

## COAL COMBUSTION - OLD AND NEW CHALLENGES\*

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The burning of coal has been practised for centuries and misunderstood for centuries. Therefore, the challenges facing the coal combustion community are as diverse and heterogeneous as coal itself. They range from overcoming the institutional resistance and social unacceptability of coal to understanding the exact nature of an active site on a char surface. The path to expanded and acceptable coal use, and this surely has to be the ultimate objective of coal combustion research, leads through a maze of interrelated conceptions and misconceptions regarding what coal is, what it is not and how it behaves.

From a combustion viewpoint coal's legacy is most assuredly that it is not natural gas and therein lies the essence of the problem. It is an unfortunate fact for expanded coal utilization that, in nature, conventional fossil fuels occur as gases, liquids, and solids. The ease with which these fuels can be recovered, handled, processed, analyzed, characterized, and burned decreases in the order listed. This translates directly into the ease of extraction of energy in a useful form, usually as heat. Notice that solid coal is at the end of the list. Therefore, in order to understand the problems facing the coal combustion community it is not only appropriate but essential to compare and contrast aspects of the preparation and combustion of coal's chief competitors, natural gas and petroleum-based liquids.

### The Competition

Natural gas occurs in a form convenient to transport over long distances in pipelines and short distances through pipes. In general, onsite storage capacity at the user station is neither required nor desirable. A continuous supply of fuel from a pressurized pipeline is but a valve turn away. As a fuel, natural gas is relatively easy to ignite with a spark or, more commonly in industrial practice, with a pilot flame and requires a burner of relatively simple construction. Once ignited, it burns rapidly and cleanly. High combustion intensity translates into small, hence inexpensive, combustion chambers. Clean combustion depicts the absence of significant quantities of solid and gaseous pollutants in the products of combustion, which avoids the need for complicated firing strategies and expensive pollution control equipment. In short, natural gas is by any measure of evaluation a premium fossil fuel for energy production.

Unlike natural gas which is composed predominantly of the single simple molecule methane, petroleum-based liquid fuels are available in a range of molecular compositions. The size and structural features of the constituent molecules determine to a large extent the quality of the liquid as a fuel. In general, quality decreases from light, low boiling distillate to heavy, high boiling residual oils. Heavy fuel oils are, for example, too viscous to flow under normal conditions and must be heated prior to pumping. Whereas transportation through pipes and pipelines is possible, transfer lines often require heat tracing to keep the liquid flowing and prevent clogging. This is particularly the case in cold climates, since viscosity is strongly temperature dependent. Liquid fuels, which generally emanate from petroleum refineries, require on-site storage capacity to ensure that a limited continuous supply in both a valve turn and pump switch away.

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Ignition of liquid distillates is a relatively straight forward procedure and subsequent combustion is intense. Ignition is more difficult for heavy oils but is still a routine procedure with current technology. Often, startup on natural gas or distillate oil to raise the combustion chamber temperature precedes firing of heavy oils. The phenomenon of ignition is related to the rates of heat generation in, and heat loss from, the flame and less heat is lost to a hot combustion chamber than to a cold one. Of course, heat generation derives from the reaction kinetics and mechanisms, subjects of significant scientific debate. Liquid fuel burners have some degree of complexity associated with the need to atomize the liquid prior to combustion. The principal role of the atomizer is to produce liquid droplets of small size to aid ignition and increase combustion intensity. Liquids combust by first vaporizing, then the fuel vapors ignite and burn in a stable envelope around the shrinking liquid droplet. Combustion occurs essentially homogeneously in the gas phase, hence, intensity is high and combustion chambers are only marginally larger than those into which natural gas is fired. Fuel oils, particularly the heavier grades, do not burn as pollutant free as natural gas but as considerably less troublesome than coal. In general, most petroleum-based liquid fuels are categorized also as premium fossil fuels for energy generation.

### **Coal's Legacy**

By comparison, coal has unique handling and combustion problems. Because of lingering opposition to coal-slurry pipelines, transportation of coal is restricted currently to road, rail and barge. Over short distances, coal is transferred by belt conveyors or moved pneumatically. Unless the user plant is located in close proximity to the mine mouth, extensive on-site storage capacity is a necessity. Moreover, a complex coal preparation plant is required to produce fuel in a form commensurate with the mode of combustion. Most of the preparation involves regulating particle size and mineral matter, both of which affect combustion behavior. In general, coal preparation and handling systems are costly and cumbersome. In short, a continuous supply of coal requires a complicated, integrated preparation process containing a multitude of valves and pump switches.

