CENTRE for RESEARCH and TECHNOLOGY HELLAS
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OVERVIEW of LOW TEMPERATURE CARBONISATION

Present Status – Properties, Yields and Utilisation of LTC chars – Survey of Various Methods – Pre-treatment Conditions & Effects – Advantages, Economic & Technological Development

Report prepared for

ISFTA – May 2004
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Survey of Various Methods – Pre-treatment Conditions & Effects
– Advantages, Economic & Technological Development

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WorkPackage # 4: Promotion of Low-Temperature Carbonisation (LTC) Technology

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EXECUTIVE SUMMARY

Low temperature carbonisation is essentially a process for producing fuels-solid, liquid, and gaseous – from coal and other carbonaceous material, such as biomass and organic wastes. The products from the various low temperature processes, however, vary in quantities and qualities depending on the coal used and the nature of the treatments to which it has been exposed. The differences can be of great significance in the selection of a process for a given application, and careful study is necessary before a choice can be made.

The process descriptions have shown that many methods for the low-temperature carbonisation of coal are available for use, and from them chars with a wide variety of characteristics can be obtained. One desirable characteristic of char is its smokeless quality. This property, together with suitable form characteristics, has won for the lump char produced by certain processes important places in the domestic fuel market in Great Britain and in other European countries. The second product, also of major importance, has been the large volume of tar. During 1920s there was a common expectation that the values of these products, especially the chemicals to be derived from the tar, plus possible economies in the carbonisation processes as compared to high-temperature coke ovens, would lead to attractive profits from a low temperature plant. Engineers and inventors have proposed a great amount of work and innumerable designs. Of all the processes worked on during that period, only one-the present Disco process - survived.

There are five necessary conditions to be met if a low-temperature carbonisation process is to be rendered technically and economically feasible:
1) Universal applicability to different types of high-volatile coals
2) Production of a firm, abrasion –resistant product
3) Maintenance of desirable temperature conditions through the system to produce a uniform tar
4) Maintenance of an output which is economically attractive, a condition dependent on rabidity of heat transfer and involving oven design and the thickness of the coal bed
5) Smooth, trouble-free oven operation

The two processes in regular commercial use are the Disco process of the Consolidation Coal company, this plant being located near Pittsburgh, Pennsylvania, and the stoker carbonisation process, operated by Shawinigan Chemicals Ltd. in Canada. In addition to these plants, the Parry process has been operated experimentally on lignite by the Texas Power and Light Company at the power installation for the Aluminum Company of America at Rockdale, Texas. The Lurgi-Spulgas process also has operated in North Dakota for some time on the local lignite. A process for producing a metallurgical formed coke from Wyoming subbituminous coal was announced, and production of a low-temperature char is one of the first steps. This plant is operated by Food Machinery and Chemical Company and the United States Steel Corporation at Kemmerer, Wyoming. In Europe, Japan, and other parts of the world, the Kruoo-Lurgi, Lurgi-spulgas and Rochdale gas re-circulation appear to be most widely in use, but work is proceeding at a number of locations on smaller-scale units of other carbonisation methods.

Although many processes have been realised, they have been operated on different coals, in different ways, and for widely different objectives. Little correlation between them is available. Each process has often functioned on pretty much an experimental scale, without reaching the more critical stage of successful commercial practice. The engineer entering the low-temperature field finds it almost a virgin one, which encompasses first the selection of a process for a given need, then the design and engineering with the identification and definition of the problems to be faced, and finally plant operation and meeting the unexpected developments. For these reasons commercial evaluations of processes are still very difficult.
Low temperature chars differ markedly from high-temperature cokes in appearance and properties. Generally, pieces of low temperature chars are black to dull grey, with an open, porous structure. They are softer, more friable, and weigh less than high-temperature coke. Both the apparent and true densities are lower. The volatile content of the char usually lies in the range from about 20% down to 9 or 10%. When heated, a char gives off much hydrogen, and the gas burns with an almost non-luminous flame. Because of the high volatile content and the open structure, char ignites readily and burns freely. These qualities make a lump char an ideal fuel for the open domestic grade. The quality of a charcoal can be represented by a great many other properties including charcoal yield, fixed-carbon content, moisture content, calorific value, elemental composition, hardness (abrasion resistance), compressive strength, bulk and true densities, surface area, porosity and pore volume distribution, electrical resistivity, and reactivity. The high reactivity of chars necessitates their thorough quenching or cooling unless they are to be used very shortly. In general, the properties of chars are greatly affected by the coals from which they have been produced and by the conditions to which they have been exposed during carbonisation. Commercial utilisation of low temperature chars includes domestic or industrial fuel, blast furnace fuel, reducing agent, electrodes and other uses.

Product distribution and how it is influenced by coal pyrolysis conditions have been studied. Lignite and bituminous coal are exhibiting quite different behaviour. Lignite products up to 700K are mainly water. Subsequently the evolution of CO2 and tar starts. At slightly higher temperatures CO, hydrocarbons and H2 are observed due to defunctionalisation. The hydrocarbons mainly consist of methane and ethylene. Based on char and gaseous products analyses, it is clear that the lignite char becomes relatively enriched in C, since even at the highest temperature only about 20% of the original carbon is lost. In the case of bituminous coal product distribution is dominated by tar. H2O is formed at relatively low temperature, below 700K. Carbon content profile is fundamentally different than that of lignite and it continuously decreases over the whole temperature range. Up to 50% of the originally present carbon is volatilised eventually. The hydrogen profile shows an analogous pattern that is in good agreement with the predominant evolution of tar. Apart from coal type,
pyrolysis conditions that affect product distribution are the temperature, retention time, heating rate, pressure and particle size distribution.

Economic and market prospects of LTC technologies are quite promising, since such fuel pre-treatment technology fits into the European, North American and global market developments, fulfills technical and cost efficiency demands, and meets International Protocols and Agreements. Among key-points of LTC development is the market tendency for expansion of small- and medium-sized power generation plants. Traditional plants can easily be retrofitted by LTC add-on module, whereas old plants can cost efficiently upgraded and still meet the new environmental norms and regulations.

Potential LTC project development offers economical benefits, such as decreased total cost of the end-product Clean Coal, environmental control cost savings, increased profits due to efficiency improvement, and reduced raw material cost, since domestic renewable sources and agricultural wastes utilisation is feasible.

Major technical and scientific benefits of LTC technologies employment include (a) preventive environmental protection, (b) improved boiler efficiency, (c) total cost savings that may exceed 10%, (d) reuse of industrial residuals, since flexible multi fuel operation is possible, (e) enhancement of fuel diversity and security, (f) elimination of environmental and health care risks, and (g) reduced boiler corrosion, resulting in maintenance cost savings. One of the major LTC advantages is fuel availability, since it offers potential utilisation of alternative biomass and food industry wastes, whose disposal may present today high cost and technical deficiency, such as (a) waste vegetable oils (co-combustion), (b) non usable – non recyclable residuals from bio-diesel production, (c) biomass residues, such as straw pellets and husk, (d) energy crops, such as short rotation wood and sawdust, (e) biomass waste, i.e. demolition wood, and (f) meat meals from animal by-product processing industry. Among known limitations that LTC technologies present are (a) reduced efficiency in treatment of feed streams with very high moisture content and (b) removal of all heavy metals from low grade fuels.
Development of R&D projects and future implementations may be beneficial for most countries, including EU Members, Accession Countries, East European and Asian Countries. In general, adoption of LTC technologies offers significant advantages, among which are:

(1) It is a preventive option that compared to “end-of-pipe” solutions results in:
   a. Guaranteed and competitive long-term clean energy supply
   b. Providing compatibility between development of open – liberalised energy market and the objectives of the sustainable environmental protection
   c. Providing overall cost savings for sustainable clean energy production

(2) Improved overall safety and recycle-reuse of industrial material streams

(3) Flexible feed choice application from regionally available feed supply by converting trash (low grade fuels) into cash (clean energy)

(4) Improving overall cost efficiency for sustainable green energy production

(5) Supporting schemes for reduction of greenhouse gases as per Kyoto Protocol and European Commission commitments
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OVERVIEW of LOW TEMPERATURE CARBONISATION

Present Status – Properties, Yields and Utilisation of LTC chars – Survey of Various Methods – Pre-treatment Conditions & Effects – Advantages, Economic & Technological Development

Part I.
Status of low temperature carbonisation
Properties, Yields and Utilisation of low temperature chars

Report prepared for

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1. Status of low temperature carbonisation

Low temperature carbonisation is essentially a process for producing fuels—solid, liquid, and gaseous—from coal. The products from the various low temperature processes, however, vary in quantities and qualities depending on the coal used and the nature of the treatments to which it has been exposed. The differences can be of great significance in the selection of a process for a given application, and careful study is necessary before a choice can be made. Although the recovery of chemical products from the tars and oils has frequently been proposed as a major objective, no installation in which they can be separated on a commercial scale appears in prospect.

As a producer of fuels, a low-temperature process is required to turn out products competing with similar types of fuels also available in liberal supply. Of the low-temperature products, the char represents the largest proportion of the original coal, and on that account its movement must be assured. Its cost is determined not only by the cost of the original coal but also by the cost of carbonisation. To offset this additional processing cost, the coal must either be cheaper than the competing fuels, or the char should be possess an advantage, such as better size, shape, analysis, reactivity, or other property. One example of the use of a cheaper coal would be the conversion of a low-grade coal, such as lignite or brown coal, into an industrial or domestic fuel, which is competitive with higher grades of coal. This condition will assume increasing importance as depletion of better grades of coal continues. The manufacture of formed cokes for metallurgical use, in which low-temperature carbonisation is a primary step, should eventually be able to utilize such lower grade coals, and consequently is of potential importance to the steel industry.

One desirable characteristic of char is its smokeless quality. This property, together with suitable form characteristics, has won for the lump char produced by certain processes important places in the domestic fuel market in Great Britain and in other European countries. Attention, moreover, is being given in Great Britain to conserving domestic coals of carbonisable quality for use in coking. In many localities where smoke-control regulations are in effect, smokeless fuels are being demanded, and consumers may find they do not have...
appliances including devices for gas ignition. For these consumers, reactive low-temperature chars, possibly in the form of briquettes prepared from non-coking coals—which are under development, should be useful. It is expected that they will receive a premium price.

The situation of the tar is similar. Because the tar will compete with fuel oils, it should at least be comparable in characteristics with them and possess no property which will render it less desirable. The presence of excessive quantities of solid char in some tars has materially reduced their value. In Germany and Japan during World War II little petroleum was available, and the military demand for aviation fuels was acute and partially satisfied by motor fuels derived from the low-temperature tars produced in those countries. With the now adequate availability of petroleum products, the demand for the tar fractions for these uses has disappeared. Where petroleum in normally available, low-temperature tars have had difficulty in competing successfully.

The volume of the gas is small, and its heating value is often low because of dilution with combustion products. On a therm basis, the yield is about half that produced in high temperature processes, and most of the gas is frequently consumed internally in the plant where it is made. The volume left has been insufficient to make the process of interest as a producer for city distribution.

The process descriptions have shown that many methods for the low-temperature carbonisation of coal are available for use, and from them chars with a wide variety of characteristics can be obtained. The second product, also of major importance, has been the large volume of tar. It was long a common expectation that the values of these products, especially the chemicals to be derived from the tar, plus possible economies in the carbonisation processes as compared to high-temperature coke ovens, would lead to attractive profits from a low temperature plant. These opinions have been largely responsible for the great amount of work and for the development of the innumerable designs, which have been proposed by engineers and inventors. Not only was much money spent in the legitimate development of new processes, but low-temperature carbonisation also became a fertile field for cranks and promoters, especially during the 1920’s. In 1931, Soule [1]
estimated that over $50,000,000 had been spent in the United States alone on low-temperature processes during the previous 10 years. Of all the processes worked on during that period, only one—the present Disco process—survived. As a result of the widespread disillusion and the shattering losses, work on low-temperature carbonisation almost dried up in the United States early in the 1930’s.

The experience in Great Britain and on the Continent has been similar. However, due to the different pattern of fuel availability in these countries, especially the absence of the ample supplies of oil and gas that have been developed in the United States, work on low-temperature carbonisation continued at a higher level of activity.

In spite of the years that have elapsed since the early interest in low-temperature carbonisation, review of the experience of the 1920’s can be of value to modern investigators. The disappointments and disillusionments resulting from low-temperature carbonisation were well summarized by Soule. He had just finished a long period of service with one of the largest commercial installations of a low-temperature process, which had been made in the United States and which had just ended in failure.

Low-temperature projects of the period were divided into three classes, according to their primary objectives: first, the development of low-temperature char as an improved domestic fuel; second, the use of low-temperature carbonisation as a step in the complete gasification of coal to manufacture a cheap industrial gaseous fuel; and third the replacement of bituminous coal as a boiler fuel by low-temperature char.

