

COAL CARBONIZATION: CURRENT AND FUTURE APPLICATIONS

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INTRODUCTION

Carbonization is defined as "The destructive distillation of organic substances in the absence of air, accompanied by the production of carbon and liquid and gaseous products". The coke produced by carbonization of coal is used in the iron and steel industry and as a domestic smokeless fuel.

Only a limited range of coals produces acceptable metallurgical cokes. These coals are in the bituminous rank range but not all bituminous coals are caking coals. Prime coking coals are expensive and not always available nationally. It is predicted that remaining indigenous coals available for coke making are poorer in coking quality (1). In addition, coke ovens need to be rebuilt and, in most parts of the world, profits from the steel and iron business are insufficient to provide the necessary capital. In future years, we will have to extend our technology even further and find new blends or raw materials and optimum operating conditions in order to reduce costs. This will only be possible with a better understanding of the fundamental aspects of the coking process, i.e. properties of coals and their functions in the coal-to-coke conversion. This understanding is also necessary because of the gradually increasing stringency of requirements made on coke by modern industrial practices, i.e. maximum output with maximum efficiency.

In this paper, the authors attempt a preview of the future of coal carbonization from the viewpoint of past and current experience.

COAL PETROLOGY

Greatest use of coal petrology has been made in the area of coke making. Most steel companies have adopted petrographic relationships, based on coal rank (reflectance) and type (maceral composition), for use them for preliminary evaluation of coals and for coal-blend design. Coal macerals have their own carbonization behaviour, some of them, e.g. part of semifusinite and most of the micrinite and fusinite, remain relatively inert whilst others, e.g. vitrinite, resinite and part of semifusinite, gasify, soften and become porous, hardening ultimately into coke (2, 3). However, maceral behaviour is more complex than originally thought and as Kaegi *et al.* (4) showed, they behave quite differently depending on what other macerals are in contact with them. Quick *et al.* (5) proposed the use of a tandem system of fluorescence and reflectance microscopy for the recognition of reactive vitrinite and inertinite to provide a better assessment of carbonization behaviour.

RHEOLOGICAL PROPERTIES OF COAL

Several tests are available to evaluate the suitability of coals for coke production (6). Coking coals become plastic when heated; consequently, a wide variety of tests have been introduced to measure the plastic and swelling properties of coal (7). The most common are the free-swelling index test to determine the agglomerating and swelling characteristics of heated coal, the Gieseler plastometer for assessing the plastic properties and the Audibert-Arnu dilatometer to record volume change as a function of time.

CHEMISTRY OF COAL CARBONIZATION

A significant development in understanding carbonization processes was made with the discovery of mesophase in the plastic stage of carbonization leading to graphitizable carbons, as observed by optical microscopy (8). The development of spherical mesophase particles (Figure 1) from an isotropic mass and their progressive growth and coalescence eventually to form anisotropic structures is well established for pitch-like precursors (9-11). Essentially, during the carbonization process, dehydrogenative polymerization of aromatic molecules occurs, with a consequential increase in average molecular weight (12). The final coke structure (Figure 2) is related to the properties of mesophase at the time of solidification and these, in turn, are dominantly dependent upon the chemical properties of the parent material. Coke quality improvements are dictated by the quality of the parent feedstock which predetermines the optical texture of the resultant coke.

In contrast to pitch-like materials, carbonization of coal produces mesophase in the form of very distorted spheres which do not show observable coalescence because of their high viscosity. These differences in behaviour could be attributed to inhibiting effects of elements such as nitrogen, oxygen and sulphur and to the influence of particulate inert matter in the coal (13).

COMMERCIAL CARBONIZATION

The coals selected for blending for blast furnace coke give the highest coke physical strength with acceptable chemistry, acceptable reactivity and at a competitive cost. The coke must contract sufficiently for easy removal from the oven and swelling pressures must be acceptable.

Strength and reactivity

The chemical composition of coke can be controlled within desirable limits by coal selection and control of carbonizing and quenching conditions (14). The desirable physical properties of coke are less defined in that furnace operation is still not completely understood. The physical quality of coke is usually defined in terms of its size and by empirical parameters representative of its resistance to breakage and abrasion. The classical strength tests (shatter test or drum tests, Micum, Irsid) are size dependent and it is difficult to derive relationships linking the results of one test to another (15).