The term coal is used generically here to describe what is, in fact, a vast array of rank and types of coal. There is general familiarity with the concept of classifying coals according to rank, for example, lignites compared to bituminous coals or anthracites. Within a given rank of coal, however, constituent molecules can vary widely in shape and composition depending to a large extent on the biological and geological history of the coal's formation. That is, coals of a given rank can vary widely in type. A consequence of these variations is that it is tenuous at best to make blanket statements about combustion behavior, there always being an exception to the rule. Therein lies one of the major problems facing the coal combustion community. It is being asked to describe, in detail, the chemical and physical transformations occurring in a material that is, at best, ill-defined. Predictive capabilities will never be fully established without an adequate model of coal constitution. With this in mind, some gross generalizations are now provided.

Coal is a relatively difficult fuel to ignite. Preceding combustion is some degree of devolatilization, a well studied but rather nebulous process whereby gases, vapors, and liquids are released from the heating coal particle. To what extent the volatiles are actually produced prior to release (as distinct from preexisting in the coal matrix) and what fraction of the total volatiles escapes the coal particle are just two of the multitude of still unanswered questions. Furthermore, the kinetics of the pyrolysis processes have eluded even the most fervent researchers for a number of good reasons. Firstly, without knowing the constitution of the starting material it is rather difficult to write anything other than the most simple chemical equations to describe the transformation. Furthermore, despite a plethora of claims and counterclaims, the temperatures of rapidly devolatilizing (and combusting) coal particles have not yet been established unequivocally.

It is often assumed, but by no means proven as being applicable to all situations, that the released volatiles ignite and burn in the gas phase thereby providing heat to ignite the residual char. The char then burns heterogeneously as a solid. It is to the detriment of coal as a fuel that solids combustion is

an order of magnitude or more slower than homogeneous gas phase combustion. That is, once ignited, coal particles are relatively slow to burn, and slowness translates directly into size and expense.

It is probably true that a little more is known about the fundamentals of char combustion than of the pyrolysis behavior. Several decades of study on relatively pure carbons has produced a clearer picture of the high temperature interactions between carbon and oxygen. What has not been established, however, is a definitive theory with predictive capability of what constitutes an active site for carbon oxidation, how this relates to structural features and the role of inorganics in determining the reactivity of a coal char.

Generally, it is not possible to start up a combustor on coal. Combustion of natural gas or light liquids usually precedes switching to coal which is fed through a burner that requires both complexity and ruggedness to handle abrasive air-entrained solids. Ignition temperatures are coal and combustor specific and cannot yet be predicted a priori. Since particle combustion times are long, large and expensive combustion chambers are required. This is the principal reason that retrofitting combustors designed for oil or natural gas to fire coal requires capacity downrating, or, more often, cannot be achieved at all. Obviously, conversion in the other direction from coal to oil or gas firing presents less difficulty and has been performed extensively in the past.

One troublesome product of pulverized coal combustion is ash, mostly micron and submicron sized fly ash. Efficient collection and disposal of this potential pollutant by baghouses and precipitators is an expensive operation. The properties of fly ash that affect collection capacity are only partially understood. Gaseous pollutants, in particular sulfur oxides and nitrogen oxides, abound from the combustion of all but premium and therefore expensive coals. Mechanisms and kinetics of sulfur and nitrogen release from the coal and their conversion to  $SO_x$  and  $NO_x$ , respectively, need to be established definitively. In short, coal in its naturally occurring form is a long way from fitting the bill as a premium fossil fuel as far as convenience and cost of energy extraction is concerned.

### **Convenience Reigns Supreme**

Needless to say, convenience has reigned supreme in the history of energy generation, at least in the U.S. Hence, although up to 80 percent of the nation's fossil fuel resources are solids (mainly coal), resources of premium gaseous and liquid fuels have persistently been depleted even in cases where solid fuels would serve as well. Take the case of steam generation in industrial boilers as an example. Installation of new capacity or reconversion of existing equipment has not been directed en masse towards coal firing. Despite persistent price hikes and intermittent interruptions in the supply of natural gas and petroleum-based liquids, convenience dictates continued reliance on these fuels.

Much of the opposition to coal is institutional in nature. It is not always sufficient to justify the use of coal combustion on economic considerations alone. The perception lingers, for example, that coal is dirty to look at, dirty to handle, and dirty to burn. While no combustion technology can overcome the first two, emerging technologies can and will help to cast the third aspersion aside.