Justification for manufacture of low-temperature char as a domestic fuel had been based on the false premise that it could be made to sell at a cheaper price than bituminous coal. Actually, low-temperature chars at that time were unable to sell for less as a domestic fuel than did high-temperature coke. Disregarding the differences in operating techniques, there were two primary reasons for this condition: the lower by-product credits for the low-temperature process, and the difficulties attendant on the transfer of heat into the coal charge.
The lower by-product credits for low-temperature carbonisation were due to the smaller yield of by-product gas. This smaller yield was of vital importance, and was rarely offset by the increased yield of tar. Hence, with a lower by-product return from the low-temperature process, it was still more essential to operate at a lower cost in order to for the low-temperature tar to complete with high-temperature ovens. These assumed that operating economies proved illusory. Transfer of heat into coal is a very slow operation, and at low temperatures, without agitation of the charge, the rate of transfer is reduced well below that a high temperatures.

Two alternatives were available, according to Soule, both of which seriously affected investment and operating costs, either coking the coal in thin layers or mechanical agitation of the charge. A thin layer construction, however, basically similar to high-temperature practice, multiplies flues and retorts and penalized the process with an excessive investment burden. To offset this, agitation of the charge was employed. Although this practice was often successful in reducing the number and cost of retorts, as a result of the agitation a weak, crumbly, light coke, much inferior in size and strength to that from the fixed retorts, was produced.

This situation left the low-temperature operator with two alternative products, a lump coke with a high investment in stationary retorts, or an inferior coke from agitated retorts of high throughput and lower investment and operating costs. Up to 1931, no commercial enterprise had been able to operate profitably on either basis.

Soule considered the two other applications for low-temperature carbonisation- a step in complete gasification of coal and the production of solid fuel for boiler use- more rational in conception. Complete gasification involved production of a low heating value gas for industrial use. His experience led to the conclusion that this should be a two-step operation, with lump char made in stationary retorts and then transferred directly, while the hot if possible, to a blue-water-gas generator in which the gasification step took place. Size and properties of the char would be of lesser importance. The low-temperature tar would be a by-product of the process, but since 1931, its value only slightly exceeded that of petroleum fuel.
oil, the credits from its production were not enough to offset the higher costs of the lump char.

In discussing the manufacture of a boiler fuel, Soule leaned toward an operation in which finely divided coal was passed continuously through a retort, heated either externally or internally. Apparently he had in mind an approach to the present day fluidised process. Two of the difficult operating problems he visualized were the separation of the finely divided dusts entrained with the outlet gases and the cooling of the finely divided coke. However, the char could not compete as a boiler fuel unless it could be produced for less money than the cost of the competing coal or other fuel. Finally, great difficulty was incurred in developing applications for the fractions or components of the low-temperature tar, and this problem still appears to be unsolved.

With the advent of fluidised or entrained processes operating on finely divided coals, in units capable of handling large tonnages, some of the difficulties inherent in the earlier retorts that plagued Soule and his colleagues have apparently been reduced or even eliminated. The char from the new processes is, of course, produced in a finely divided form, suitable for industrial firing. To obtain a product in lumps, the fine char must be briquetted, and since the binder will give off smoke on heating, a second carbonisation step must be applied to the briquettes. This means that instead of a single operation—the fluidisation step—three will be required, fluidisation, briquetting, and final carbonisation.

The alternative to producing a lump char by briquetting is to carbonise a coking coal in such a way that it forms lumps. Soule also indicated that some of the difficulties which could be encountered. In a high-temperature oven, the coke shrinks and pulls away from the walls of the oven as carbonisation is completed. In a low-temperature slot retort, carbonisation may be complete when the char has actually expanded, so that discharge of the coke by pushing may be entirely impractical. To permit discharge from these low-temperature ovens, in a number of cases the walls have been considerably tapered. With other retorts, the walls have been made movable so that they can be swung aside to permit the char to drop out onto a car or conveyor. These measures can add materially to the cost of an installation.
In carbonising a coking coal in a continuous retort, heating eventually puts it in a plastic condition where it balls up into masses that harden on the walls or screws of the retort. These accumulations may result in irregular operation or even cause production to cease. Accumulation of irregular masses on the surface causes uneven temperature conditions in the retort walls with subsequent warping and distortion. Many attempts have been made to cope with such accumulations [2]. Building heavier retorts has usually proved impractical, and periodic disassembly for cleaning has often proved to be the ultimate answer, with a resulting serious loss in production. Pre-treatment of the coal can be employed, but it adds to the cost of the operation.

On the basis of his experience with low-temperature processes, Price listed the requirements he considered necessary for successfully producing a char to be used in place of low-volatile coal in the mix for blast furnace coke [3,4]. These requirements are deserving of consideration in connection with any new installation for producing a char:

1) The prime requisite is that the char produced in the retort will make a satisfactory grade of coke, the satisfactory use of the coke in the blast furnaces being the criterion of its quality
2) Agitation of the coal within the retort appears to be desirable to prevent the coal from caking and to insure efficient and uniform heating of all coal particles
3) The retort should have as few interior projections as possible consistent with proper working to prevent build-up of carbonaceous material
4) The process of feeding coal and discharging char should be continuous
5) Access to the interior of the retort for inspection and cleaning should be easy
6) Control of heating should be adequate for handling of different grades of coal and for producing the desired quality of char there from, and the retort should be efficient in its consumption of fuel
7) It should not require that the coal be closely sized nor than all fines be removed from the coal for its satisfactory operation
8) It should have a relatively high rate of daily throughput in order to reduce the number of separate units required to simplify operation practice
9) It must be capable of being operated at sufficiently high-temperatures to insure the production of char of the proper chemical and physical properties
10) It should be of sufficiently rigid construction to withstand deformation in event of emergency shutdown
11) It should be of simple and rugged design making possible low repair, maintenance and operating cost
12) Installation cost should be as low as possible consistent with good quality of construction and with the other requirements as stated
13) It should be provided with adequate dust collectors in order that the tars and liquid products be not unduly contaminated by char dust

Hansen [5] has named five necessary conditions to be met if a low-temperature carbonisation process is to be rendered technically and economically feasible:

6) Universal applicability to different types of high-volatile coals
7) Production of a firm, abrasion –resistant product
8) Maintenance of desirable temperature conditions through the system to produce a uniform tar
9) Maintenance of an output which is economically attractive, a condition dependent on rabidity of heat transfer and involving oven design and the thickness of the coal bed
10) Smooth, trouble-free oven operation

Low-temperature processes in the United States and Canada have operated largely on an experimental basis, with two exceptions. The two processes in regular commercial use are the Disco process of the Consolidation Coal company, this plant being located near Pittsburgh, Pennsylvania, and the stoker carbonisation process, operated by Shawinigan Chemicals Ltd. in Canada.
In addition to these plants, the Parry process has been operated experimentally on lignite by the Texas Power and Light Company at the power installation for the Aluminum Company of America at Rockdale, Texas. The Lurgi-Spulgas process also has operated in North Dakota for some time on the local lignite. A process for producing a metallurgical formed coke from Wyoming subbituminous coal was announced, and production of a low-temperature char is one of the first steps. This plant is operated by Food Machinery and Chemical Company and the United States Steel Corporation at Kemmerer, Wyoming.

In Europe, Japan, and other parts of the world, the Kruoo-Lurgi, Lurgi-spulgas and Rochdale gas re-circulation appear to be most widely in use, but work is proceeding at a number of locations on smaller-scale units of other carbonisation methods.

These few plants provide a limited engineering basis for the selection or installation of a low-temperature process to meet a given need. Although many processes have been tried, they have been operated on different coals, in different ways, and for widely different objectives. Little correlation between them is available. Each process has often functioned on pretty much an experimental scale, without reaching the more critical stage of successful commercial practice. Numerous factors, such as handling the dust in the outlet gases and cooling the char, although possibly of lesser importance in pilot installation, can become serious problems when a process becomes mature and a full-scale plant is required. The engineer entering the low-temperature field finds it almost a virgin one, which encompasses first the selection of a process for a given need, then the design and engineering with the identification and definition of the problems to be faced, and finally plant operation and meeting the unexpected developments. For these reasons commercial evaluations of processes are still very difficult.

Little significant information on the costs of low-temperature processes is available, but reliable estimates are important in considering an installation. Parry [6] has given a useful relation between the costs of the coals employed, the cost of processing, and the value of the tar. For the use of the char as a power-plant fuel he expressed this relation by an equation, which can be transported into several forms:
\[ P = VT - C(1 - R) \]

where \( P \) = net cost of processing and handling fuel to the burner, which equals the total processing cost, including crushing, drying, carbonisation, tar condensing, and handling of materials, minus the total estimated cost of burning raw coal, including crushing, pulverizing, and handling; \( C \) = cost of coal delivered to the processing plant; \( R \) = net heat ratio, char/coal, obtained from assay and analysis; \( T \) = tar yield, gallons per ton, obtained from assay; and \( V \) = break-even value of the tar, cents per gallon.

A.D. Little [7] has estimated the gross receipts, which might be derived per ton of North Dakota lignite from the char and the products potentially available from the tar. The use of lignite as a fuel has been on a comparatively small scale because of its high water content—almost 50%—and its tendency to heat spontaneously in a pile, which makes storage difficult. From the $3.03 gross receipts shown by A.D. Little should be subtracted the various costs chargeable against the operation: the cost of the lignite itself, any transportation, costs of processing and of refining the tar, the costs of transportation of the various products to the market, fixed charges on the investment, and taxes.
2. Properties, Yields and Utilisation of low temperature chars

2.1 Properties of low temperature chars

Low temperature chars differ markedly from high-temperature cokes in appearance and properties. Generally, pieces of low temperature chars are black to dull grey, with an open, porous structure. They are softer, more friable, and weigh less than high-temperature coke. Both the apparent and true densities are lower. The volatile content of the char usually lies in the range from about 20% down to 9 or 10%. When heated, a char gives off much hydrogen, and the gas burns with an almost non-luminous flame. Because of the high volatile content and the open structure, char ignites readily and burns freely. These qualities make a lump char an ideal fuel for the open domestic grade. The volatile matter content of a “good-quality” charcoal depends on its use. Charcoal intended for domestic cooking typically contains 20-30% VM (with a value of 40% being marginally acceptable), whereas metallurgical charcoal often contains 10-15% (or less) VM.

Simple calculation of charcoal yield provided by a kiln, in terms of efficiency of the carbonisation process, is intrinsically vague because it does not reflect the fixed-carbon content of charcoal product, which varies widely. A more meaningful measure of the carbonisation efficiency is given by the fixed-carbon yield, which represents the efficiency realised by the pyrolytic conversion of the ash-free organic matter in the feedstock into a relatively pure, ash-free carbon. If chemical equilibrium is attained in the kiln, the fixed-carbon yield should approximate the theoretical carbon yield that results from a thermochemical equilibrium calculation. Carbon yields close to the maximum that can be obtained from a kiln operating at atmospheric pressure have been reported. It should be noted that the metallurgical industry employs the fixed-carbon content of charcoal to determine its price. Occasionally, a “charcoal carbon yield”, $y_C$, is reported, as

$$y_C = y_{char} \times \left(\frac{\%C_{charcoal}}{\%C_{feed}}\right),$$

where

$\%C_{charcoal}$ : is the carbon percentage in dry charcoal

$\%C_{feed}$ : is the carbon content of the dry feed
Because CO$_2$ and CO are evolved during the fixed-carbon analysis of charcoal, there is no simple relationship between fixed-carbon yield and charcoal-carbon yield. Beyond its proximate analysis, the quality of a charcoal can be represented by a great many other properties. These include moisture content, calorific value, elemental composition, hardness (abrasion resistance), compressive strength, bulk and true densities, surface area, porosity and pore volume distribution, electrical resistivity, and reactivity. [8]

The high reactivity of chars necessitates their thorough quenching or cooling unless they are to be used very shortly. If a warm spot is retained in the center of a lump, the head can spread, with eventual ignition of the entire mass, unless preventive action is taken. Hot, finely divided char can also ignite and cause serious trouble.

The test frequently used abroad for determining the reactivity of cokes or chars is the C.A.B. (critical air blast) method. The test is a measurement of the minimum current of air with which a given size of char will continue to burn after ignition under standardized conditions. The smaller the volume of air, the greater is the reactivity of the char, or coke, tested. Pound [9] has published the following average values, shown in Table 2.1, for several solid fuels, based on determinations in his laboratory.

<table>
<thead>
<tr>
<th>Sample</th>
<th>C.A.B. value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Char</td>
<td>0.017</td>
</tr>
<tr>
<td>Anthracite</td>
<td>0.039</td>
</tr>
<tr>
<td>High-temperature coke</td>
<td>0.057</td>
</tr>
</tbody>
</table>

He further found he could almost double the C.A.B. value of char by heating it at 900°C for 2 hr, thus decreasing its reactivity. The C.A.B. method is merely one among many such reactivity methods.
The properties of chars are greatly affected by the coals from which they have been produced and by the conditions to which they have been exposed during carbonisation. With the infinite variety of coal materials, ranging all the way from peats or lignites up to the bituminous coals which have been processed, and with the hundreds of carbonisation processes, each with its variety of conditions (among which temperature and time are especially important), few meaningful data correlating these variations with the properties of chars, and their relation to the different applications are available. Major problems in the commercialisation of a low-temperature process have been adapting it to produce a satisfactory char from the coals available and selecting the coal giving the optimum products.

