PULVERIZED CHAR COMBUSTION IN A LABORATORY SCALE FURNACE

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INTRODUCTION

Due to the existing energy situation, many different coal gasification and liquefaction processes are in various stages of development at the present time. All these processes produce at least some charbonaceous residues or chars as by-products or perhaps coproducts. In some cases however, the char is used on site internal to the particular coal conversion scheme being employed. Other processes will have a negligible net yield of char for external consumption, and still others may produce char in considerable quantities amounting to as much as 50 percent of the raw coal feed to the plant. A study performed by Battelle Memorial Institute (1) stresses the need for markets to be found for the char produced from conceptual full-scale coal processing plants. They cite as examples yields of 4.4 million tons of char per year from a Consolidation Coal Company liquefaction plant producing 55,000 barrels of liquid petroleum per day at a depth of conversion of 50 percent, and 1.7 million tons per year from an FMC COED plant processing 3.5 million tons of coal per year. The ability to efficiently use these yields of char is of serious concern if full scale plant operation is to become a reality. The long term objective of the present continuing study is to determine the combustion characteristics of various coal chars in order to determine the suitability of the chars produced from various coal conversion processes for use in conventional combustion chambers, especially water-wall utility boiler furnaces.

The work reported here represents only the first stage of this continuing research project, and as such, serves mainly to make some general observations about the combustion of various coal chars and to identify the pertinent questions and research goals that will be examined in future critical detailed experiments.

EXPERIMENTAL APPARATUS

The experimental flames discussed in this report were generated in a vertical furnace termed the plane-flame furnace. The present version of the furnace is patterned after the one previously used by Howard and Essenhigh (2). The furnace is of square cross section, 16.5 x 16.5 cm inside dimensions, with the chamber walls constructed of insulating brick (6.35 cm thick) and encased in transite sheet insulation (.48 cm thick). The combustion chamber is approximately 2 meters long and is topped by a two-row staggered tube bank of water cooled tubes. On top of the tube bank assembly is a sheet metal mixing chamber, a pyramid in shape and 72 cm high. The pulverized fuel and primary air enter the top of the mixing chamber through a jet directed vertically upward, and the secondary air enters through a jet directly opposed to and impinging on the pulverized fuel and primary air jet. The two air streams and the pulverized fuel mix thoroughly in the mixing chamber and the resulting dust cloud then passes through the staggered tube bank into the combustion chamber below. The tube bank ensures that the dust cloud remains at ambient conditions until it actually enters into the combustion chamber proper and forces the resulting flame to stabilize in the square cross section combustion chamber. The flames produced in this manner will stabilize with a flat flame front somewhere along the extent of the combustion chamber, with the flame itself extending throughout the remaining portion of the chamber and passing out the flue connecting section to the stack. A set of observation and sampling ports is distributed along the vertical axis of the furnace on the front wall and wall thermocouples, flush with the inside of the furnace wall, are located on the back wall.

The essential feature of the plane-flame furnace is the absence of recirculation currents, i.e., backmix flow in the combustion chamber. Howard (3) verified this fact with detailed helium tracer experiments. No recirculation of hot combustion products means that the history of material gathered at a particular point in the furnace is able to be unambiguously specified.

The flames produced in the plane-flame furnace are stabilized by radiation heat transfer, with conduction and convection playing completely negligible roles. The incoming dust cloud emerges from the tube bank and the fuel particles are heated by radiation from the combustion zone lower in the chamber. The gas is primarily heated by conduction from the hot particles, and when the mixture ignites and proceeds to combust, the hot dust cloud in turn radiates back to the incoming cold cloud and the whole process continues in a selfsustaining manner.

The design and operation of the plane-flame furnace is characterized by the following summary description:

1. No recirculation of combustion gases.
2. All secondary air is added to primary air/fuel mixture before entrance to combustion chamber.
3. No supplementary fuel is fired with the experimental fuel being studied.
4. No preheat of secondary air or primary air/fuel streams.
5. Combustion environment in a utility boiler is simulated.

