figures 18 to 20. Dolomite is only half calcium carbonate and limestone is almost all calcium carbonate; therefore twice as much dolomite is required at any given mole ratio to be equivalent to the limestone. To attain a 75 percent reduction in sulfur oxides from a 70-inch-deep bed burning Ohio coal with limestone would require a calcium-sulfur ratio of 1.2, or about 12 pounds of limestone per 100 pounds of coal (fig. 19). To attain the same reduction with dolomite would require only a 0.8 calcium-sulfur ratio, or about 15 pounds of dolomite per 100 pounds of coal.

From their testing, Exxon has summarized the requirements for attaining sulfur oxide emissions of less tha 1.2 lb/million Btu's. The data are presented in table III. They show no difference in the calcium-sulfur ratio needed for calcined limestone or dolomite, but they do point out the difference in the quantities of sorbent required. One explanation of why dolomite reacts as well as it does is that under the high pressures and temperatures in a PFB reactor, the magnesium compounds in the dolomite oxidize and break down. This provides a more porous particle structure for most of the calcium carbonate to calcine and to then react with the sulfur oxide gases (ref. 24).

CONCLUSIONS

As a result of 2 years of experimental work on pressurized, fluidized-bed reactors and over 4000 test data readings, the following conclusions were reached:

1. To produce high-temperature, high-pressure, clean gases for a gas turbine using a PFB system, two factors are most important: (1) to have a highly efficient solids-removal separator in the gas line between the PFB reactor and the turbine; and (2) to minimize the solids content in the combustion gases leaving the reactor. The latter is more easily attained by burning Pittsburgh coal rather than Ohio coal, by keeping the combustion airflow (i.e., gas velocity) to a minimum, and by maintaining as large a freeboard volume in the reactor as possible. In regard to the gas-solids separator the most important thing is that any unit have an efficiency based on the input gas solids loading. Too high a loading for a given unit will result in poor separation efficiency. Compact stages within stages, as compared with stages in series, resulted in minimum gas temperature drop.

2. The nitrogen oxide emissions in the exhaust gases did not prove to be a serious problem at the operating conditions tested. Although these emissions are a function of the excess oxygen in the gases and the bed temperature, the Lewis testing was done in such a way that the maximum Federal limits were not exceeded.

3. The sulfur oxide emissions in the exhaust gases depended on a number of factors: (1) whether the fluidized reactor bed was new or used material; (2) the coal burning rate and its sulfur content; (3) the type of sulfur reactant used in the fuel; (4) the fluidized-bed depth; and (5) the bed temperature. At equilibrium bed conditions the sulfur emissions were mostly a function of the coal burning rate and how much sorbent was injected with the coal. To stay within the Federal guidelines required greater quantities of calcium-containing sorbent with higher coal burning rates.

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Test series	Combustor depth, in.	Type of coal used	Type of sorbent used	Sorbent-coal ratio tested	Coal-air ratio tested	Attained test bed temperature, ^{OF}	Attained test combustor pressure, psia	Number of test changes	Number of combustor heat exchangers	Steady-state test, hr
E F G	70 99 70 46	Pitt. No. 8	Limestone	0.09 - 0.15 .0615 .0615 .0630	0.05 - 0.10 .0610 .0305 .0609	1508 - 1848 1701 - 1886 1600 - 1904 1609 - 1880	60 - 80 40 - 80 76 - 81 49 - 80	15 12 22 24	4 4 0	
I J K	46 70	Ohio 	Dolomite Dolomite Limestone	.0625 .0613 .0620	.0608 .0611 .0611	1587 - 1858 1648 - 1835 1671 - 1881	79 - 80 79 - 80 60 - 82	15 9 16		
L M N T3		Pitt. No. 8 Pitt. No. 8		.0613 .0620 .1213	.0711 .045 .0405 .0405	1678 - 1823 1661 - 1870 1845 - 1865 1885 - 1928	80 - 81 78 - 81 60 - 81 72 - 80	6 11 8 0	0	 164
T4 T5 T6		Ohio Ohio Pitt. No. 8	Dolomite Limestone		.0708 .0708 .0405	1832 - 1849 1836 - 1872 1836 - 1915	73 - 79 77 - 82 71 - 82		4 4 0	13 12 29
T7 C1 C2		Pitt. No. 8 Ohio Pitt. No. 8			.0405 .0708 .0607	1824 - 1875 $1800 - 1884$ $1769 - 1950$ 1216	78 - 82 80 - 81 79 - 81		042	401 16 30
C4		Pitt. No. 8 Pitt. No. 8	•	.12	.0405	1716 - 1920 1789 - 1887	77 - 81	•	0	100

TABLE I . - LEWIS 1978 AND 1979 PFB COMBUSTOR TESTING

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TABLE II. - TYPICAL MATERIAL BALANCE

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[Lewis PFB combustor burning coal with limestone.]