2.2 Utilisation of low temperature chars – Domestic or Industrial fuel

The properties of an ideal char for domestic use were listed by Pound [9]:

1) Not friable or subject to breakage during transportation and handling
2) Suitable in size for the grate, and clean to handle
3) Readily ignitable with paper, wood, or other normal means, with the ability to catch quickly when a low fire is replenished
4) Smokeless in burning
5) Low as possible in ash
6) Although higher in price than coal, superior to coal in heating and radiation efficiency, and consistent in quality

The use of chars for domestic fuels finds its principal application Europe. In the United States, gaseous and liquid fuels have largely displaced solid, smokeless fuels. Much domestic char is produced in the form of briquettes. In Great Britain, for instance, a great deal of domestic heating is still by open grate, and briquettes or lumps of char have proved attractive in this service.

Very little char appears to be used for industrial firing, this application especially being limited by the lack of good outlets for the tar. The results of preliminary combustion trials,
according to Phillips [10], indicated that standard fuel analyses were inadequate for predicting furnace performance with the chars now being produced in certain low-temperature carbonisation processes. These chars, which he expected would become increasingly available as boiler fuels, were dry, fine, porous materials, usually containing less than 20% volatile matter. To aid in the selection of proper combustion equipment, Phillips [10] studied their classification according to the ease of ignition and speed of carbon utilization by means of three test procedures:

1) an ignition temperature test
2) a method for correlating the loss of unburned carbon in furnaces with the heating value of the volatile matter in the char
3) the determination of the relative combustion rates of fuels

In the ignition temperature studies, which measured the temperature at which a sample of the pulverized fuel produced a flame in air, low temperature chars with 10 to 20% volatile matter exhibited ignition characteristics similar to those of low-volatile bituminous coals. In tests on the losses of unburned carbon made with different fuels in the same furnace, the carbon losses tended to decrease as the heating value of the volatile matter decreased. The usefulness of this method of correlation appeared to be restricted to the natural fuels, and additional information was required before such fuels as low-temperature char could be compared to with these fuels of known performance. Results of the burning rate tests indicated that the low-temperature chars would burn in commercial furnaces at rates similar to fast-burning coals.

2.3 Utilisation of low temperature chars – Blast furnace fuel

Char, even in lump form, has been unsuitable for direct use in the normal blast furnace or cupola, owing to its low strength and high reactivity. Its use in high-temperature coke mixes or in briquettes, which are subsequently coked, has been necessary before a char could be applied satisfactory. In several plants, chars with volatile contents in the range of approximately 15 to 20% -near that of the low-volatile coal displaced - have been combined
with the coal blend to the high-temperature ovens to produce a satisfactory metallurgical coke. The experience of operators, both in the United States and abroad, has indicated that the substitution of a slightly greater amount of char of the proper quality for the low-volatile coal has given good results.

Price [11] has listed desirable requirements for a coal to be used in the production of suitable chars:

1) Capability for producing the proper quality of char from the point of view of coke improvement
2) Low ash content
3) Freedom from excessive gumminess on heating, or susceptible to blending to minimize such a tendency
4) Low content of sulphur, phosphorus, and other objectionable impurities
5) Low moisture content, to avoid pre-drying
6) Low delivered cost

Past work indicated that the following characteristics, other than the coal selected, required consideration in the production of a satisfactory coke when char is used as a component in the blend:

1) Amount of char—if more than approximately 20% is used, the resulting coke is grainy in structure and has lost strength
2) Pulverization of 100% through an 1/8” screen has usually been adequate
3) Volatile content should preferably be at a point between 15 and 20% and variations from the selected value of more than 1% impair the effectiveness of a char
4) Physical properties, especially surface condition

Physical characteristics have appeared largely depended on the coal, the carbonisation temperature, and the time in the retort. Better cokes have usually been prepared from chars produced as cenospheres or with a distorted surface, rather than from a char with an unaltered
appearance. In general, carbonisation temperatures approaching 750°C have resulted in better chars than those produced at lower temperatures of about 500°C.

Reed et al. [12] produced metallurgical coke of good quality by coking in test ovens coal blends including char made from Illinois coals. Neither the percentage of char nor the uniformity of its volatile matter content in the range studied was found critical. However, the quality of the high-volatile coal used for blending was of great importance. Comparisons with chars produced in several types of retorts indicated that retort design and consequent operating procedure influenced the properties of the coke.

Burstlein [13] patented a method by which durain, fusain, and chlarain were separated from a coal prior to high-temperature coking. These constituents were then precarbonised to char 450° to 750°C and mixed with the balance of the coal before it was charged into the coke ovens. The constituents were separated by grinding the coal and screening.

Char was added to the coal mix in proportions of 15 to 25% by Petit [14] for the production of metallurgical cokes. Small spheres of 1 to 3 mm diameter, which were produced by a fluidised process, gave excellent results. Char from rotating horizontal retorts, however, proved unsatisfactory for the preparation of a good coke.

Japanese experiments on the use of char in the coal mixes used for the preparation of blast furnace coke have been described [15]. After World War II in Japan, it was necessary to produce coke for blast-furnace use, with at least an 85 drum index value, by using only domestic high-volatile coal. Hisada also described experiments in which coke with a strength of over 90 on the drum index was obtained commercially by coking a mixture of a coal of high fluidity blended with about 30% of char containing 20% volatile matter, and ground finer than 0.3 mm in a tube mill. The optimum volatile content for the char proved to be 18 to 20%. Results of operation with the high-temperature coke produced from the blend in several blast furnaces were very good. Use of the char in the coal blends increased the proportion of carbon monoxide in the gas and the viscosity of the tar from the coke ovens.
Work in Australia describes the use of char in production of metallurgical coke [16]. When low temperature carbonisation has been used as a step in the manufacture of a high-quality metallurgical coke from a non-coking coal, pre-carbonising the coal at low or medium temperatures has been the primary step. The char produced was crushed, if necessary, and the fine particles briquetted, usually using a coal-tar pitch. The briquettes were then carbonised in vertical ovens at temperature sufficient to impart the strength required for metallurgical use, and at the same time drive-off the volatile matter from the binder. The use of a coal-tar pitch of softening point 65 to 75°C, usually cutback with an equal quantity of tar oil, has been mentioned. Baum [17] produced metallurgical coke by briquetting low-temperature char with a few percent of a swelling-type coal and a binder, and carbonising the briquettes at high temperature in direct contact with hot gases.

The successful production of coked briquettes, from an Australian subbituminous coal, was found to depend on securing full preshrinkage of the chars [18]. They shrank approximately 20% over the temperature range, 500 to 1000°C, so it was necessary to carbonise the raw coal to at least 800°C to preshrink the char adequately. Thereafter, the char was briquetted with a suitable binder and carbonised at 500°C, which gave a strong coked briquette.

Low-temperature carbonisation was used as a prior stage in the production of a carbonised fuel, according to a patent of Berry [19], which disclosed three distinct stages of heating and carbonisation, comprising:

1) Heating the coal to 275 to 400°C, during which only moisture but no tarry vapours were evolved
2) Continuing heating to approximately 500°C to produce the low-temperature char
3) Conversion of the char to high-temperature coke by heating to 900°C maximum

According to Malcor [20], a small char (Coalite) was used directly as a fuel in a low-shaft blast furnace at Ougree, with excellent results. This char was made at Bruay from long-flame coal and had the following analysis: moisture 15% by weight, ash 12%, and volatile matter 9 to 12%. With dust-free St. Pierremont ore, the combustion of char was about the same order
of magnitude as with breeze, so that the recovery on the basis of the fixed carbon was about 13% higher with the char. This indicated that the volatiles in the char played a part in reduction of the ore. The char appeared more reactive than breeze.

2.4 Utilisation of low temperature chars – Reducing agent

The reducing agent commonly used in the production of such elements as phosphorous and copper from their ores has commonly been a high-temperature coke. However, low-temperature processes, especially those of which are a first step in the manufacture of briquettes, make possible the utilization of non-caking coals. In this way, lower rank coals, especially the lignites and sub-bituminous coals in the extensive western coalfields, may be made available for wider commercial applications. The interest in this direction is indicated by the increasing attention in which these western coals are employed.

2.5 Utilisation of low temperature chars – Electrodes

Shea produced Electrodes from coal in a process, which included carbonisation of the coal at a temperature between 300 and 550°C [21]. The volatile matter was reduced to between 8 and 20%. The char was crushed, the particles briquetted under pressure without a binder, and the briquettes heated to a temperature in the same range. After a final crushing, the final char was cokes at a temperature above 700°C.

2.6 Utilisation of low temperature chars – Other uses of chars

Low-temperature carbonisation has often represented a primary step in the complete gasification of coal. Non-coking coals have charged to carbonising chamber located above the gas producer. Such processes have been discussed by Gumz and Hubmann [22]. During World War II, some char was gasified in portable gas producers to drive motor vehicles, both in Japan and Germany.
Additional uses of char have been described, the service depending on some special property of the char, such as its reactivity. Nunderloh [23] pointed out that soda-treated lignite coke could be used satisfactory as a substitute for charcoal in chemical processes, such as carbon disulfide production. This finding was based both on experiments and on industrial experience.

Low-temperature carbonisation was employed in laboratory tests as a step in the production of ethylene and aromatics from lignite. Texas lignite proved much superior to North Dakota lignite and was carbonised at a temperature not over 550°C. The gases at the exit of the retort were immediately cracked at about 800°C to produce 6 lb of benzene and 40 lb of ethylene per ton of dried lignite.

Berg and Atkinson [24] have pointed out that non-coking coals are distinguished in great abundance throughout most of the country, and the char that would result from their carbonisation is thus readily available.

Because low-temperature carbonisation can be applied to other carbonaceous solids, such as sawdust, coal fines, lignite, or peat, with the production of useful chars, the process may also be utilized to upgrade some of these materials, when they cannot be utilized directly with satisfactory results [25].
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CENTRE for RESEARCH and TECHNOLOGY HELLAS
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OVERVIEW of LOW TEMPERATURE CARBONISATION

Present Status – Properties, Yields and Utilisation of LTC chars – Survey of
Various Methods – Pre-treatment Conditions & Effects – Advantages, Economic &
Technological Development

Part II. Survey of Competitive Carbonisation Methods

Report prepared for
OPET Network

EUROPEAN COMMISSION

ISFTA – May 2004
1 Low temperature carbonisation process

Low temperature carbonisation of coal may be defined as carbonisation in the absence of air, which is carried on to a final char temperature below 800°C. This process aims to convert the dirty black rock called coal to clean fuels, liquid fuels or pipeline gas, and more valuable chemicals.

The principal problems faced by these processes are how accomplishing the following:

- Provide the necessary process heat
- Handle the caking tendencies of the solids
- Separate the resulting three phases (solids, tars, gas)
- Upgrade each of three phases to useful products
- Corrosion-erosion
- Deal with toxic products and by-products
- Accomplish all these tasks efficiently and economically

The initial change that occurs in carbonisation is the breakdown of the coal substance to simpler, distillable material. This breakdown is a function of the type of coal and of temperature, which, in turn, is directly dependent on the energy transferred to and into the solids. Therefore the basic controlling factor in these processes is either the rate of transfer of heat into the coal solids, or the rate of the decomposition reactions. The finer the grind of the coal, the more likely it is that the reaction rate will be controlling.

The low temperature carbonisation processes can be classified according to the design of the retorts, the character of the solids carbonised and the character of the char produced. The design of the retorts differs according to the operation, batch or continuous, the layout, vertical or horizontal, the stages, single or multiple and finally the heating method, indirect, direct or both.
2 Classification of low-temperature carbonisation processes

The great number and variety of the processes available for the low temperature carbonisation of coal make necessary some system of classification. The main factors on which classifications may be based are the general design of retorts, the character of the coal carbonised, and the character of the char or char product produced. However, the classification following in next paragraphs is based mostly on the physical characteristics of the retorts.

2.1 Vertical retorts

Vertical retorts embrace many different carbonisation devices, possibly more than any other arrangement, and they include both batch and continuous types, with some processes resembling existing types of high-temperature coke ovens. In vertical retort processes, either batch or continuous, the coal is charged at the top, as with a high-temperature coke oven, but discharge has usually been from the bottom. Sizes of retorts are based on securing the maximum throughput per unit of capital cost. Because coal and char are very poor thermal conductors, and penetration of heat through layers of these materials into the center of the charge is slow, retorts indirectly heated by conduction of heat through the retort walls are usually narrow, resulting in relative small capacities per retort. To increase rates of heat transfer and carbonisation, some retorts have been made of steel, and in other resort has been made to internal heating of the charge. Internal heating is affected by circulating hot gases, either the products of combustion of a fuel or preheated gases, such as make-gas, though the retort. Carbonisation has thereby often been accelerated, and the throughput of a retort increased materially.