The last item in the summary above is not obvious since a chamber roughly half a foot on a side and seven feet tall does not seem to resemble a boiler furnace that might be forty feet on a side and perhaps a hundred or more feet high. Both chambers have residence times on the order of 1 second and peak gas temperatures around 1500 - 1600°C. Thus, the plane-flame furnace does simulate quite adequately the combustion environment in a full scale utility boiler.

PLANE-FLAME FURNACE RESULTS

After the reconstruction of the plane-flame furnace, the first fuel burned in the furnace was a Pittsburgh Seam bituminous coal, in fact, the same fuel that Howard (2, 3) studied. The flame front for the bituminous coal stabilized approximately 5 cm down from the bottom of the tube bank, as Howard had observed. This initial firing on bituminous coal served to verify that the reconstructed furnace behaved similar to Howard's original. As seen in Fig. 1, a variation on particle size was run with one fraction consisting of all particles above 88 microns, another with all particles above 44 microns and the last with all particles below 44 microns. Amazingly, no shift in flame front occured at all, but with a definite shift in wall temperature level present in the case of the smallest size fraction. It should be emphasized that most results presented in this study are displayed as plots of back wall thermocouple temperature versus distance down from the bottom of the tube bank. Suction pyrometer temperatures were obtained for some specific points for various fuels and those readings will be clearly specified as not being the usual wall thermocouple temperatures.

The next fuel burned in the plane-flame furnace was a Bureau of Mines char prepared from a high-volatile bituminous coal (Utah King A mine) in an entrained carbonizer at the Bureau's Grand Forks Station. The VM (volatile matter) content of this char is 5.1 percent. Only a 20 pound sample of this char was available, meaning that only one short run could be made. In Fig. 2, the wall temperature profiles are shown for two different total air flow rates. The general shape of the profiles is radically different from the bituminous coal profile in Fig. 1. For the higher air flow rate, the flame front was approximately 55 cm from the tube bank, while for the lower air flow rate the flame front stabilized at about 40 cm. Also, it should be noted that the wall temperatures for most of the top half of the chamber become depressed and the ones in the bottom half are elevated with increasing air flow rate. This general tendency will be seen through the presentation of the results here.

The next fuel burned was an FMC COED char with a VM content of around 2.5%, the parent coal being an Illinois No. 6 coal (Peabody No. 10). Fig. 3 shows two wall profiles for this COED char, one for all particles less than 44 microns and the other for about 45 percent (by weight) smaller than 44 microns. The shift in flame front from 55 cm back to 37 cm from the tube bank is quite significant since only a relatively minor change of particle size distribution was involved. Recall that the bituminous coal showed no flame front shift for considerably more radical changes in particle size.

In connection with the study of coal chars, other low volatile fuels, specifically anthracite coals, were burned in the plane-flame furnace. Fig. 4 shows the wall profiles for some anthracite runs. The regular grind anthracite profiles in Fig. 4 correspond to times soon after (early) and about 1 hour after (later) the preheated chamber was switched over to the anthracite coal. This particular anthracite is actually an anthracite silt with about 8.5 percent VM which is fired in utility boilers in the Hunlock Creek Station of the Luzerne Electric Division of U.G.I. Corporation. The silt is very finely ground with about 75% smaller than 44 microns and only 4 percent larger than 88 microns. The wall temperatures fell progressively with time until after about 90 minutes after switchover, the flame extinguished. At no time did the wall temperatures stabilize, thus this run was under strictly transient conditions. Also plotted in Fig. 4 is the profile for an ultra-fine grind Anthracite (5-10 Microns) for approximately the same firing conditions. This profile was stable without a doubt in this case, with the flame front located about 40 cm from the tube bank.

Two different low volatile fuels also became available for experimental purposes after the anthracites were run. These two fuels are Exxon chars produced at the Exxon Baytown Research and Development Division. One char is produced from a Wyodak coal (Wyodak Resources Development Company Mine) and the other from an Illinois coal (Monterey Coal Company No. 1 Mine). Preliminary runs with these chars (7-9 percent VM) indicated that their combustion characteristics were more like bituminous coal than the other low volatile fuels described previously. It was decided to run the previously described COED char and the two Exxon chars under controlled conditions to see how these three fuels would behave under similar experimental conditions.