In this context, it is not difficult to follow the argument in favor of, say, fluidized-bed combustion of coal. It alleviates many of the problems associated with conventional coal combustion technologies, in particular those related to fuel quality and pollution control. Fluidized-bed combustors can handle without difficulty materials so low in heating value that they have been discarded or disregarded as fuels in the past. This decreases the extent of expensive coal processing in order to meet fuel specifications. In addition, fluidized-bed combustors can be operated in a mode so that acid gas emissions are not of regulatory concern. The net effect of fluidized-bed combustion technology is to narrow the so-called ease-of-combustion gap between natural gas and coal, thereby making coal combustion a more acceptable and attractive proposition.

## Current Combustion Technologies

Coal-burning equipment has been available to the industrial market for close to a hundred years. Historically, combustors have been equipped with mechanical stokers. These are multi-purpose devices which feed coal on to a grate within a combustor and remove the ash residue. In addition, the grate acts as the support structure for the burning bed of coal. The concept of continuous and automatic stoker firing is attributed to James Watt who patented it in 1785. He was concerned with smoke emissions, imperfect combustion, and capacity limitations associated with hand firing or manual stoking. Concerns have not changed much in the last 200 years.

About sixty years ago, a revolutionary development occurred in coal combustion technology. To overcome problems of boiler capacity versus size restrictions associated with stokers, pulverized coal-firing technology was developed for steam locomotives. The technology was quickly adapted to stationary utility boilers and today, stoker firing is no longer a serious competitor in the utility market. Pulverized coal firing offers greater flexibility in furnace design and has been embraced in the trend toward large units. The present market for small stoker-fired units is quite poor.

Coal combustion rates are determined to a large extent by the size of the particles fed to the combustor. Pulverized coal particles are typically less than 75  $\mu\text{m}$  in diameter, that is, they are relatively small. In contrast, stokers normally feed a size range between 0.5 and 1.5 centimeters, relatively coarse particles. As a consequence, efficiency loss due to unburned fuel in a pulverized coal combustor is typically less than 1 percent compared to about 5 percent for a spreader stoker.

Pulverized coal combustors do have limitations, however. In particular, a high-grade fuel (high heating value and low mineral matter content) is generally required. Moreover, pulverized coal combustors have, in general, high levels of solid and gaseous pollutants in their products of combustion.

## Emissions from Coal Combustors

Three principal classes of emissions from coal combustors have been judged significant from an air quality standpoint: particulate matter, sulfur oxides ( $\text{SO}_x$ ) and nitrogen oxides ( $\text{NO}_x$ ). Historically, particulate matter has been the focus of attention since it can be easily seen and therefore is easy to label as a public nuisance. It is comprised of fly ash and unburned carbon. Particulate matter release is controlled to some extent by either electrostatic precipitation or fabric filtration. Oxides of sulfur and nitrogen are of concern because they are precursors to acid rain and can cause ill health effects in humans. In addition, oxides of nitrogen participate in atmospheric reactions that produce photochemical smog.

Acid rain has emerged as one of the top environmental issues of the 1980s. According to the experts, it is caused by the atmospheric conversion of gaseous  $\text{SO}_x$  and  $\text{NO}_x$  into sulfuric and nitric acids. In the eastern United States where most attention is focused, man-made emissions of acid gases are said to dwarf natural emissions. In the case of  $\text{SO}_x$ , emissions are said to be dominated by a relative handful of sources. Coal-fired power plants are being labeled as the prime culprits, with the 20 largest plants producing about a quarter of all  $\text{SO}_x$  emissions in the region.  $\text{NO}_x$  emissions, on the other hand, are attributed to both power plants, not necessarily those firing coal, and motor vehicles.

All fossil fuels are being blamed for releasing  $\text{CO}_2$  into the atmosphere, a fact which will get no dispute from combustion scientists. What will stir debate, however, is whether or not such release of  $\text{CO}_2$  is causing global warming. This debate and the associated research effort in the next decade is likely to dwarf the current preoccupation with the causes and effects of acid rain.

The sulfur content of different coals varies widely. It is unfortunate but a fact of life that many of the high-sulfur coals are located in the eastern half of the U.S. The east is also the region that is under scrutiny for the effects of acid rain. Furthermore, it is the region of the country that is most intensive in the use of fossil fuels. All these factors work against utilization of local, inexpensive, and abundant coal and drive users toward transported (and often imported) natural gas or petroleum-derived liquids.

Sulfur atoms are either chemically bound to the organic fraction of coal or found in mineral constituents such as pyrite. Coal preparation plants can reduce mineral matter content, hence, inorganic sulfur content. They can do little, however, to reduce organic sulfur content. There is some renewal of interest in the concept of superclean coal and coal water slurry fuels produced either by chemical and/or biological treatment methods. The concept here is to add cost to the fuel thereby producing a so-called premium fuel and reduce the burden on the combustor and down stream cleanup devices. This is likely to continue to receive attention, particularly for gas turbine and diesel engine applications.