Vertical retorts have been used for carbonising a wide variety of coals, caking and non-caking, and various sizes, such as slack, lumps, and briquettes. However, all types of vertical retorts are not adapted to all coals. Thus, when caking coals are plastic the usually flow together to form large masses which adhere to the retort walls. Unless the plastic properties are reduced by a prior oxidation or other treatment, such a coal is usually best carbonised in a
batch retort designed to permit ready discharge of the char. For continuous retorts, sticking of char in the retorts cannot be permitted, and only coals possessing a low caking power should be introduced.

Construction materials for vertical retorts range from steel to various refractories. Higher temperatures to speed up carbonisation of the charge are, of course, applicable with refractory retorts. Similar methods of heating, indirect, direct or a combination of both are used with both types of materials.

2.2 Fixed bed retorts – Indirectly heated fixed bed retorts

Fixed bed retorts consist essentially of chambers, which are heated indirectly by circulating the products of combustion of a fuel gas through flues built in the walls. Because of the lower temperatures and the consequent slower rate at which carbonisation proceeds, retorts must be much narrower in width than coke ovens to obtain a satisfactory rate of production. Typical processes take place in indirectly heated fixed bed retorts are:

- Krupp-Lurgi process
- Brennstoff-Technik
- Cellan-Jones ovens
- Carmaux process
- Otto retort
- Weber process
- Phurnacite process
- Parker retort

2.3 Krupp-Lurgi process

The Krupp-Lurgi [1-4] process was developed in Germany during the 1930’s, and up to the close of World War II a number of plants, each with a daily capacity of several hundred tons, had been constructed in that country. During the war the process proved important to
Germany because the low-temperature tar from the Krupp-Lurgi plants was a useful source of the fuel oils required by the German military forces. In recent years, the availability of petroleum oils appears to have greatly diminished the need for the process.

Figure 1 shows a cross section of a Krupp-Lurgi coke oven. The coal was carbonised in units, each consisting of six narrow carbonisation cells contained between seven rectangular steel heating cells, though which were recirculated hot gases. A unit of six carbonisation cells constituted an oven, and each cell was approximately 10.5 ft long by 6.9 ft high by 3 in. wide at the top and 4 in. wide at the bottom. Each oven had a volume of about 125 ft$^3$ and carbonised about 2.75 metric tons/charge.

![Figure 1](image)

**Figure 1** Diagrammatic drawing of a Krupp-Lurgi low-temperature coke oven.

The ovens were constructed entirely of steel plate. After several years of operation, the German ovens were in good condition and a life of 10 to 12 years was expected. The ovens were heated by recirculation of the hot gases through the heating cells. The combustion gases from a common, gas fired combustion chamber were recirculated by a fan through ducts running to and from the oven. The circulating gases flowed into the flues at the bottom of each heating cell, up between the steel walls, out at the top, and back to the furnace. At regular intervals, the direction of the combustion gases through the flues was reversed to heat uniformly the walls of the chambers. The temperature of the heating gas entering the heating
cell was about 610 to 620°C and leaving the cell at about 560 to 580°C. The drop in temperature of about 40°C represented sufficient heat to carbonise adequately the coal in a cell, both at the bottom where it was 4 in. thick and at the top where it was only 3 in. thick.

Figure 2. Flow diagram for recovery of low temperature char, tar, ammoniacal liquor and benzine from Krupp-Lurgi low-temperature carbonisation plant.
The flow diagram of Figure 2 shows the manner in which the gas from the ovens was treated to recover tar, ammonia liquor, and light oils. The quantities on the low diagram, of coal carbonised, char, and other products made, apply to 24 of the newer ovens installed during the war (1943) at the Wanne-Eickel plant of Krupp Tribstoffwerke G.m.b.H. The plant at Wanne Eickel then contained 56 ovens with an annual capacity of 204,000 metric tones of char, 1200 tons of fuel oil, and 1920 tons of motor fuel.

Normally at Wanne Eickel a blend of high-volatile Ruhr coking coals was carbonised: about 25% Hannobal coal with 22% volatile matter, and 75% Bismarck coal with 33 to 34% volatile matter. The charge mixture usually contained 8 to 10% moisture, 30% volatile matter, and 5.4% ash. The coal size for Krupp-Lurgi ovens was preferably 77% minus 1/8 in. and about 5% minus 100-mesh. By stamping the coal during charging, the density of the charge could be increased to about 8% over the normal density of 48 lb/ft$^3$. A fuel gas containing 220 to 250Btu/ft$^3$ was burned in the combustion chambers, and the heat input amounted to 970 to 1050 Btu per pound of coal charged.

About 3350 ft$^3$ of gas were produced per ton. The yield of dry char amounted to about 84% of the dry coal. The larger coke was consumed in a Fischer-Tropsch plant, which produced water gas with a CO:H$_2$ ratio of 1.35 to 1.50:1. The smaller sizes were used in house heating and in the small mobile gas producers.

German coal, containing 27% to 38% volatile matter, have been of most interest for use in the Krupp-Lurgi ovens because they have given chars of good quality plus high yields of tar and light oil. When gas coal containing 30% volatile matter, 9% moisture, and 5.8% ash was carbonised, the produced char was composed of 7.2% ash and 9.3% volatile matter. The tar yield was 6.4% and the light oil yield 0.67% of the weight of the dry coal. Others chars from ovens in the Saar ranged in drum stability from 50 to 80%, their apparent specific gravities were 0.8, and the porosities 42 to 45%. The volatile matter ranged from about 12% for the large sizes to 16-18% for the small sizes. About 12% of this char was −1/2 in. breeze, which was used in the coal mix to high-temperature coke that was nearly as good as coke from mixes in which special Ruhr coal has been blended.
In the high-temperature coke oven, the plastic zone moves steadily inward from the walls after charging in completed until it meets the center of the oven shortly before it is pushed. However, in the narrow Krupp-Lurgi ovens, at the lower temperatures, no definite plastic zone existed, but all the coal in the entire oven was in a plastic condition at one time. Formation of a good low-temperature char was attributed by the designers of these ovens to the rapid and simultaneous increase in temperature throughout the plastic mass (350 to 450°C). The faster the temperature increased in this range, the more complete was fusion of coal constituents with increase in the strength of the char. Studies have shown that the temperature started to rise in the middle of the narrow, low-temperature ovens shortly after charging, even when they charged with coal containing several percent of water. Special tests indicated that in \( \frac{1}{2} \) hr all the water had been driven off from a coal containing 8%, in 2 hr the coal was entirely in a plastic state, and after 3 hr it had been converted to char. Unlike high-temperature coke, which breaks in the center of the oven, the char normally showed no tendency to divide in the center of the oven; the pieces were uniform from wall to wall. Analyses showed that virtually the same volatile content in the char from the center of an oven as from the walls. Furthermore, retention of char in the oven for an additional 2 hr after carbonisation has been completed changed the properties very little.

Krupp-Lurgi retorts have been used not only for carbonisation of coals or coal mixtures, but also for briquettes. As applied to brown coal and lignite, the Krupp-Lurgi ovens were a part of a three-stage process. The coal was first passed counter-current through a stream of hot gases at about 1000°C. At this temperature, it disintegrated into a powder, which was compressed in a ring-roll press at a pressure of 2000 to 2500 atm into briquettes. The hard and very dense ovoids were finally carbonised in the Krupp-Lurgi ovens. In Germany, at the end of the last war the production of briquetted char from lignite was reported to be 3800tons/day.
2.4 Brennstoff-Technik

The objective of the Brennstoff-Technick [5,6] process was to achieve good yields of char in lump form from weakly coking coals by rapidly heating the coal in very narrow ovens. Both experience and tests have shown that the plasticity of a coal increases with the rate of temperature rise. The relationship is illustrated by Macura in Figure 3, and shows the pronounced increase in plasticity of a coal when the rate of heating was increased from 1.43 to 14.6°C/min [5,7].

![Figure 3](image)

**Figure 3** Curves showing the relationship of plasticity to the rate of temperature rise on coal carbonisation.

In the Brennstoff-Technick process, the desired high heating rates were secured, not by the use of high temperatures, but primarily by making the ovens narrow, usually in the range of 60 to 125mm. The narrower ovens were used for the weaker caking coals. It was claimed that these ovens might be used to process high volatile, weakly caking coals, which could not be carbonised in ordinary coke ovens.

Brennstoff-Technick ovens were constructed of steel to benefit by the much higher heat conductivity of iron as compared to silica refractories. The temperatures in the retorts seldom exceeded 650°C, a level at which the coke has not shrunk sufficiently for easy pushing. Because of this condition and of the complications which pushing the multiplicity of narrow
ovens would entail, the heating walls, which contained the flues, were pivoted at the top so that they could be swung aside, thus allowing the coke to drop out into the quenching car.

A battery of Brennstoff-Technick ovens consisted of several blocks of ovens, each block usually containing twelve ovens. All the ovens in a block were charged simultaneously from a car running along the top of the battery. Carbonisation of the oven charge, usually about a ton in weight, was complete in approximately 2 hr if the coal was briquetted and 4 hr when unbriquetted fine coal was charged. After carbonisation has been completed, the walls were pushed apart by a mechanical device, and the coke char was dropped into the quenching car and was quenched with water.

The ovens were heated by hot combustion gases, which were recirculated by means of a blower through the flues of the walls and then through a combustion chamber. In this chamber, fuel gas was burned with air and the products of combustion were added to the recirculating gases to maintain them at the desired temperature. The ducts, which conducted the hot combustion gases in and out of the main flues of a block of ovens also served as the trunions for the movable walls. Figure 4 shows the general arrangement of a block of Brennstoff-Technick ovens. Yields of products obtained by coking a weakly caking coal having a volatile content of about 30% are shown on Table 1. After the heating requirements for the retorts and coal dryers were met, only about 700,000 to 800,000 Btu in the surplus gas per ton of coal processed was available for distribution.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Product yields from Brennstoff-Technik process</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Yield per ton (dry coal)</td>
</tr>
<tr>
<td>Char</td>
<td>1500 lb</td>
</tr>
<tr>
<td>Tar</td>
<td>19.7 gal</td>
</tr>
<tr>
<td>Light oil</td>
<td>2.6 gal</td>
</tr>
<tr>
<td>Ammonia</td>
<td>2.2 lb</td>
</tr>
<tr>
<td>Total gas</td>
<td>4550 ft³</td>
</tr>
<tr>
<td>Gross heating value of gas</td>
<td>680 to 793 Btu/std ft³</td>
</tr>
</tbody>
</table>
In Germany, Brennstoff-Technick ovens were employed in a number of trial plants, of which the Berlin-Neukolln Gas Works was the most important [8]. This plant, with a capacity to carbonise as much as 50 to 60 tons of coal or briquettes per day, was operated from 1938 to 1944. The tests included the use of ribbed walls, which transferred the heat into the heart of the charge, and considerably increased the heating surface of the chambers so that the coking time was reduced from appreciably and the production rate increased. Successful results in the Berlin-Neukolln plant led to the design of three large plants, each with an annual capacity of 500,000 tons. However, at the close of the war, work on these plants, all in various stages of completion, was suspended. In a plant erected at Marienau in the Saar, however, work on the process was continued. For a number of years, about 154 tons of coal were carbonised each day, not as slack, but in the form of briquettes, with cellulose sulphite as a binder.
Figure 4. Brennstoff-Technik low-temperature oven for carbonization of bituminous coal fines.
Scherer [9] has reported the processing of weakly caking long-flame coals in Brennstoff-Technik ovens, each with a width of 90 mm and a carbonisation time of 2.5 hr, to produce 110 tons of briquettes per day. The briquettes from the retorts were subsequently heated in a shaft retort to about 700 to 800°C by means of hot gases passed upward through the retort, thereby reducing the volatile matter 3-5%. The final coke had a drum resistance of about 90% and was suitable for both metallurgical and domestic use. The tar represented 11% by weight of the raw coal. In addition, 2470 ft$^3$ of gas having a heating value of about 843 Btu/ft$^3$ were produced from the ovens. In the shaft retorts, 7600 ft$^3$ of lean gas (heating value 225 Btu/ft$^3$) were produced per ton of coal.

2.5 Cellan-Jones ovens

In 1941, an experimental battery containing fifty-one low-temperature coke ovens was erected in Great Britain [11,18]. These ovens were built of special, high-alumina fireclay brick, and resembled conventional high-temperature coke ovens in both design and operation. Each oven was heated by burners operated on the underjet principle, with the flames burning in a horizontal combustion flue located below the level of the oven floor. The hot gases flowed from the combustion flue up through the vertical flues, which were divided to centrally by a thick brick partition to give a hairpin arrangement. The brickwork partition provided a reservoir of heat. The gases leaving the vertical flues were conducted through a horizontal sole flue into a vertical waste-heat flue located remote from the oven end. More heat could be supplied to the sole flue by an extra burner. No regenerators were provided and the flue gas was not reserved.