The conditions chosen for the standarized runs were net heat input close to 100,000 Btu/hr and preset stepped total air flow rates. The proximate analyses, moisture contents, heating values and fuel feed rates are shown in Table 1. (see next page) The total air flow rate values chosen are given below along with their designated symbols: A, 8.76 SCFM; B, 11.55 SCFM; C, 13.76 SCFM and D, 17.79 SCFM. For this series of runs, each fuel was pulverized so that 45 percent was finer than 44 microns. Figures 5 - 7 represent the wall temperature profiles for the COED, Exxon Wyodak and Exxon Illinois chars respectively. Figure 5, the FMC COED plot, contains plots for only conditions A and B, the other conditions could not be stabilized. However, in Figure 5 the same COED type profile is seen as in Fig. 3 discussed earlier. Again the flame front positions are quite far removed from the

TABLE 1

EXPERIMENTAL CONDITIONS FOR STANDARDIZED CHAR RUNS

CHAR TYPE	FMC	EXXON WYODAK	EXXON ILLINOIS
% VM	2.97%	8.84	7.83
% FC	76.03%	69.91	68.17
% ASH	21.00%	21.25	24.00
% MOISTURE	2.23%	6.18	3.51
LHV (BTU/LB)	11,362	10,446	10,094
FEED RATE (LB/HR)	8.65	10.36	10.22
NET HEAT INPUT (BTU/HR)	98,280	108,220	103,160
STOICHIOMETRIC AIR FLOW (SCFM)	15.92	14.85	17.93

tube bank and a region of nearly linear wall temperature rise is followed by an almost isothermal zone. Samples of particulates were taken at a port 123 cm from the tube bank to obtain an estimate of the extent of particle burnoff. Table 2 has the burnoff values tabulated for all three chars.

TABLE 2

BURNOFF PERCENTAGES FOR STANDARDIZED CHAR RUNS (POSITION 123 cm)

CHAR TYPE	FMC COED	EXXON WYODAK	EXXON ILLINOIS
% BURNOFF CONDITION A	18.63	29.35	15.84
% BURNOFF CONDITION B	31.67	53.10	19.92
% BURNOFF CONDITION C	--	--	41.17
% BURNOFF CONDITION D	--	77.43	47.47

Profiles for the two Exxon chars are plotted in Figures 6 and 7. Here the wall profiles look more like bituminous coal profiles at low air flow rates and tend to become more like the COED profiles at the highest flow conditions. For both the Exxon chars, the flame never moved more than 23 cm down from the tube bank.

One addition solid sample measurement was taken for the Wyodak char under condition A. A sample was taken just above the visible flame front (3 cm from tube bank) and the resulting burnoff value obtained from this sample was 2.37%. Due to the minute fluctuations of the flame front with time, a 5 minute sample might be expected to show some burnoff slightly greater than zero.

Also several suction pyrometer temperature measurements were taken during these standardized runs. The first reading was for COED (A) and showed the flame temperature 330 degrees above the local wall temperature of approximately 1250°C. A pair of suction pyrometer readings was taken at distances of 2 cm above and below the visible flame front for the Wyodak (C) char. A difference of about 400 degrees in flame temperature was observed in moving a total distance 4 cm across the flame front.

DISCUSSION

The first point to be treated here is the use of the plane-flame furnace as a qualitative tool to order fuels according to their suitability for combustion purposes. As was mentioned earlier, the regular grind anthracite silt as burned by UGI failed to stabilize, while the COED char can be stabilized at low air flow rates. Taking account of this experimental fact, FMC and UGI arranged a full scale COED char firing of one of UGI's anthracite burning boilers (4). The parent coal of this char is a Utah high volatile - B bituminous coal from the King Mine. The tests were successful with the essential result being that when firing the COED char 8 percentage points in boiler efficiency were gained (73 percent to 81 percent) even with the minimum excess air limited to 45 percent due to the boiler control system. At lower values of excess air, the gain in efficiency might have been still greater. Thus, the plane-flame furnace has already been used as a test to order fuels according to combustion suitability.