As coal burns, most (often 90 percent or more) of the sulfur is converted into  $\text{SO}_x$ . Historically, gaseous  $\text{SO}_x$  emissions have been "controlled" by either dispersion or reduction. Dispersion of pollutants through tall stacks is an old-slight-of-hand which is no longer acceptable. Reduction by flue-gas desulfurization (FGD) is a band-aid solution, according to some, often creating as many problems as it solves. There is a general movement towards alternative ways to combat acid rain, and it is not difficult to understand why. According to industry figures, maintenance costs for FGD systems are 20 times higher than those for the rest of the power plant, and this has led some to call FGDs the most costly and least reliable pieces of equipment in the utility industry. Operational problems aside, there is the challenge of what to do with the sludge produced. In fact, disposal of all residues associated with coal combustion systems is likely to be the focus of significant regulatory attention.

While  $\text{SO}_x$  emissions can be related directly to fuel sulfur content,  $\text{NO}_x$  emissions originate from two different sources. During combustion,  $\text{NO}_x$  forms in high-temperature regions in and around the flame zone. Oxidation of both atmospheric nitrogen (thermal  $\text{NO}_x$ ) and fuel-bound nitrogen (fuel  $\text{NO}_x$ ) occurs. The rate of thermal  $\text{NO}_x$  formation is influenced by temperature and the oxygen concentration. Reducing flame temperature and excess air, therefore, can help control thermal  $\text{NO}_x$  formation. This is achieved in practice by performing combustion in stages, appropriately called staged combustion, and recirculating some of the products of combustion. Fuel  $\text{NO}_x$  formation is related to fuel nitrogen concentration and is most significantly influenced by the oxygen concentration in the combustion zone. In an oxygen-deficient atmosphere, fuel nitrogen can be converted to  $\text{N}_2$  rather than  $\text{NO}_x$ , particularly the nitrogen contained in the volatiles. Details of reactions kinetics and mechanisms are still sketchy.

### **Fluidized Bed Combustion**

Debate is currently raging over the issue of whether or not the effects of acid rain are serious enough to warrant costly emissions control programs. When the ayes prevail, the question will be reduced to what control strategy should be implemented. While the current preferred technology may still be FGD or some form of limestone injection, a promising alternative technology for new construction and retrofit applications is fluidized-bed combustion.

Burning coal in fluidized-beds usually involves injecting the fuel into a bubbling or circulating bed of calcined limestone or dolomite and ash. Combustion occurs both within and above the bed and the gaseous pollutants produced can be captured by the bed material. Until recently, this technology was aglow with praise, but it has now been relegated by some for "supported implementation" until economic and political circumstances change. What this means, in essence, is a time delay until petroleum supplies are again interrupted or prices hiked.

The principle of fluidization is conceptually simple and mathematically complex. To state the obvious, solid particles can be moved by a fast-flowing stream of air or other gas and it is this simple principle upon which fluidized beds operate. Consider a chamber open at the top with a wire mesh at the bottom. On the mesh is a bed of inert granular particles. From beneath, a stream of air is being blown up through the mesh. If the air flow rate is low, air will percolate between particles in the bed without disturbing them to any great extent. If the air velocity is increased, a point will be reached when the particles move upward in unison and the bed will occupy an expanded, yet still well-defined, space. The particles, in essence, are now being supported by the air rather than the grate. At still higher air velocities, the particles begin to mix violently within the envelope of the bed and adopt the appearance, and some of the properties, of a boiling liquid. The bed is then said to be fluidized in a conventional bubbling mode.

Were the air velocity to be increased even further, granular particles of bed material would become entrained in the air and be blown or carried from the bed. The bed would then lose its integrity. In practice, granular particles come in a range of sizes intimately mixed. Fluidizing velocity depends, among other things, on particle size. Hence, operating limits for a boiling bed are set, on the one hand, by the minimum velocity needed to keep the largest particles fluidized and, on the other hand, by the maximum velocity that can be tolerated before an excessive number of small particles are blown from the bed.

This highlights not only the principles of fluidized bed operation but also those of fixed bed (stokers) and entrained-flow (pulverized) combustors. In fixed beds, coal particles are large and the grate-supported bed is relatively undisturbed by the percolation of combustion air through it. In entrained-flow combustors, small pulverized coal particles are carried at high velocities into a turbulent fireball in a combustion chamber. In fluidized-bed combustors, medium-sized coal particles are injected into a turbulent bed, and burn within and above the bed in intimate contact with the bed material.