These British ovens, carbonising from 5 to 6 tons of coal per charge, had a daily throughput of 200 tons with a brickwork temperature not exceeded 750°C. The Vane Tempest coal charged contained 7% moisture, 32% volatile matter, and 5% ash-with 85% of coal passing the ½ in. screen. The yield of coke was from 72 to 73% of the coal charged, with not more than 7% of it breeze. In addition, 16 gallons of tar and 5500 to 6000 ft$^3$ of gas, with a heating
value of 680 Btu/ft$^3$, were produced. About 10 to 15% of the gas was surplus. The coke was sold for domestic use and as a producer fuel. It contained up to 8% volatile matter, and possessed a bulk density of 36 lb/ft$^3$ and a Critical Air Blast reactivity value of approximately 0.03. The ovens are no longer operating.

### 2.6 Carmaux process

In the Carmaux [4,6,9,10] process, which was operated in Southern France, coal briquettes were carbonised in tall, narrow, vertical retorts after having been subjected to a preliminary heating for about 4 hr in the presence of air at 250°C. This treatment was designed to oxidize the surface of the briquettes with formation of a thin “skin”, thus preventing their softening or sticking when they were processed in the slot retorts. The preliminary heating chamber was a smaller, cylindrical vessel located above the main retort, and here the raw briquettes were heated by direct contact with a mixture of air and hot combustion gases ascending from the flues of the retort. An exhauster was used to pull the gases through the bed of briquettes.

The main retorts were constructed of steel and were heated by means of flues located in the long slides. Steam, produced in a waste-heat boiler, was introduced into a char-cooling chamber located at the lower end of the retort. Here is first cooled the char, then it carried the heat from the char up into the retort, and finally it swept the vapours evolved from the coal out of the retort and thus prevented their cracking on the retort walls.

The coal used was of 6-mm size and contained 25% volatile matter and 10% ash. About 6% pitch was added to serve as binder for the briquettes. When pre-treatment of the briquettes in the upper chamber was completed, about one-third of a charge in a retort was discharged from the bottom, and the contents of the pretreating chamber were dropped into the retort. The pre-treatment chamber was then recharged with fresh briquettes.
2.7 Otto retort

An Otto [8] low-temperature carbonisation retort was developed from the Otto vertical chamber retort used for gas manufacture. The charge of coal briquettes was heated indirectly by gas burned in twin vertical flues, a system which was reported to provide good control of temperatures and which gave maximum tar yield and optimum char properties. Non-caking coals were desirable charge materials. An experimental retort with a capacity of 4 tons of coal per day was operated in Germany from 1938 to 1944, and from it briquettes suitable for use in generators and a dust-free low-temperature tar was obtained. Although a larger plant was designed from the results of the experimental retort, the close of the war prevented the installation.

2.8 Weber process

Another process in which briquettes of char or of a non-caking bituminous coal were carbonised in vertical-slot, batch retorts was the Weber [2,8] process. A flow sheet of the process is shown in Figure 5. The coal was elevated to a crusher, from which it passed through a feed hopper to a mixer. Here binder comprising sulphite pulp liquor plus an aqueous suspension containing about 1% clay were added with heating and vigorous agitation. The briquettes, formed from the paste in a roll press, were dried on a slowly moving belt until most of the water had been expelled. They were then carbonised at a low temperature to produce “very reactive, dense, hard, and smokeless-burning briquettes or ovoids”.

Because the binder possessed no softening tendency, as does pitch, the briquettes should not become plastic and stick together, thus obstructing the retort.

### 2.9 Phurnacite process

The “Phurnacite” [8,11] process carbonised coal briquettes in tall, vertical slot ovens at low or medium temperatures to produce a smokeless domestic fuel. The ovens were provided with vertical flues and recuperators, with the products of combustion of a fuel gas recycled through the flues. The briquettes were charged in at the top of an oven. After carbonising for about 4 hr, the door at the bottom was removed and the hot ovoids discharged onto the inclined oven floor and into the quenching car. An installation erected in South Wales in 1942 consisted of two batteries, each containing forty ovens, with a daily capacity of about 1000 tons of the raw ovoids.

### 2.10 Parker retort

The Parker retort [12,13] for the production of a smokeless domestic fuel was an early development in British low-temperature carbonisation practice. The first installation was made near Barnsley in 1927 and was followed by two further installations. A plant at Bolsover, which went into operation in 1936, was enlarged to 19 batteries with a total of 728
retorts. Another at Askern near Doncaster contained 9 batteries with 360 retorts. A retort consisted of a multiplicity of small, vertical tubes with a tapering cross section in which the coal was carbonised. These tubes were about 9 ft long, with a diameter of 4-½ in. at the top and 5-¾ in. at the bottom. Twelve of these tubes contained in a single monobloc casting of special cast iron formed a retort, and forty retorts were combined into a battery heat. The retorts were heated only by radiant heat, which was supplied by the combustion of fuel gas in chambers arranged so that they alternated with the retorts throughout the battery. The temperature of the retorts was maintained at approximately 650°C, and approximately 4-½ hr were required for carbonisation. Products of carbonisation were withdrawn from an off take near the top of the retort. A cooling chamber, large enough to hold the char from the two retorts, was located below each pair of retorts. This chamber was fitted with double doors at the bottom, an inner door to support the char, and a water-sealed door below to prevent admission of air. Average yields of products per ton of coal carbonised at Bolsover were as follows:

<table>
<thead>
<tr>
<th>Product</th>
<th>Yield</th>
</tr>
</thead>
<tbody>
<tr>
<td>Char</td>
<td>1500 lb</td>
</tr>
<tr>
<td>Crude tar</td>
<td>16.5 gal</td>
</tr>
<tr>
<td>Crude light oil</td>
<td>3.5 gal</td>
</tr>
<tr>
<td>Aqueous liquor</td>
<td>30.0 gal</td>
</tr>
<tr>
<td>Gas heating</td>
<td>4000 cu ft</td>
</tr>
<tr>
<td>Gas heating value</td>
<td>700 Btu/cu ft</td>
</tr>
</tbody>
</table>

The char, termed “Coalite” is sold for smokeless domestic fuel.
3 Fixed bed retorts – Directly heated fixed bed retorts

In addition to the fixed bed retorts which are heated by transmission of heat through the walls, a number of other retorts have been developed in which carbonisation is effected by direct contact of the coal with hot gases such as:

- Rexco process
- Records “Coalene” retort
- Karrick retort

3.1 Rexco process

In the Rexco [13,14] process, a low temperature char in the form of lumps has been produced by carbonising a static charge of the coal at approximately 700°C in large capacity, vertical chambers which were heated internally by combustion of a portion of the charge. The coals used in this process were weakly coking or noncoking, and were charged in the form of lumps, 4 in. by 2 ½ in., and 2 ½ in. by 1 in. The retorts were vertical, cylindrical vessels, about 11 ft in diameter and 25 ft high, consisting of a mild steel shell lined with firebrick, with a capacity of about 34 tons of charge.

When a retort was filled, about 23 tons of the larger size lumps were charged into the lower portion of the retort where they were supported on a grate. The coal flowed down and was distributed uniformly by means of a telescopic chute, thus preventing the shattering of the lumps, which would otherwise occur if the coal were allowed to fall freely. Above the larger lumps were laid about 11 tons of the 2-½ in. by 1 in. coal. The telescopic chute was gradually withdrawn as filling of the retort proceeded. When charging was complete, gas burners above the top layer of coal were ignited, and the hot products of combustion were drawn downward through the charge by an exhauster, thus carbonising the coal in the retort. The volatile products of carbonisation evolved from the coal joined with the hot gases, and the mixture passes downward through the grate to the tar recovery equipment and the exhauster. From the exhauster a portion of the cool, tar-free gas was recycled to the dome, which formed the
top of the retort. Here it entered through the two tangential burners and was burned with air to provide the heat of carbonisation. Carbonisation proceeded regularly for 8 or 9 hr until it was substantially completed in the upper carbonised mass to complete carbonisation of the lower portion of the charge. Accordingly, the air to the gas burners in the combustion chamber in the dome was shut off, and gas merely recirculated through the charge for about 6 hr more to complete carbonisation. During the combustion stage, the amount of air to the burners was strictly controlled to reduce to a minimum the excess air, which burns a portion of the charge.

As the hot carbonisation zone moved downward through the retort, the volatile products, which were evolved mixed with the hot gases, but the combined gases immediately entered a cool layer of coal so that secondary cracking of the tars was prevented. When carbonisation was complete, the circular grate was withdrawn, and the char fell onto the inclined coke bench, which was suited under the retort. The Rexco char had been substantially cooled by means of the circulated gas by the time it was discharged from a retort. It was screened into the sizes required by the domestic consumers. The entire operation required approximately 16 ½ hr, made up as follows: preparing and charging the retort 1 ½ hr, carbonising the upper portion of the charge 8 hr, carbonising the lower third of the charge and cooling 7 hr. Since about 34 tons of coal were carbonised per charge, the daily throughput of a retort was about 45 tons.

The retort was simple and sturdy in construction, without flues or heat transfer surfaces. The original Rexco plant consisted of two retorts, which were installed about 1936 by the National Carbonising Co., Ltd. at Mansfield in Nottingham to supply the char, termed “Rexco”, as a fuel for domestic use. Increasing demand for the product led to installation of a third retort of the same dimensions as the two original ones. The new retort was improved by redesign of the combustion chamber so that the heating gas was burned in two separated stages. With the new arrangement, better control of carbonisation temperature to maintain it below 700°C was possible. In addition, the overall time of carbonising a charge could be reduced to 13½ hr, so that approximately 55 tons of coal was the daily throughput. The general arrangement of a retort is shown in Figure 6.
Figure 6  Diagrammatic arrangement of a "Rexco" plant.
Average analyses when carbonising a noncoking coal of medium quality, which were obtained during 11 years of operation, are shown in Table 2.

**Table 2** Rexco process-analyses of coal charged and char produced.

<table>
<thead>
<tr>
<th></th>
<th>Coal</th>
<th>“Rexco” (Char)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture, %</td>
<td>5.3</td>
<td>5.4</td>
</tr>
<tr>
<td>Volatile matter, %</td>
<td>34.6</td>
<td>6.6</td>
</tr>
<tr>
<td>Fixed Carbon, %</td>
<td>55.2</td>
<td>80.8</td>
</tr>
<tr>
<td>Ash, %</td>
<td>4.9</td>
<td>7.2</td>
</tr>
</tbody>
</table>

Yields of products per ton of the coal were: char, 1360 lb; tar oil, 19 gal; and gas, with a heating value of approximately 140 Btu/cu ft, 3 million Btu.

The “Rexco” char was dense, hard, and slow burning, with a high degree of reactivity. The results of burning test made on Rexco by the Fuel Research Station of the Department of Scientific and Industrial Research are shown in Figure 7. It indicates that the duration of intense fire with Rexco was more than twice that with the standard coke. Briquettes made from coal fines with a pitch binder could be carbonised in a Rexco retort with elimination of the smoke-producing quality of the briquettes.

**Figure 7** Fuel Research Station tests on “Rexco” fuel. Duration of intense fire is twice as long as with a standard low-temperature char.
A somewhat similar carbonisation process was patented by Shaha. It involved partial combustion of the coal at the bottom of the retort to furnish the hot gases for converting the upper portion of the charge into a soft coke.

The semi continuous Horner process described by Churcill bears an even closer resemblance to the Rexco process. Hot gases from a combustion chamber above the charge on a grate were drawn down through the coal while the charge was raised progressively. The finished semi coke was removed intermittently from the coking chamber by means of a horizontal slicer bar. When the entire charge was coked, the grate was lowered, a new charge added, and the cycle repeated.

3.2 Records “Coalene” retort

In the Records “Coalene” retort [15-17], fine coal was coked by a batch process in an inclined retort by direct contact with steam at about 10-lb pressure and superheated to from 540 to 650°C. The coal charge, which was admitted into the retort through an upper door, was carbonised by direct contact with the hot stream, as well as indirectly by hot gases from a combustion chamber located below the retort. Before introduction into the retort, the steam was superheated in a steam coil located in the combustion chamber. Steam and volatile products were withdrawn from the top of the retort and condensed. The char was discharged through the bottom door of the retort on completion of coking, and was quenched. Figure 8 shows a diagram of a retort.
A small, eight-retort plant, installed at Wellington, Utah noncoking coals. This plant was designed to carbonise the coal charge in about 1.5 hr with a yield of from 1300 lb to 1600 lb of char and 40 gal of tar per ton of coal. Much of the gas was used for process heat.

Additional designs developed by the inventors included a special, vertical, cylindrical container in a perforated basket was charged before the superheated steam was introduced. Two of these retorts with a total capacity of from 15 to 20 tons of coal per day were installed in Tacoma, Washington. A third design was a vertical cylinder in which the coal was carbonised continuously in the same general way.