It should be noted that the Bureau of Mines has burned the 5 percent VM Bureau of Mines char and the 2.5 percent FMC COED char in their 500 pound per hour experimental furnace. They found in the case of the COED char (6) that for a secondary air preheat temperature of 600°F, 14 percent of the furnace heat input had to be supplied by auxillary natural gas at a primary air/char preheat temperature of 250°F to obtain stavility, decreasing to no natural gas at a primary air/char temperature of 450°F. For the 5 percent Bureau of Mines char approximately 15 percent of the heat input had to be supplied by natural gas with no primary stream preheat and a 700°F secondary stream preheat for a stable combustion condition. As mentioned earlier, the plane-flame furnace uses no preheat of either input stream and no supple-fuel. Based on arguments based on radiative heat transfer scaling effects, the small ceramic-walled plane-flame furnace tends to be a better utility boiler simulation than the medium sized (12 ft x 7 ft x 5 ft) water-cooled Bureau of Mines furnace.

Secondly, the subject of particle size was mentioned several times in the previous section. The fact that the regular grind UGI anthracite (75 percent finer than 44 microns) could not be stabilized and that the ultra-fine grind anthracite did stabilize is of general importance in the area of low volatile fuel combustion. Smith and Tyler (5) have studied the reaction of a semi-anthracite with oxidizing atmospheres with four distinct size fractions being used. Only when they dealt with their 6 micron size fraction did the results indicate complete penetration of oxidant into the pore structure. This fact together with the 40 kcal/mole activation energy for the 6 micron fraction versus the 20 kcal/mole value for the 22, 49 and 78 micron size fractions clearly points to the value of around 6 microns where pure chemical control takes over from the combined chemical and pore diffusion region. This size limit for coals of similar microstructure seems to account for the difference in behavior between the two anthracite samples burned in the plane-flame furnace.

Certainly, the most important topic of discussion here is the extreme difference in the combustion behavior displayed by the two Exxon chars and bituminous coal on the one hand and the anthracites and the other chars on the other. The key point in characterizing the combustion behavior of fuels in the plane-flame furnace is the distance between the flame front and the water-cooled tube bank. The argument behind this decision is based on the thermal ignition theory of flowing systems as first proposed by Vulis (8). In this theory the position of the flame front is determined by the heat balance criterion being simultaneously applied to a set of control volumes composing the flowing mixture from the cold inlet to the chamber exit. For a given stoichiometry, set of reaction kinetics and flow conditions, the temperature profile of the entire flame can be solved for by the simultaneous solution of all the heat balances of the elemental volumes in the system. The solution of these equations must be iterative because each volume element is coupled by radiation to every other volume element in the chamber and to every element of surrounding wall area. A computer simulation of plane-flame furnace behavior has been tested in a simplified form using the char kinetics of Field (9,10) and the Hottel zone method of radiative analysis (11). Preliminary solutions do mirror the effects of increased air flow rates on lowering the earlier temperatures in the flame, displacing the flame front from the tube bank, and elevating the later temperatures in the chamber. Reactivity variations show the trend that higher reactivity fuels will stabilize closer to the tube bank. This fact should be expected because a highly reactive fuel can release more heat per unit volume at a given temperature and consequently for the same rate of heat loss to a cold heat sink. Thus, flame front position is a sound basis on which to compare fuel reactivities on a semi-quantitative basis.

The major question is why do the two Exxon chars behave differently than the other low volatile fuels. The historical approach to characterizing fuel reactivity by simple testing has been by proximate analysis, specifically volatile matter. On a dry ash free basis, the chars and anthracites have the following VM values: ultra-fine anthracite, 10.1%; U.G.I. anthracite silt, 6.9%; Bureau of Mines char, 5.9%; Exxon Wyodak char, 11.2%; Exxon Illinois char, 10.3%; and FMC COED char, 3.8%. The VM content concept obviously cannot explain the observed differences in combustion behavior since all the values tend to be very closely grouped and the lower reactivity fuel will sometimes have the higher VM content of a pair of these fuels.