Conventional strength indices are being supplemented by new coke heating procedures which provide a better simulation of the behaviour of coke in the blast furnace. These include various "solution loss" reaction tests and hot strength tests (16). A major objective of modern coke making is to improve the properties of coke at high temperatures, especially its strength after reaction with CO_2 and its reactivity (17, 18). Table 1 shows the BSC coke specifications for large blast furnaces. Many tests have been developed to measure coke reactivity and a large and diffuse amount of literature is available on this subject. A synoptic report describing the high temperature properties of coke has been published by the Commission of the European Communities (19).

Coking pressure

It is established that some coals can damage coke oven walls because of either excessive pressure developed during carbonization or insufficient coke contraction at the end of the coking process. This problem has lately become a matter of importance due to coal preheating and the widespread acceptance of tall batteries which increase the bulk density of the coal charge, so affecting coking pressure, contraction and coke oven life. The Spanish National Coal Institute (INCAR) has

developed (20) a laboratory method of predicting expansion and contraction behaviour during carbonization, based on a modification of the early Koppers laboratory test.

Prediction of coking properties

Much effort has been made to predict coke strength based mainly on measurements of vitrinite reflectance and maceral analysis. These predictive techniques have two common premises: i) for any given coal there is an optimum blend of reactives and inerts which will give the best coke, ii) percentages representing this optimum blend or ratio vary with rank. Different approaches have been followed in terms of analysis and treatment of coke conditions and of the particular index of coke strength determined (21-23). The Japanese assessed the suitability of a single coal to be used in a blend from maximum fluidity values and Ro (24). CRM Belgium developed a new prediction method (25) applicable only to the conventional wet charge process which relates coke strength indices to inert content of coal, caking ability of the reactive components and the maximum Gieseler fluidity of the blend.

Predictions for volume charge and carbonization pressures based on petrographic data have also been formulated.

CURRENT COKING TECHNIQUES

As mentioned before, producers of metallurgical coke are experiencing a shortage of prime coking coals and suitable high volatile coals at a time when more stringent coke quality demands are being made as iron is being produced in larger, faster-driven blast furnaces. The conventional wet charging process of making good quality coke is very dependent on coal quality. To solve this problem several techniques have been tested.

Additions of caking substances

The aim of this process is to produce metallurgical coke of good quality by adding reformed non-coking coal or petroleum heavy residue to coal. This technical approach does not produce any particular problems in the coking process itself and its success depends upon making inexpensive binders. Major binder manufacturing processes that have already been industrialised or are in the pilot plant stage are in Japan. Coal tar pitch has been found to be a satisfactory substitute for medium volatile coking coal in blends in coke oven tests conducted in West Germany (26).

Selective crushing

It is known that selective crushing of many types of coal and blends can produce an improvement of coke quality (27) but commercial development has been obstructed by the non-availability of suitable equipment to screen wet fine coal with large throughput. Nippon Steel Corporation has developed a large wet coal screening equipment (110 t wet coal/hour) which is efficient and commercially available (28). The process minimizes the production of very fine coal particles and, at the same time, selectively crushes inertinite-rich particles into smaller sizes than the vitrinite-rich particles. In this way, the inertinite-rich particles are evenly distributed in the coking blend, the bulk density is increased and coke quality is improved (29).

Partial briquetting

Briquette blend coking process (BBCP) is most commonly used in Japan as a technique to use low grade coals (30). Non-caking and poorly caking coals can replace coking coal giving coke with a suitable strength. However, blending conditions must still be optimized and briquette manufacturing costs reduced.

Stamp charging

This technology is applied in China, Czechoslovakia, Poland, East Germany, Rumania, France and West Germany. In India a plant is being built and the USSR recently decided to implement it. This process has been used for many years in Europe to produce coke from high volatile coals and, at the present time it has been successfully incorporated into the new high ovens (31). The coke oven feed is compacted in a stamping machine and is charged into the ovens horizontally. Stamping increases bulk density of the charge and improves coke strength, especially resistance to abrasion.

Formed coke

Formed coke processes (32-33) consist of manufacturing briquettes from non-caking or weakly caking coals which, after suitable carbonization, can be fed directly into blast furnaces. This avoids the construction and maintenance of expensive coke oven batteries. The briquetting may be hot or cold and the binder is usually a pitch. Several tests have been carried out in the blast furnaces with formed coke replacing conventional coke but, at the present time, the process does not have significant application.