3.3 Karrick retort

The Karrick process was basically much like the Records process. Superheated steam at about 650°C and 10-psi pressure was passed into the top of a vertical, cylindrical retort about 10 in. in diameter and 10 ft long. The volatile products were removed at the bottom. Both batch and continuous retorts were operated by the U.S. Bureau of Mines in a pilot scale, but the process never advanced beyond this stage. The results of the work indicated that steam is an expensive heating medium.
Continuous vertical retorts operate with coal charged continuously in the top and char removed at the base. The height is sufficient so that carbonisation is completed by the time the coal reaches the base. Some retorts are narrow chambers similar to the slot ovens, the majority are round or ellipsoidal shafts, usually a single shaft, but some are multiple or concentric shafts.

Continuous vertical retorts have been heated, either indirectly by hot combustion gases passing through flues in the retort walls, or directly by means of hot gases ascending through the coal charge. Narrow, slot-type ovens or thin layers of coal have been required where indirect heating alone was applied, but this method of heating has been supplemented in many retorts by introduction of hot gases or steam at the base of the retort. Introduction of a hot gas accelerates carbonisation of the charge by carrying heat up uniformly through the mass of descending coal. When the coal is carbonised by direct contact with the hot gases, the cross section area of the retort may be made a great deal larger than would be possible if it were indirectly heated. The hot gases, introduced for direct heating of the retorts, include the products of combustion of fuel gas, steam, or product gas, re-circulated after preheating. Retorts using steam or product gas usually have been indirectly heated as well.

When hot gases are introduced directly, the volatile products of carbonisation mix this heating medium, and additional provision is required in the gas-processing equipment for cooling this much larger volume and treating it to separate the tar. If steam is used as the heating medium, the treatment is quite simple. When the mixture of gases is cooled, the steam is condensed and may be separated readily from condensed tar by decantation. The presence of products of combustion in the final mixture of product gas from a retort obviously lowers its heating value materially, although its total heat content will not be altered. A lean gas is less desirable as a fuel than the higher heating-value gas, which would be produced without the dilution from products of combustion.
The charge to retorts carbonised by direct contact with hot gases must be sufficiently open to permit the passage of these gases, and briquettes or a lump coal is required. Examples of continuous vertical retorts are:

<table>
<thead>
<tr>
<th>Vertical retorts</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Indirectly &amp; Directly Heated</strong></td>
</tr>
<tr>
<td>Koppers continuous vertical ovens</td>
</tr>
<tr>
<td>Didier-Werke retorts</td>
</tr>
<tr>
<td>Rochdale process</td>
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4.1 **Koppers continuous vertical ovens**

Vertical chamber ovens, heated by the combustion of fuel gas in flues, were developed by H. Koppers A.-G. [2] in Germany for carbonising nut coal at low temperatures by a continuous process. The coal to be carbonised was fed in at the top and cooled char removed at the base. Heat generated by burning gas in flues of silica brick was transferred to the charge by conduction through the hot brick walls. The difference from usual coke-oven practice consisted in the continuous movement of the coal downward through the retort, rather than its intermittent charging and horizontal pushing at relatively long time intervals. Furthermore, the use of a noncaking coal was necessary to avoid the provision of mechanical means for removing the char from the retort. The introduction of large quantities of steam into the continuous oven below the carbonising zone was a third point of difference. The resulting water gas not only increased the gas yield, but also, in conjunction with the
uncomposed steam, served to augment the transfer of heat to the charge. The Koppers method, therefore, was a combination of external and internal heating.

Information is available on a plant that was under construction at the close of World War II at the Oheim mine of Berwerksverwaltung Oberschlesien, Katowice (Kattowitz). This plant had a capacity for carbonising 1,000,000 metric tons/yr of Upper Silesian, weakly caking bituminous coal.

**Figure 9** Diagrammatic drawing of a Koppers continuous, vertical, steaming oven
The arrangement of oven, flues, and regenerators (Figure 9 and 10) was somewhat similar to that of conventional, Koppers high-temperature coke ovens, with the banks of flues separating the oven chambers. The regenerators, however, were located beside the ovens, one above the other, instead of below them. Dimensions in feet of each oven at the Oheim plant were as follows: total chamber height, 37.7; length of chamber, 11.5; width of chamber at top, 0.8; width of chamber at a point 18.7 ft below the top, 1.1; and width of chamber at bottom, 1.1. The oven walls were lined with firebrick, but the heating flues were constructed of silica brick.

Figure 10  Cross section through Koppers continuous, vertical, steaming coke oven.
The fuel gas was burned in the flues, and the hot gases were passed alternately upward and downward through the flues and into the upper and lower regenerators. Automatic reversing equipment controlled the direction of flow of the gases. To maintain a uniform temperature over the entire height of an oven, additional heat could be obtained by use of auxiliary burners. The heat from these burners was directed into either the upper or lower regenerator as needed, thus raising its temperature and that of the air it preheated during the next reversal. Product gas from the ovens or producer gas generated from a portion of the char was used for under firing the ovens.

Above each unit of fifteen ovens was a coalbunker from which coal was dropped into the feed hopper on the top of each oven chamber. From the feed hopper, the coal was fed continuously into the carbonising chamber. The char was discharged into a bottom hopper, from which it was withdrawn periodically into a tilting cart for transportation to a special quenching station.

To precool the char entering the discharge chamber, a portion of the product gas, free of tar and light oils, was returned to this chamber at a point just above the char-discharge outlet. At the same time, water injected into the char was converted into steam, which mixed with the recirculated gas. In this passage through the char in the chamber, the steam formed water gas, which increased the volume of low-temperature carbonisation gas.

The gases flowed from each oven through two standpipes into the collector main and were conducted on through indirect coolers and then through a mechanical or electrical tar separator. The free ammonia was removed in the final cooler, from whence the gas passed to the light-oil adsorption equipment, and then to a gas holder.

When carbonising Upper Silesian, high-volatile, bituminous, nut coal containing 5% moisture and 7% ash to produce char with 3% volatile matter, results were as follows: coal throughput per oven per day, 25 metric tons; char yield (3% moisture) 64%; low-temperature tar yield, 9.2%; low-temperature light-oil yield, 0.8%; heat consumption per pound of coal,
990 Btu; gas yield per short ton (2000 lb) of coal, 10,330 standard cu ft, with a gross heating value per standard cubic foot of 470 Btu (equivalent to 2250 Btu per pound of coal). The combined tar and light-oil yield amounted to 87% of that found by the Fisher assay.

Prior to the installation at the Oheim mine, several plants of this type had been installed for other services, including calcining petroleum coke, carbonising ovoid briquettes or weakly caking high volatile bituminous coal at 700°C, and production of mixed gas (distillation gas plus water gas) from noncaking, high-volatile, bituminous nut coal.

Ceramic refectories were considered to have an advantage over iron as a construction material, because of the wider range in temperatures that was possible. The ceramic ovens were not limited to a maximum temperature of about 600°C, the maximum for iron. It was also claimed that recirculation of the gases raised the throughput of the ovens 50% above that of ovens that had no gas recirculation.

Several installations of the continuous Koppers ovens were utilized in Japan during World War II, and Reid has reported on their performance. The temperature in the vertical flues ranged from about 1000°C at the bottom to 850°C at the top, with 800°C as the estimated maximum temperature of the charge, which would classify the Koppers process as “medium-temperature” carbonisation. The large volume of steam, which was added below the carbonising zone was the only gas introduced into the retort. Because contact with the heated walls tended to decompose tar vapours, the yield of tar in the Koppers ovens would be lower than with internally heated processes carbonising at the same temperature, but by direct contact with the hot gases alone. These ovens produced a high yield of gas, which was usually employed as synthesis gas, the flues being heated with producer gas generated from a part of the char.

At the plant at Ube, Yamaguchi-ken, the Koppers ovens were arranged in two banks, each bank consisting of thirty-six ovens divided into six batteries of six ovens each. A battery was 26 ft long and was separated from the adjoining battery by a narrow, air-cooled chamber. The overall width of a bank was 25 ft, including the regenerators, and the height of the
carbonising zone was 31 ft. Each oven was 12 ft 9 in. long, and tapered in width from 10\(\frac{5}{8}\) in. at the top to 11\(\frac{3}{4}\) in. at the bottom, to facilitate descent of the charge. The volume of each oven was approximately 350 cu ft, so that about 0.001 ton of coal was carbonised per cubic foot of oven volume per hour. The daily rated capacity of the seventy-two ovens was 600 tons of coal, and 21 hr were required for passage of the coal through an oven. The regenerators were of conventional design and were reserved on a 30-min cycle. When rich coke-oven gas burned in the oven flues, only the air was preheated; but with the leaner producer gas, the air and gas passed through separate passages of the regenerators.

Steam at 300°C was introduced at the bottom of the carbonising zone. It amounted to about 360 tons/day, and about 144 tons were converted into water gas at the expense of about 175 tons of char containing an estimated 55%. The rate of heat transfer from the hot refractory walls to the charge of coal was increased by the addition of the steam, the quantity of product gas was increased, and the retort temperature was decreased to from 700 to 850°C, although the temperature in the flues might be as high as 1000°C. About 800 to 1200 Btu per pound of coal was required for carbonisation and the water-gas reaction.

The tar recovery and by-product-processing systems were similar to those used in coke-oven practice. Dust in the off take gas was not particularly troublesome and was removed along with the tar by passing the gas through the usual coolers and centrifugal tar separators.

The formation of clinker in the lower sections of the ovens, and the development of cracks in the walls, permitting leakage of air into the oven chambers and the combustion of part of the char, were the two principal difficulties encountered in operating the plant. The clinker gradually accumulated on the face of the refractory, interfering with heat transfer and allowing its temperature to increase to 900 to 1000°C. The masses of clinker eventually became so thick that daily poking was required to maintain a continuous downward flow of char. Another difficulty was the clogging of the regenerators with dust, which was entrained in the dirty producer gas, so that cleaning at 3-year intervals became necessary. The presence of 660 grains of hydrogen sulphide per 100 cu ft of product gas caused severe corrosion in the off take and main gas lines. In operation, it was possible to maintain the rated capacity of
the retorts, approximately 600 tons of coal per day, only for short periods. At an average rate of about 480 tons of coal per day, continuous operation of the plant was possible.

4.2 Didier-Werke retorts

Continuous, ceramic, vertical chamber ovens were used for the low-temperature carbonisation of noncaking coal in a commercial plant erected at Didier-Werke [2,18,19] in Berlin. It consisted of twenty-four chambers and was designed to carbonise 300,000 metric tons of noncaking coal per year. The ovens resembled Woodall-Duckham, continuous vertical retorts, although the heating arrangement was modified to adapt them to low-temperature carbonisation. Recirculated, tar-free, product gas was introduced into each oven above the char extractor. By recirculating a larger or smaller volume of this gas, the velocities of gases and vapours expelled from the coal could be regulated to control within wide limits the length of time during which vapours and gases remained inside the chamber, and thus to adjust tar quality and yields. Sections through Didier retorts are shown in Figure 11.
This oven was not considered satisfactory for a caking coal. To carbonise coal evenly within a reasonable time, a narrow oven chamber was required—about 8 in. in width. Because the contraction of low-temperature char is less than that of high-temperature coke, the friction between the wall faces and the surface of the char mass formed from a caking coal would be too great to be overcome by the weight of the charge.

The low-temperature carbonisation step was only one part of a three-stage process, which yielded briquetted chars. The process comprised:
1. Carbonisation of a noncaking coal of less than 2-in. size at 550 to 650°C to remove the tar in the continuous, vertical chamber ovens;
2. crushing and briquetting the crushed char in a roll press, after addition of 5 to 8% of pitch (produced by distillation of the tar) as binder, together with 10 to 15% of a coking coal; and
3. carbonisation of the briquettes in either continuous or intermittent vertical chamber ovens at 900 to 1000°C. These second higher-temperature ovens were similar to those normally used in European gasworks practice, except for recirculation of clean make-gas up through the charge. Briquettes suitable for domestic or metallurgical use could be produced, depending on the conditions of operation. In blast-furnace trials, briquettes produced from lignite chars were found to be entirely suitable as fuel.

4.3 Rochdale process

The use of heated gases to carry the heat rapidly through the charge was the basis of the Rochdale process [20], which was developed by the British North Western Gas Board. By introduction of the gas, the coal throughput of gas-making, continuous, vertical retorts, which could otherwise be reduced, was reported to have been increased by as much as 100% above the nominal figure for which the retorts were originally designed. In normal operation of continuous, vertical retorts, steam is admitted at the base to reclaim the heat from the hot coke before it is discharged. The steam then reacts with the coke higher up in the retort to produce carbon monoxide and hydrogen, which combines with the gases evolved from the coal and increases the volume of gas produced. In the Rochdale process, the steam was replaced by gas, either blue water gas or producer gas, both of which have been successfully applied. Because these gases do not react with the char to any extent, all the available heat from the char is transferred to the coal charge for carbonisation.