For the chars, the quantity known as the fixed carbon conversion percentage is a quantity of usual interest. This percentage is defined as the absolute change in fixed carbon content from raw coal to char based on a unit weight of dry coal divided by the original absolute weight of the fixed carbon content of a unit weight of dry raw coal. The fixed carbon (FC) conversion figures for the chars are as follows: Exxon Illinois char, 45.1%; Exxon Wyodak char, 45.7%; Bureau of Mines char, 30.5%; and FMC COED char, 16.4%. As a comparison, Stacy and Walker (12) studied five FMC COED chars whose FC conversion percentages ranged from 9% to 23%. Thus, the COED char studied here can be considered typical. A pattern starts to emerge from the above values for the chars. The two Exxon chars have received quite heavy gasification treatment, the Bureau of Mines char probably an intermediate treatment and the COED char only very light treatment. The COED process is known to involve multistage pyrolysis in fluid bed reactors at temperatures from 320 °C to 870 °C and a light partial combustion (about 10% by weight combustion loss) to heat the pyrolysis stages (12). Only when the FC conversion concept is linked to the concepts of available surface area and pore size distribution does it take on any physical significance in relation to char reactivity.

Two different gas adsorption areas have come into common use to characterize particulate fuels. The area calculated from nitrogen adsorption at 77 °K is considered to represent the external area of the particle plus the areas in pores larger than about 5 Angstroms, while the area calculated from carbon dioxide adsorption at 298 °K is considered to be the best approximation to the "total surface area of coals" (13). Stacy and Walker (12) report that for the COED chars they tested, the nitrogen areas were typically lower by about 400 m^2/gm than the carbon dioxide areas. Typical values reported are 100 m^2/gm for nitrogen and 450 m^2/gm for carbon dioxide. Other chars tested in the same study (HYGAS and CO_2 Acceptor) had both adsorption areas of comparable values averaging around 400 m^2/gm . The two Exxon chars are reported to have nitrogen areas in the 300-425 m^2/gm region for typical char products (14). Thus, the COED chars have far smaller nitrogen areas than carbon dioxide areas, and the Exxon chars have nitrogen areas that are 3 to 4 times the COED nitrogen areas. At the present time carbon dioxide areas for the Exxon chars are not readily available, but this does not hinder the logical pattern described above. Based on the above area figures, the two Exxon chars would be expected to be much more accessible to the attack of reactant gases than the more highly microporous COED char.

Considering anthracite coals, Gan et. al. (15) have recently shown that for all high rank raw coals (percent carbon on daf basis greater than 83% or from HVA to anthracite) the nitrogen areas are certainly less than 10 m^2/gm with corresponding carbon dioxide areas of 200 to 450 m^2/gm . On a surface area basis, it would be reasonable to assume that the regular grind U.G.I. anthracite would not be able to stabilize in the plane-flame furnace when other low volatile fuels with much less pore volume percentages contained in micropores were having some difficulty in stabilizing. Quantitatively, anthracites have been established as having 75% or more of their open pore volumes contained in micropores, with the remaining volume split between the transitional and macropores (15). The COED are reported to only have about 30% of their open pore volume contained in micropores and apparently negligible transitional porosity (12). Therefore, adsorption surface areas and pore size distributions seem to give some solid physical background to the observed reactivity differences found when the low volatile fuels are burned in the plane-flame furnace.

In support of this viewpoint, scanning electron micrographs have been taken as part of this low volatile fuel research project. The Exxon chars, the FMC COED char and the ultra-fine grind anthracite were photographed at magnifications of up to 10,000X. The Exxon chars have voids in the particles which are of the same order of magnitude as the particle dimensions themselves and some lacy fan-like structures have been observed. The COED char shows some voidage, but the apparently solid areas on the particle exteriors are flat planar surfaces with sharp corners and definite clevage edges. The Exxon chars are seen to be almost completely amorphous structures, while the COED char appears to have a high degree of order and organization in some of regions of the particle exterior. The ultra-fine anthracite shows no surface details even down to a scale as small as 500-1000 Angstroms. The SEM results seem to agree quite well, at least in a qualitative sense, with the surface area and pore size distribution argument presented above.