Fluidized-bed combustion affords some advantages over other modes. When operated at atmospheric pressure, however, and this is the current state of the art, boilers employing fluidized beds have no particular thermodynamic advantages over conventional boilers. Their attractiveness and potential for success lie in their ability to meet air pollution standards at a lower cost than conventional systems, particularly those equipped with FGDs. The development of pressurized fluidized bed combustion of coal, on the other hand, is being driven by improved cycle efficiencies, particularly in connection with gas turbines.

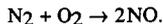
Low emissions levels in fluidized beds are inextricably linked to low bed temperatures, 700 to 900 C, compared to flame temperatures of 1600 to 1800 C in pulverized coal boilers. The lower temperatures are a prerequisite for efficient sulfur capture by the bed material, normally calcined limestone, by the endothermic reactions:



Calcium oxide particles are porous with sulfation occurring to some extent over much of the surface. SO<sub>2</sub> is the predominant sulfur oxide in combustion gases, usually accounting for 80% or more of the sulfur-containing species, hence, equation 1 predominates. Calcium sulfate does form preferentially on outer particle surfaces, however, eventually creating an impervious skin which stops further reaction. The relationship between sorbent composition, combustor operating conditions and sorbent performance is the subject of considerable enquiry at the moment.

A second major pollutant that suffers retarded production at temperatures less than 900 C is thermal NO<sub>x</sub>. Thermal NO<sub>x</sub> is a term coined to specify those nitrogen oxides, mainly NO, formed in

combustion systems in which the original fuel contained no chemically bonded nitrogen atoms. Generally, it is thought that the NO is formed from N<sub>2</sub> in air by the following overall equation:



According to equilibrium considerations, significant NO would be found only at temperatures considerably in excess of 900 C. However, the route to NO formation involves several radical species including O and N atoms and OH radicals. Consequently, equilibrium calculations provide only a conservative estimate of actual NO concentrations as a function of temperature. Nevertheless, it is generally recognized that thermal NO<sub>x</sub> formation is not of great consequence at the operating temperatures of fluidized-bed combustors.

Although not directly temperature-related, reactions destroying NO<sub>x</sub>, which does manage to form in fluidized-bed combustors, are quantitatively significant. Most of the NO<sub>x</sub> formed originates from nitrogen contained in the fuel. Once formed, it can be converted to N<sub>2</sub> by heterogeneous reactions with bed char and sorbent. There is some evidence that these reactions are catalyzed by CO, one of the major gaseous products of carbon combustion. In summary, the consequence of having a limestone- and char-containing bed at low temperatures is the elimination of regulatory concern about NO<sub>x</sub> and SO<sub>x</sub> emissions. This is the feature that may control the destiny of atmospheric fluidized-bed combustors.

Some combustion experts try to sell the concept of fluidized beds on the basis of high heat transfer rates to in-bed water tubes. From an economic point of view, this is offset to a large extent by high tube erosion rates, and some now question the wisdom of inserting tubes into the bed at all.

An additional advantage of low-temperature coal combustion is suppression of slag and deposit formation. Both are attributed to the presence of various inorganic constituents in coal which undergo physical and/or chemical transformations at high temperatures. The presence of slag and deposits causes capacity downrating of the boiler and operational problems. The behavior of inorganics during combustion may be the single most important operational issue in the coal combustion community at the present time.

The current trend in medium size units is circulating fluidized beds. In circulating fluidized-beds, gas velocities are much higher and a consequence is that coal and bed material are elutriated from the bed and recirculated through the combustor back to the bed via a hot primary cyclone. Combustion of the coal occurs throughout its passage, hence, combustion efficiencies are high and SO<sub>2</sub> removal is more efficient. Fluidized bed technology is undergoing the inevitable growing pains associated with commercialization, particularly with respect to tube failures and bed agglomeration. Materials handling problems have also surfaced in units firing low grade fuels. The problems are not insurmountable and should be overcome with further research and development. Fluidized bed combustion will then take its place alongside existing technologies, and will then face the common enemy, a lack of knowledge of the fuel being burned in the combustor.

### The Challenge

Coal combustion research is and should be driven by necessity. A thorough knowledge of how coal burns is predicated on the desire to mitigate the environmental impact of coal combustion, overcome operational difficulties and develop predictive capabilities. This will only be achieved once a detailed understanding of coal constitution has been obtained. Then the task of describing, in detail, the transformations that occur when coal is burned can be launched in earnest. Applications may have changed over the decades but the challenge has not. It is, indeed, a formidable one.