The Rochdale process was used to produce a highly reactive fuel of domestic grates. A coal of low or moderate swelling index in small nut sizes was carbonised in continuous vertical retorts at temperatures below the normal temperatures of gas-making retorts. Into the base of the retorts, blue water gas was injected in amounts of 18,000 to 18,500 cu ft per ton of coal
carbonised. Temperatures in the combustion chambers were usually maintained around 890°C, which was probably equivalent to a 700°C maximum temperature in the charge. Although carbonised at a low temperature, the volatile matter of the char was low, about 3 to 5%. The heating value was about the same as that of normal coke, but the bulk density was lower, about 17lb/cu ft, instead of the 19lb/cu ft with ordinary gas coke. The char ignited easily and burned completely in the open grate. In appearance it was dull, without the silvery sheen of gas coke. The price of the low-temperature char was expected to command a premium price.

4.4 Lurgi-Spulgas retort

In the Lurgi-Spulgas retort [2,3,6,21], coal, usually in the form of nuts or briquettes, is dried and then carbonised as it continuously descends through the retort in direct counter-current contact with ascending heated combustion gases. A retort is divided into two chambers, an upper chamber for drying, and a lower one in which the coal is carbonised by contact with gases at about 600 to 700°C. These two chambers are connected by several vertical pipes, through which the coal descends from the upper chamber to the lower. The hot gases used for drying and for carbonising are produced by burning gas in two separate external combustion chambers, and the products of combustion from these chambers are introduced into the dryer and the carboniser, respectively. The hot char is finally cooled by direct contact with cool product gas before it is withdrawn at the bottom of the carboniser. Uniform distribution of incoming coal is necessary, and a distributing mechanism is provided for this purpose at the top of the retort. Many Lurgi-Spulgas plants were built in Germany shortly before World War II to obtain from brown coal the tar from which motor fuel can be made.
A flow sheet of a Lurgi-Spulgas plant at Offleben in Germany is shown in Figure 12. In this plant, raw brown coal, containing about 54% moisture, was transported from the open mine to a storage and feed bunker, and from thence to a crushing plant. There, by means of roller screens and “Titan” breakers, the coal was reduced in size to 0 by 20mm. Before raw coal
was introduced into the Lurgi-Spulgas retorts, it was dried in a separate dryer to a moisture content of about 15% by contact with combustion gases produced by burning a portion of the product gas. The dried coal was then briquetted in a press without the use of a binder, and the briquettes were elevated to the top of the low-temperature retorts, of which there were 10, each with a capacity of 300 tons of briquettes per day, and dropped into the upper drying section.

In the top zone of the Lurgi carboniser, the briquettes were dried to 0.5% moisture or less, with the drying gases entering at about 220°C and leaving at 60 to 80°C. As drying of the briquettes progressed, they disintegrated into dry granular pieces which descended by gravity through the large down-pipes into the lower section, which was about 10 ft in diameter and 20 ft high. Here carbonisation took place the presence of oxygen-free combustion gases having a temperature of 600 to 700°C. The maximum temperature reached by the briquettes was 500°C. Granular char was discharged from the carboniser through a lock and cooling was completed by contact with water and air in a revolving cylinder before transporting it to the loading bunkers.

The mixture of combustion gases and volatile products leaving the carboniser passed first through a scrubber, in which dust and heavy tar were removed, and then through an electrical precipitator to separate light tar. The tar-free gases from the electrical precipitator were cooled in indirect coolers to condense middle oil and were scrubbed with an absorbent oil to recover light oil. The heating gases from the two chambers were kept separate and were recirculated back to the combustion chambers. In one plant, sulphur was removed from the hot gases circulated through the dryer. Yields of products expected per ton of dried briquettes prepared from a normal brown coal were: char, 45% by weight; tar, 12.5% by weight; and gas, 4600 cu ft with a heating value of 225 Btu/cu ft.

The char from the retorts contained from 10 to 12% volatile matter and up to 20% ash. It was very reactive, and at the Bohlen plant was cooled in nitrogen to avoid ignition. The size consists of the char product varied with the briquetting method used. Char from ordinary plunger presses usually contained about 20% breeze, 50%6 to 20 mm size, and 30% over 20
mm. Char from briquettes produced in ring roll presses contained only about 5 to 8% below 6 mm in size. The char was screened into suitable sizes for industrial and domestic purposes. The fine material was used in boiler plants and in gasification process, the middle size mainly in gas producers, and the large size as a domestic fuel and in central heating plants. This char was reported to be extremely reactive after aging, so that it required special precautions to avoid spontaneous ignition and explosions.

A Lurgi-Spulgas carboniser was installed at Lehigh in Stark Country, North Dakota, for carbonising lignite, which was mined there, and data on this operation have been published. As mined, this lignite contains about 40.3% moisture, 6.7% ash, and 0.78% sulphur.

The lignite was first crushed, and the +5/8-in. size was dried in the drying section of the Lurgi unit by direct contact with the products of combustion from burning a portion of the gas produced in the carboniser. Although the temperature of these combustion products was approximately 250°C, the temperature of the lignite was much lower about the boiling point of water—because of its high-moisture content. Under the conditions in the dryer, the lignite disintegrated, and the material descending to the carboniser ranged from about 0.25 to 0.75” in. size. The dried lignite gravitated through eight pipes, each about 16in. in diameter and 5 ft long, which were arranged to provide a seal against the upward flow of volatile products. Upward leakage was hindered by maintaining the pressure of the gas in the carboniser at slightly less than that in the dryer.

Products of combustion at about 700°C from burning additional fuel gas were circulated through the carboniser to heat the lignite in its descent to about 590°C and to effect carbonisation. In the lower section carboniser, before the char was discharged, it was cooled by recirculating cool product gas through it. Volatile products were withdrawn from the top of the carbonising chamber and were cooled and condensed for recovery of the tar, which was distilled into creosote and a pitch binder.

The finished char was briquetted with pitch, from the tar produced, or with asphalt. About 100 tons of briquettes were obtained from 250 tons of raw lignite. The briquettes were of the
pillow type, $2^{\frac{1}{8}}$" square by $1^{\frac{5}{16}}$" thick, with a weight of 3.5 oz. Until the briquettes had cooled and hardened, they were fragile and required gentle handling. Although the char itself was smokeless, the binder caused a little smoke when the briquettes were burned.

The Lurgi-Spulgas system has also been used successfully in several Japanese plants, particularly as a source of liquid fuels during World War II. At the Wakamatsu Works of Nissa Ekitai Neuryo K.K. four carbonisers were installed, each with a design capacity for carbonising 300 tons of Takamatsu coal per day. On the basis of the heat in the coal changed to the carboniser, 65.6% appeared as heat of combustion in the char, 16.1% in the liquid products, and 5.1% in the excess gas, giving a total thermal efficiency of 86.8%. The heat losses-13.2%-included 0.5% in the waste gas, 0.2% as sensible heat in the char, 8.5% as radiation and other unmeasured losses, and 4% as sensible heat in the gas leaving the carboniser.

According to Reid, the plant was shut down for inspection after the first 13 months of continuous operation, and the internal condition of the carboniser and the various units in the byproduct-recovery system were observed. Severe spalling of the brick lining in the upper part of the dryer section had occurred, probably because of attack on the mortar by sulphurous and sulphuric acids. Moisture had penetrated and caused swelling, with the result that the bricks originally 4.53 in. thick were reduced by spalling to only 2.56 in. In the lower part of the dryer, temperatures were higher and so such damage had occurred. The inside of the steel shell of the dryer was badly corroded, and the cast-iron main connecting with the blower for circulating gas also was seriously corroded. The steel duct making up the gas circulation system was perforated and required replacement. Although sulphur has usually been removed from the gas used for heating in the dryer zone, this precaution had not been taken with this unit, and the only protection for the steel work was the application of an acid-resisting paint. The paint film was inadequate and corrosion had occurred.

Since start of operation in 1941, this plant carbonised metric tons of Takamatsu coal to produce 396,529 tons of char, and 16,872,000 gal of total liquid fuels-divided into 1,977,000 gal motor gasoline, 1,243,000 gal semi-diesel oil, and 13,652,000 gal diesel fuel.
oil. These yields represent production by three retorts only, because one retort was kept idle as a spare. The coal may be classed as a high-volatile C bituminous coal. The caking tendency was slight, only 1.5 to 2.0, as measured by the Campredon-Gray method. The output of the carboniser was determined by the caking power of the coal used, because this property affected the pressure drop through the carbonisation zone and consequently the mass flow of hot gases. Strongly caking coals could not be used because of the formation of lumps of char, which interfered with the flow of the charge by gravity through the carboniser and upset uniform gas flow conditions. Non-caking coals could not be used either with complete satisfaction because of size degradation, the excessive volume of fines blocking the flow of carbonising gases. For instance, tests made with coal from the Ube field, having a caking index of zero, showed a pressure drop through the carboniser of 3.15-in.water gauge, and the maximum input obtainable was 250 tons of coal per day. With Takamatsu coal having a caking index of 1.5 to 2.0, the pressure drop was 1.57 to 1.97 in. water gauge, and a throughput of 350 tons per day could be maintained. The char had adequate strength for the uses to which it was put, principally as a fuel for portable gas producers on automotive vehicles. Its high reactivity made it a fair substitute for charcoal in this application.

Joh discussed the manufacture in Lurgi carbonisers of char for addition to the coal mix charged into coke ovens. Char containing from 18 to 20 % volatile matter was the most suitable as a constituent in blends to be used for blast-furnace coke, and was produced in a Lurgi carboniser operating at 500°C.

Production of char in two Lurgi retorts in New Zealand has been described by Jones. Each of these retorts, which were erected in 1931, had a daily normal throughput of 140 tons of the fine sizes of Waikato coal, a subbituminous coal of early Tertiary age. The char was screened and the +\(\frac{3}{8}\) in. size was sold as fuel for slow-combustion stoves. The smaller sizes were briquetted to form 2\(\frac{1}{2}\) oz ovoids, using either coal-tar pitch or bitumen as binder. These were popular for use as a domestic fuel and in bakers’ ovens. A portion, to which 5 to 10% coking coal was added, was used for locomotive fuel. The briquettes possessed a high reactivity and retained their shape in the fire. After being properly ignited, they were hard and coherent. In the briquetting operation, the char, usually at about 80 °C was sprayed with the binder, and
the mixture was then heated in a fluxer with saturated steam and sent to the briquetting press. The briquettes from the press were cooled by water sprays for 3 min on a wire-mesh belt, and cooling was completed by the briquettes standing overnight in bins.

The time of passage through the retort was approximately 20 hr, including 5 hr in the dryer and 5 hr in the carboniser. The char was discharged from the retort at approximately 200 °C onto a rubber belt on which it was cooled with water sprays. The amount of fines (-\frac{1}{8}\text{-in.}) in the coal exerted an important influence on the operation of the retorts because of its effect on the flow of hot gas. The most important factor in carbonisation was the volume of hot gas circulated through the carboniser, so long as its temperature was above 600 °C, because without sufficient gas the coal could not be carbonised. Moreover, too large a proportion of fines limited the cooling of the char in the lowest section of the retort. With 40% -\frac{1}{8}\text{-in.} coal in the feed, the maximum throughput of a retort was only 118 tons of coal per day, but this increased to 151 tons when the proportion of fines dropped to 20%.

All possible precautions were required in the design of the coal handling and charging equipment in order to maintain a uniform mixture of fines and the coarser particles of the cola. When separation occurred, the hot gases followed the path of least resistance, and the finer sizes of coal were imperfectly carbonised. Even with a uniform mix, differences in the volatile contents of the different sizes of char occurred. In one instance an increase from 7.9% in the +\frac{1}{8}\text{-in.} size to 10.2% in the -\frac{1}{16}\text{-in.} material was observed.

Retorts required periodic shutdowns for overhauling and cleaning. With ample coal and labour, each retort was expected to average about 9\frac{1}{2}-mon operation each year. Regular cleaning of the different parts of the retorts was especially important when carbonising coal containing a large proportion of fines, and about 5 hr a week were lost in cleaning. After a 10-mon continuous campaign, the thorough cleaning of a shutdown retort required four men for about a month. From 9 to 10 days were consumed in shutting down a Lurgi retort, because gradual cooling by recirculating an inert gas was necessary. Starting up took 6 to 9 days. After a temporary shutdown due to mechanical troubles, the retort could be restarted.
immediately, if the shutdown had lasted for only about 12 hr or less. With shutdowns lasting for more than 16 hr, additional gas had to be provided to reheat the retort.

Although Jones found that a Lurgi-Spulgas retort was limited to noncaking coals, such as subbituminous or brown coals, the addition of a small proportion of coking coal to the subbituminous coal was suggested as a means for increasing the size of the char. Several experiments in the New Zealand plant yielded char in larger pieces, although it was less strong than the carbonised coal briquettes.