Material going into reactor	Material coming out of reactor				
Old bed material, 1b: 200 - 250 Lime, percent 60 - 70 Silica, percent 7 - 10 Sulfur compounds, percent 7 - 10 Carbon percent 1 Calcium carbonate, percent 1 Coal, 1b/hr: 40± 30 Fixed carbon, percent 49 Volatiles, percent 36	Bed material (same analysis as old bed material), lb/hr 515 Combustion gases, lb/hr: 700±300 Carbon dioxide, ppm 80 000±20 000 Carbon monoxide, ppm 80 000±20 000 Carbon monoxide, ppm 15±10 Hydrocarbons, ppm 15±10 Nygen, ppm 100 000±30 000 Nitrogen oxides, ppm 250±100 Sulfur oxides, ppm 400±200				
Ash (silica, iron oxide, and alumina), percent 10 Moisture, percent 3 Moisture, percent 2 - 3 Sulfur, percent 2 - 3 Limestone (or equivalent), lb/hr: 5±4 Calcium carbonate, percent 97 Magnesia, percent 1 Silica, percent 1 Miscellaneous, percent 1 Air, lb/hr 700±300	Fly ash, 1b/hr:3.4±2.5Calcium oxide, percent25Silica oxide, percent35Aluminum oxide, percent20Ferric oxide, percent13Magnesium oxide, percent2Sulfur oxides, percent2Titanium oxides, percent1Carbon solids, percent1				

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TABLE III. - SORBENT REQUIREMENTS^a

[To attain 1.2 lb of sulfur oxides per million Btu's in exhaust gases from PFB system operating at 1700° F.]

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Sulfur content of coal, percent	Required quantity of SO _x to be captured, percent	Required quantity of uncalcined limestone		Requir of lime	ed quantity calcined stone	Required quantity of dolomite	
		Ca/S	lb/100 lb of coal	Ca/S	lb/100 lb of coal	Ca/S	lb/100 lb of coal
2	59	1.3	8.2	0.8	5.0	• 8	1
3	73	2.1	20	1.0	9.4	1.0	20
4	79	2.8	34	1.2	15	1.2	29
5	84	3.2	51	1.3	20	1.3	40

^a Ref. 21 data.

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Figure 3. - Effect of PFB coal flow rate on solids loading in reactor exhaust gases.













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Figure 9. - Cross-sectional view of gas-solids separator for Lewis PFB system.

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Figure 10. - Effect of solids loading in reactor exhaust gases, before and after separator modification, on PFB gas-solids separator coefficient.







Figure 12. - Particle-size distribution of solids in PFB gas-solids separators.





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Figure 14. - Effect of excess combustion air on nitrogen oxide concentration in combustion gas.







Figure 16. - Effect of time into test on sulfur oxide concentration in combustion gases.



Figure 17. - Effect of coal flow rate on percentage of coal sulfur escaping PFB system.













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A 200-kilowatt-thermal, pressurized, fluidized-bed (PFB) research reactor test facility was designed, constructed, and operated by the NASA Lewis Research Center. This facility was constructed as part of a NASA-funded project to assess and evaluate the effects of the PFB hot gas effluent on aerospace alloys used in the hot gas path of gas turbine engines. This facility has to have the capability of operating over a wide range of conditions in order to provide meaningful results. The Lewis PFB reactor differed from others in that it was conical so that the combustion-gas velocity would be lower at the top of the bed than at the bottom. The lower gas velocity reduced the carryover of bed particles and allowed a wider range of testing conditions than would be possible in a cylindrical PFB reactor. Testing conditions included the type and feed rate of the coal and the sulfur sorbent, the coal-sorbent ratio, the coal - combustion air ratio, the depth of the reactor fluidizing bed, and the technique used to physically remove fly ash from the reactor effluent gases. Testing revealed that the particulate loading matter in the effluent gases was a function not only of the reactor-bed surface gas velocity, but also of the type of coal being burnt and the time the bed was operating. At least 95 percent of the fly-ash particles in the effluent gas were removed by using a unique gas-solids separator under controlled operating conditions. Gaseous pollutants in the effluent (nitrogen and sulfur oxides) were held within the proposed Federal limits by controlling the reactor operating conditions and the type and quantity of sorbent material.

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