Although it has been recently shown that a strong correlation exists between the reactivity of chars prepared by a thermal treatment in a nitrogen atmosphere and the rank of the parent coal (16), the Exxon Illinois char has completely different characteristics than the FMC COED char which was also prepared from a similar Illinois parent coal. This fact suggests that the precise method of activation of a coal char in a process other than simple heating in an inert atmosphere can have a dominating influence on the reactivity of the resultant char. Although, it has been demonstrated that parent coals of high rank tend to yield highly microporous chars even with activation treatments that tend to open up the pore structures of much lower rank materials (16). Therefore, eventhough preparation method can be the dominating influence on a resultant char's reactivity, high rank parent coals usually will cause severe problems if a highly reactive char is desired.

At this point, some details about the standardized char runs shown in Figs. 5-7 will be brought out to make their meaning a little clearer. As stated previously, the aim of the set of experiments is to run all three chars at equivalent values of net heat input and total air flow rate. The nominal net heat input was chosen to be 100,000 Btu/hr and the stepped air flow rates are 8.76,11.55, 13.76 and 17.79 SCFM, as stated previously. In Table 1 it can be seen that the net heat inputs ranged from 98,280 to 108,220 Btu/hr. It is regretable that this nonuniformity exists. but it does not alter the conclusions based on the results of the standardized runs. Also, the values of the theoretical air needed for complete combustion of each char are tabulated in Table 1. Here the Exxon Illinois char has the highest value of 17.9 SCFM, the Exxon Wyodak char requires 14.9 SCFM and the COED char value is 15.9 SCFM.

Flame front position again was the major difference between the Exxon chars and the remaining fuel, in this case, the COED char. Even with the highest values of total air flow used, the Exxon Illinois and Wyodak chars moved no further than 20 and 23 cm respectively from the tube bank. The COED char on the other hand, could not be stabilized any closer than 54 cm from the tube bank at the lowest air flow rate. For condition B (11.55 SCFM air flow rate) , the COED flame front moved to 75 cm from the tube bank and for condition C (13.76 SCFM air flow rate) the wall temperatures fell continually at all thermocouple locations, indicating imminent extinction. All three chars showed the effect of increasing maximum wall temperature with increased air flow rate.

Table 2 contains the burnoff data based on particulate samples collected 123 cm from the tube bank. For all chars the burnoff values increased with increasing air flow rate. Some care must be used in comparing burnoff levels of different chars. As mentioned previously, the theoretical air requirements for the chars are somewhat different. The burnoffs for the COED char versus those for the Exxon Illinois char are higher than would normally be expected until the difference in theoretical air requirements is taken into consideration. The Wyodak char has the highest burnoff values and also has the lowest theoretical air requirement. This fact makes interpretation of the burnoff data rather difficult when comparing any two chars. However, it can be noted that for condition D (17.79 SCFM air flow rate) the Exxon Wyodak char has 77.43% burnoff at about 19%excess air, while the Exxon Illinois char yields 47.47% burnoff at nearly stoichiometric conditions. Another observation that can be made is that for the Exxon chars, only when the air flow rate approaches stoichiometric conditions does the early portion of the wall temperature profile become appreciably depressed and sloped, similar to the COED type profile. The added air flows experienced in going from condition A to conditions B and through to D permit combustion to take place appreciably downstream of the tube bank, as evidenced by the flatter wall profiles recorded. Although the total air flow was increasing from condition A to condition D, the chamber residence time was decreasing due to the increase of cold inlet flow and the higher gas velocities present at the higher resultant gas temperatures. Future furnace runs with detailed burnoff profiles obtained along the axis of the furnace at many different locations would establish exactly where the burnoff is taking place, and burnoff rates based on segment residence times could be calculated. Accompanying suction pyrometer reading profiles at the same locations would allow reasonable gas temperatures to be obtained simultaneously.