Preheating

Preheating is a technique which substantially improves coke quality from low rank coals. It also produces an increase in oven and battery throughput (as much as 50% for the lower rank coals). Other advantages derived from the elimination of the water from the charge include more uniform heating and a reduction in the thermal shock. Applications of preheating also provide more effective smokeless charging, elimination of routine mechanical levelling and a decrease of pollution during the pushing operation. These effects of preheating may be due to modifications of the plastic properties or possibly to a permeable plastic layer which allows tar and other pyrolysis products to react with the coal (34).

The disadvantages of the preheating process are related to the difficulties of handling fine hot coal and the increased carryover of fines. Coking pressure during carbonization must also be closely controlled and, in general, coking batteries charged with preheated coals have more technical problems (35).

DEVELOPMENT OF MODERN COKE MAKING TECHNOLOGY

In recent years, the productivity of coke ovens has been increased by using higher coking temperatures, silica bricks with higher thermal conductivity and coking chambers of larger volume. In past decades, the use of wide ovens was controversial because the correlation between coking time and chamber width was not clear. In this respect, Bergbau-Forschung (BBF) carried out a large scale testing operation (36) in oven chambers of 450, 600 and 750 mm width in the range of temperature of 1300-1350°C. It was assumed that the increase in residence time, at these temperatures, was not significant and in any case it could be compensated for by increasing bulk density. As a result, an increase in productivity could be achieved by building wider oven chambers. From this research, such plants were built in Germany and operational experiences indicated that the operation of coke ovens with an effective volume of 80m³ and more, and a coke production of about 50 t per charge is possible without too much risk.

However the controversy about coke ovens width still continues. Recently, Centre de Recherches Metallurgiques (Belgium) carried out coking tests in its experimental oven in order to determine

the effect of chamber width on coke quality and on coke oven productivity (37). It was concluded that it is mainly dependent upon coal blends used. With wider chambers, good coking blends showed an improvement in fissuring resistance, significant improvements in the abrasion resistance indices and an increase in the mean diameter of coke pieces. With poorly coking blends, the mechanical strength indices and coke piece mean diameter decreased.

FUTURE OF COAL CARBONIZATION

From the above discussions it is evident that, despite considerable effort, there is as yet no practical alternative to traditional coke making. Three different approaches to coal carbonization may be adopted in the future: i) improvement of coke production by introducing modifications, ii) the building of a new coking system, iii) the development of new carbon materials with different specifications and new functions.

Improvement in existing technology includes the construction of new heating systems which reduce NOx formation, the automation of the coke oven battery and the use of computers to control the working schedule. Automation could be extended to the characterization of coking coal blends in order to save time, i.e. techniques such as neutron activation elemental analysis are being assessed (38).

The second approach, to build a new coking system, was taken into consideration by the German coking industry, in collaboration with Ruhrkohle AG and BBF GmbH. They developed a new concept in coke making technology on the basis of ecological problems, cost-effective production of coke with high productivity and specific output. To achieve this, some constraints must be considered (39): maintenance of the present coke structure, establishment of independence from the raw materials, minimization of the energy consumption, reasonable solutions to problems of environmental protection, improvement of process control and monitoring and production of by-products adapted to prevailing conditions. Noting these constraints, a research programme called "Jumbo Coking Reactor" was developed (39).

From the studies carried out, the productivity would be higher than those currently operating. The operation of only 55 reactors will be of major benefit to the environment. The length of the sealing surfaces that require daily cleaning is 60% less than those for the large capacity ovens that are now in service. The number of opening processes is reduced to only 110, against 1200 in one of the new German plants (Prosper). From the economical standpoint, this plant will reduce the total coke costs by between 10 and 20%.

ALTERNATIVES TO COAL CARBONIZATION

One of the functions of coke in the blast furnace is to provide heat in the hearth of the furnace for melting the reduced ore. In the mid-fifties oil was injected at the furnace tuyeres to provide heat to the hearth and reduce the coke rate. When oil prices rose, a number of systems were developed to inject pulverised coal. At least 16 countries have blast furnace coal injection systems. This has been one of the most rapidly expanding iron-making developments world-wide (Figure 3) (40).

Ironmakers are now considering coal injection to reduce the coke rate of blast furnaces. Reasons given are: coal is cheaper than purchased coke; for those ironmaking plants which are considering building or rebuilding coke ovens, the capital cost of coal injection equipment is less than coke oven plant; a wide range of coal types can be injected including non-coking coals. Coal

injection results in smoother blast furnace operation with less hanging and slipping than the comparable all-coke operations.

Injected coal can reduce the fuel and reductant roles of coke in the blast furnace but the provision by coke of a solid permeable bed from stockline to the hearth cannot be replaced by coal injection. As coke rate reduces, the pressure drop across the furnace increases and it is thought that the gas and liquid flow will decrease and, consequently, the iron production rate.