CONCLUSIONS

The preliminary data and discussions presented here have demonstrated that coal chars can posses significantly different combustion characteristics, with the differences being due to parent coal rank, method of preparation, etc. It has also been shown that the plane-flame furnace has superior potential for char combustion research due to its unique design. In conclusion, if the use of coal chars in full scale combustion chambers is to become a technical reality, precisely designed critical experiments,based on the qualitative behavior reported here must be performed to put even preliminary design procedures on firm scientific foundations.

Acknowledgments

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Fig. 1 -- Wall Temperature Profiles for Bituminous Coal

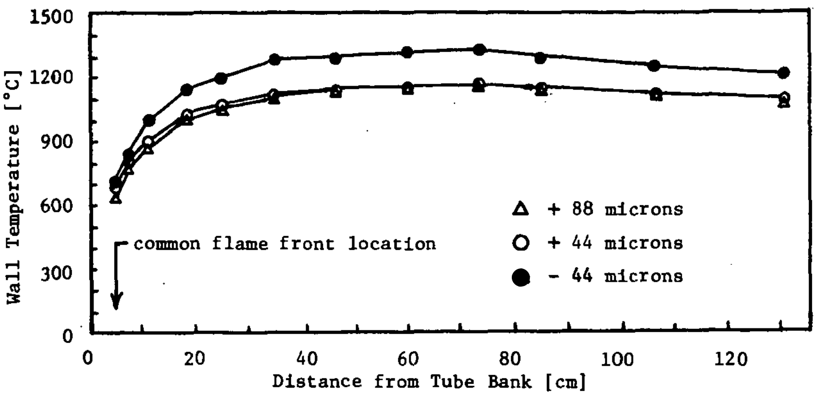

Fig. 2 -- Wall Temperature Profiles for Bureau of Mines Char

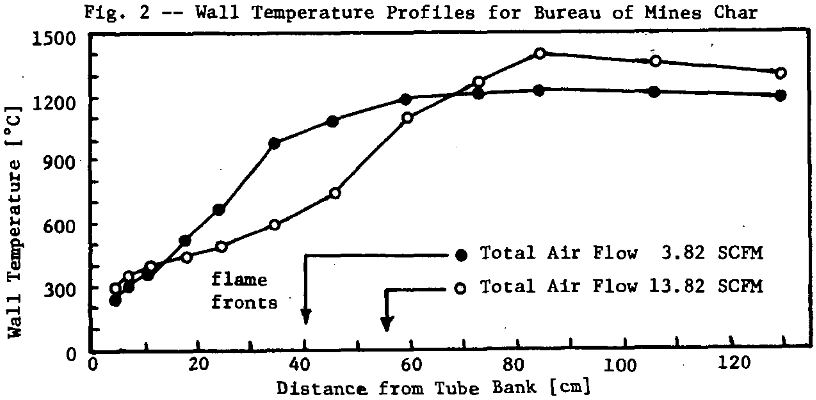

Fig. 3 -- Wall Temperature Profiles for FMC COED Char

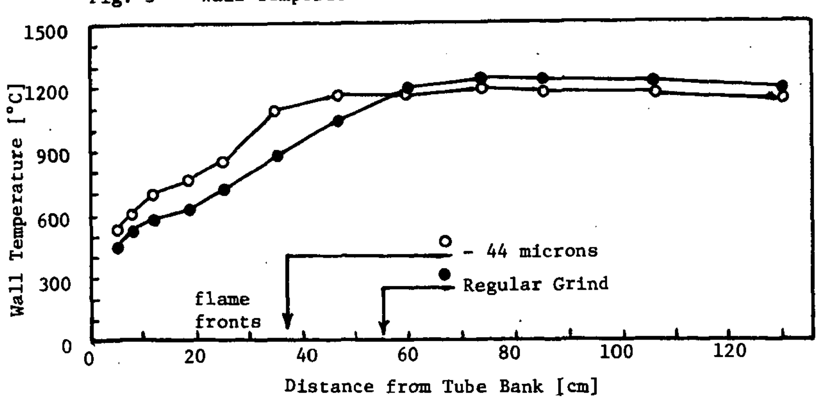

Fig. 4 -- Wall Temperature Profiles for Anthracite Coals

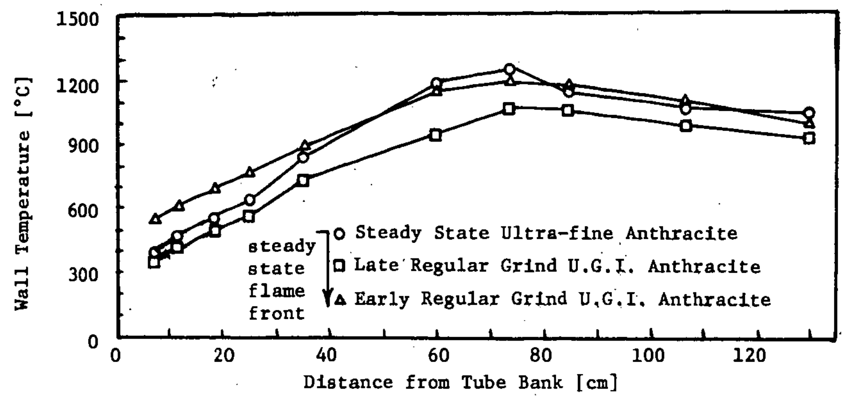

Fig. 5 -- Wall Temperature Profiles for Standardized COED Char Runs

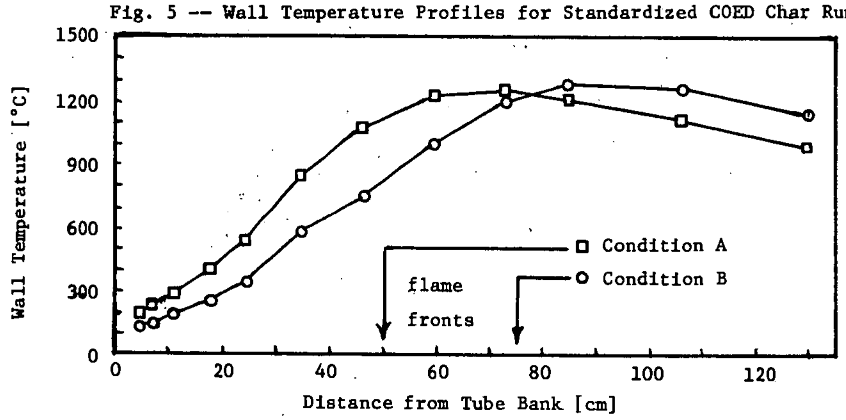

Fig. 6 -- Wall Temperature Profiles for Standardized Exxon Illinois Char Runs

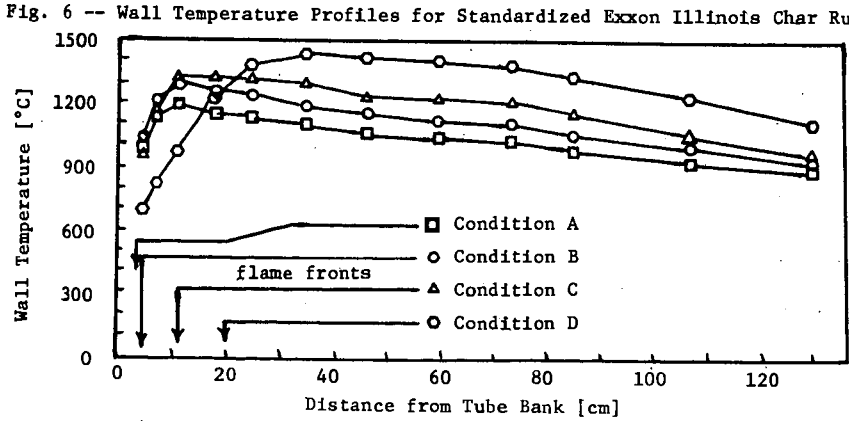

Fig. 7 -- Wall Temperature Profiles for Standardized Exxon Wyodak Char Runs