SPANISH APPROACH

The availability of coking coals in Spain is limited. ENSIDESA, the most important Spanish iron and steel industry has a coking coal consumption of about 3×10^6 t/year. 70-80% is foreign coal and only 20-30% Spanish coal. The wide diversity of Spanish coal mines and the need to import coking coal from all over the world causes major difficulties in coal blending.

ENSIDESA is using a very sophisticated blend of 15 to 20 different coals and owns a large yard for coal blending and homogenizing with a handling capacity of 8.5 Mt/year and a stockpiling facility of 800,000 tons. To predict the coke strength and to optimize the blending from an economical stand-point, a mathematical model has been developed and successfully applied by ENSIDESA (41). The Spanish Research Council (CSIC) has two Institutes for coal research. One of them, the National Coal Institute (INCAR) has an Experimental Coking Test Plant (Figure 4). The battery contains four ovens. The oven dimensions are 2.8 m in height, 6.5 m in length and 300, 350, 400 and 450 mm, respectively, in width. The heating system is independent for each and their capacity is 4-6 tons. INCAR co-operates very closely with ENSIDESA.

Some of the research carried out by INCAR is related to coking pressure, preheating, coal weathering effects on coke quality, coal blending and the use of different additives to improve coke quality. INCAR has developed and patented (20) a laboratory method for predicting expansion and contraction behaviour during carbonization, based on the Koppers laboratory test. The Koppers-INCAR laboratory test was successfully applied to the problem of Spanish coals which have dangerous swelling behaviour during coking. The contraction behaviour data obtained are related to the chemical properties of coal, i.e. volatile matter, mean vitrinite reflectance and softening point. A 2 t/h preheating pilot plant, Precarbon process, owned by ENSIDESA and built on line with the INCAR Coking Test Plant, is being used for preheating research. It has been used to study effects of adding a high volatile coal and semi-anthracite to an industrial base blend which had given good quality coke by the conventional wet coking process (42). Studies were also carried out on the effect of petroleum coke addition to coals with high and low volatile matter content and the effect on the productivity and the pressure exerted during the coking process. The improvement of coke quality from coals with a high volatile matter content by using different additives is also being studied. It was observed that aluminium additions up to 3.0 wt% of aluminium considerably improved coke strength (43).

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Figure 1. Mesophase development from isotropic pitch material.

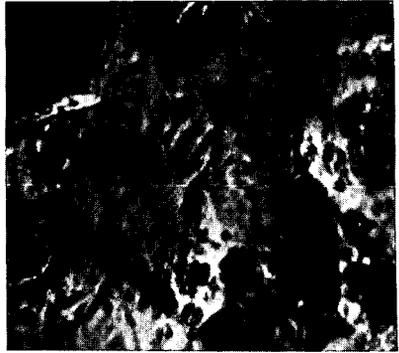


Figure 2. Optical micrograph of polished surface of a metallurgical coke.

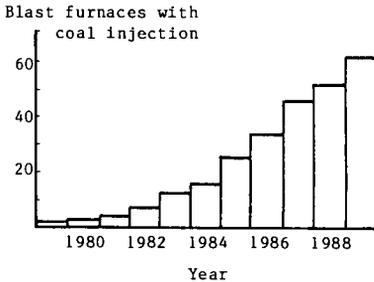


Figure 3. Development of coal injection.

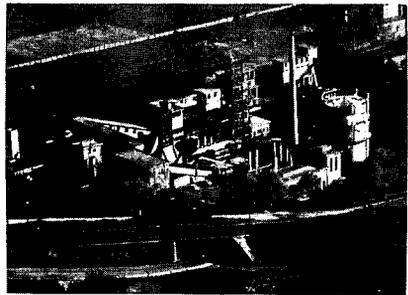


Figure 4. INCAR Coking Test Plant (Oviedo, Spain).

Table 1. The new BSC specification for large blast furnaces (18).

| | |
|---|----------|
| Mean size | 50 mm |
| Size range | 30-75 mm |
| Japanese DI ₁₅ ⁹⁰ | ≥82 |
| Japanese reactivity | ≤30% |
| Japanese post-reaction strength | ≥53% |
| Ash, d.b. | ≤10% |
| Sulphur, d.b. | ≤1.1% |
| M40 micum index | ≥80 |
| M10 micum index | ≤7 |
| Extended-micum slope | ≤0.8 |