In comparative tests on the low-temperature carbonisation of some poorly coking to noncaking Indian (Raniganj) coals, the yields of products obtained by the Gray-King assay, the Fisher assay, and in a Lurgi-Spulgas plant, with a capacity of 25 tons of coal per day, were measured by Das Gupta and co-workers. In the assays, the coke yield ranged from 1250 to 1500 lb per ton of dry coal, and the tar from 16 to 31 gal. From the Lurgi-Spulgas retort, when operated at 650 to 750°C, the yield of tar was about 70 to 80% of that produced in the Fisher assay at 550°C. The char contained 5 to 7% volatile matter.

The Kollergas process resembled the Lurgi-Spulgas process. Hard coal was carbonised continuously by recirculating hot gas produced in the process up through the charge, according to Thau. The char in the lower part of the retort was cooled before discharge by means of additional clean product gas. In a plant installed in Germany during 1938, a nut coal of 10 to 20-or 20 to 40-mm size was carbonised to produce 71 to 74% char and 6 to 9% tar. The yield of light oil was 0.9 to 1.2% of the coal. The char contained 6 to 7% volatile matter.

### 4.5 National Fuels process

The process of the National Fuels Corporation [5,6,15,22-24] was adapted to the production of char or coke in pieces of uniform size from either coking or noncaking coals. Before carbonisation a coking coal usually required an oxidation step in which the coal was brought in contact with an oxygen-containing gas at an elevated temperature to secure a uniform and
acceptable coking index. The oxidized coal was then briquetted with tar or pitch, usually that produced in the process, and the briquettes were coked. The process essentially comprised the five steps: (1) crushing or grinding the coal to pass a 30-mesh or finer screen, and drying to less than 2% moisture; (2) oxidizing, where necessary; (3) briquetting in a roll press, after fluxing with a suitable binder; (4) carbonizing, by direct contact with hot product gas; and (5) recovery and treatment of the volatile products.

Figure 13 is a flowsheet of the carbonization part of the process. The carbonizing operation was so designed that the carbonized briquettes were continuously dry cooled as they descended below the maximum temperature zone in the retort, with recovery of the bulk of the process heat. Cooling gas entered the bottom of the retort and, while ascending, simultaneously cooled the briquettes and recovered heat that was utilized higher up in the retort.

![Figure 13](image)

**Figure 13** Flow sheet for the National Fuels Process

Heated gas was introduced to the retort through tuyeres located at a level about onethird the height up from the bottom. Thgis gas, heated to the desired carbonizing temperature in an external heat exchanger, supplied the process “make-up” heat. The quench gas rising from
the bottom attained a temperature approximating the carbonizing temperature by the time it had reached the level of the tuyere gas inlet. From this level, the combined gases rose through the incoming charge, giving up much of their heat and leaving relatively cool from the top of the retort. As a consequence of the sweeping action of this progressively cooler gas, the volatile matter was removed with a minimum of secondary thermal decomposition. The combined gases were passed through a washer-cooler, followed by a Cottrell precipitator, and then a portion was reheated and recirculated to the retort. The balance, equivalent to the product gas, was continuously withdrawn from the circuit, subjected to further purification, and used as desired.

Preoxidation of the coal was a vital part of the process and made it possible to carbonize highly swelling coals by controlling the coking properties of the coal at a desirable level. Unless the coal was low in coking power and practically nonswelling, serious trouble from agglomeration occurred during carbonization. In the retort during carbonization, the briquettes were heated at the rate of 2 to 3°C/min while passing through the critical temperature range of approximately 350 to 500°C in which they softened. For an underoxidized coal, the particles oversoftened and swelled. The strength of such briquettes was insufficient to withstand the overhead load, and both deformation and sticking occurred.

For an overoxidized coal, on the other hand, the coking properties were almost completely destroyed. No agglomeration occurred within the briquettes and a product was obtained that was weak and impossible to handle without breakage. Although coals of a wide range of coking index from very low to very high are amenable to treatment by the N.F.C. process, in general, at least some coking property has been required. However, experience indicated that it could be so little as to make the coal wholly unacceptable in conventional coking practice.

The briquettes were formed under high pressure in the press, and the binder was required to give them sufficient strength to withstand charging into the retort and to sustain the overhead load until carbonization was complete. Dissolving of the coal by the binder at any point during passage of the briquettes through the critical temperature range in the retort was
undesirable. The tar from the process itself made a good binder, although other coal tars or pitches, as well as molasses and sulfite lye, have been used instead.

The carbonising temperature ranged from about 450°C to the temperatures of high-temperature coking, depending on the product desired. Use of the lowest temperature has produced a highly reactive fuel, attractive for domestic consumption—the highest being a coke of less than 1% volatile matter, suitable for metallurgical use. The carbonized briquettes were uniform size, with a weight of about 42 to 47 lb/cu ft, and an apparent specific gravity of about 1.25. The briquettes were free-burning and highly reactive. Long periods of storage in the open showed no appreciable deterioration.

The N.F.C. process was developed in the 1920’s by the National Fuels Corporation, then a subsidiary of Calco Chemical Co., Inc. It was tried in a 5-to 10-ton/day pilot plant located in New Haven, Connecticut, and later in a 100-ton/day plant located in Bethlehem, Pennsylvania. In addition, study was devoted to the N.F.C. process in Utah in connection with the extensive research program sponsored by the Utah Conservation and Research Foundation.

Wolf modified the coal tar or pitches binders for use in the N.F.C. process by addition of sulfuric acid and combining 5 to 10% of the mixture with the oxidized coal. In another patent, he treated the coal with about 1 to 2% of an inorganic oxidizing acid at temperature of 90 to 250°C for at least 5 min to reduce the swelling and caking properties of the coal. Sulfuric acid was suitable and could be added in combination with oxidizing salts.

The North Western Gas Board proposed the preparation of briquettes by a process similar to N.F.C. process. Noncoking slack was blended approximately 90% with a higher rank coal and the mixture was then briquetted with pitch. To prevent swelling of the briquettes during carbonization with loss of shape, preoxidation of the coal was necessary, and the addition of an inorganic oxidant to the coal before briquetting was considered in an effort to reduce the cost of oxidation below that of the conventional air-oxidation step. Oxygen should be evolved by the oxidant during or just before the plastic stage is reached, about 400 to 430°C.
In carbonizing 2-in. diameter briquettes made from Yalourn brown coal in Australia, trouble was experienced because of their cracking, even at a low heating rate. It was found that when a briquette was carbonized by indirect heating through the wall of the retort, the temperature of one side was raised faster than was that of the other side—a condition which increased the tendency to crack. However, by heating the retort internally by the circulation through it of gaseous products of combustion, it was possible to dry and carbonize briquettes at temperatures up to 700°C or higher—even to 900°C—without cracking, owing to the more uniform temperature conditions to which they were exposed. At higher temperatures, reaction of carbon dioxide and water vapor with the briquettes occurred.

To produce crack-free briquettes from the brown-coal char, the heating rate should be slow and carefully controlled through both the drying and distillation stages up to 600°C. Drying required not less than 5 hr, and the temperature during distillation should not be increased faster than 100°C/hr, and possibly more slowly in the case of large briquettes. Above 600°C, more rapid heating was possible. By observing these conditions, briquettes in sizes to 5 in. by 2\(\frac{1}{2}\) in by 1\(\frac{3}{4}\) in. were successfully carbonised without formation of serious major cracks and fissures.

Another treatment proposed for use in the preparation of briquettes from the Yallourn coal was to preshrink the coal particles completely before briquetting and carbonizing. This was done by precarbonizing the coal to 800°C, briquetting, and recarbonizing the briquettes at 500°C. By the preliminary carbonization to a temperature in the range 600 to 800°C, the char was adequately preshrunk, and the particles did not pull away from the pitch binder during the final carbonization. The objective in the work was the preparation of fuels suitable for foundries and other metallurgical uses.

### 4.6 Pintsch-Weber process

A carbonization process for the brown-coal briquettes or bituminous, noncaking, lump coal (free from a large proportion of fines) being fed to the chain grate stokers of a boiler
installation, was developed by Julius Pintsch [2] Kommandit Gesellschaft in Berlin before World War II. The coal was carbonized continuously as it descended through a charging hopper located above the grate by drawing through it hot combustion gases. Hot char was delivered from the bottom of the hopper onto the grate, and the gases withdrawn from the top of the hopper were returned and burned under the boiler after the tar which they contained had been recovered. The process was of such interest that the German technical-military authorities during the war attempted to have it installed on all boilers fired with bituminous coal or brown-coal briquettes to recover tar for use as fuel oil.

In operation, a coal containing a large proportion of fines was briquetted with a binder in a roll press. The briquettes were then conveyed by a slow-moving elevator provided with broad, perforated buckets to the feed bunker of the stoker. Hot gases introduced at the bottom of the elevator housing passed up around the briquettes and hardened them as they ascended to the bunker. From the bunker, the briquettes slid down into the carbonising shaft and were converted to char before discharge onto the chain grate.

Pieters described a unit which consisted of a multiplicity of internally heated vertical retorts grouped within a casing and operated with continuous production of char. The coal was first dried and then carbonised by contact with superheated steam introduced at the bottom of each retort. In the lower part of the retort with increasing temperature the coal was reported to be desulfurized by the action of nascent hydrogen which was formed from steam in contact with the hot charge. Ash was removed in a dry-cleaning tower, and the resulting low-ash and low-sulfur char was briquetted and the briquettes calcined. The final briquettes were reported to form a highgrade metallurgical or domestic fuel.

### 4.7 Electrically heated retort

In Norway, a noncoking bituminous coal, in the form of briquettes, was carbonized by the Norsk Hydro-Electrisk Kvaefslotfaktieselskab on a pilot scale in a vertical shaft retort, heated by the use of electric power [8,25,26]. The Longyear City coal was mined in Spitsbergen, and, because it was frozen as mined, it disintegrated on thawing.
The coal was crushed, and either the pulverized coal was briquetted at a very high pressure of almost 36,000 psi, or larger size was briquetted by using a pressure of from 1400 to 2850 psi with sulfite lye as binder. The briquettes were preheated to 150 to 200°C and charged into the top of the shaft retort, Figure 14. In their slow descent through the shaft, the briquettes were partially carbonized by the heat in the gases ascending from below, so that they became conductive. An electric furnace, containing two sets of three-phase electrodes was located in the lower end of the shaft—the briquettes forming the resistor element to provide the process heat necessary for completion of carbonisation.

**Figure 14** Flow sheet of electric process for the carbonisation of non-coking bituminous coal

Below the electric furnace and above the discharge point, the briquettes were partially cooled by introduction of cool product gas. The temperature of the heated gas ascending from the cooling zone was raised further in the electric furnace to 500°C, and at that temperature some
of the constituents reacted with hot carbon in the descending charge to form carbon monoxide and hydrogen. The gases continued their ascent through the shaft and provided the heat for partial carbonization of the coal above the electric furnace.

The coke product was expected to possess both the properties of high-temperature coke with 1 to 2% volatile matter and a compressive strength up to 7000 psi and, in addition, to be as reactive as low-temperature char. On the basis of the first small installation, the power consumption of the process was reported to be 40 kwhr per 1000 cu ft of gas produced, which is equivalent to almost 1000 kwhr per ton of coal carbonized. A larger plant with a capacity of 25 tons of coal briquettes per day was projected; it was estimated that it could operate on 175 kWh per ton, and that a retort with a capacity of 125 tons per day would operate on 120 kWh per ton of coal.

4.8 Petit retort

In the Petit process [16,27], the coal was heated by indirect heat as it descended continuously in a solid stream through a vertical retort, the steel walls of which were shaped to provide a zigzag path. By this zigzag arrangement of the walls, the coal was retarded in its descent. To insure the continuous, uniform descent of the coal and to delay buildup of carbon on the walls, the coal was agitated and kept in suspension by vibrating the walls. This was accomplished by means of several vibrators which were located at intervals up the height of, but outside, the retort, and which were connected to the wall by rods extending through the brick setting. A flow diagram of a retort is shown in Figure 15.
The retort was mounted in a firebrick housing and was heated by gaseous products of combustion which ascended around it. Baffles located at suitable intervals in the retort setting deflected the hot gases against the walls. The volatile products were withdrawn from the retort near the bottom, and the char was removed at the bottom through a rotary valve. This retort was developed by Daniel Petit in France. A pilot plant having a rated capacity of 25 tons/day was installed by the Colorado Fuel and Iron Corporation at Pueblo Colorado, in 1953. Other installations of similar principle are reported to have been made in Europe.

4.9 P.D.P. retort

In the effort to develop an economical source of carbon for use as a reducing agent in various processing operations, including phosphorous manufacture and nonferrous metal smelting, several pilot plants for continuous carbonization of Western coals were erected and operated in Montana by P.D.P. Processing, Inc. The process, usually known as the “Koal Krudes” process, was studied in several pilot plants, one of the last, shown in Figure 16, being described by Berg and Atkinson. It consisted essentially of four vertical, concentric cylinders.

Figure 15 Cross section through Petit retort (After J.D. Price; Courtesy of Daniel Petit)
of stainless steel. They made up the heated portion of the retort and were surrounded by a mild steel shell with an outer diameter of 58 in. The inner cylinders were, respectively, 12, 20, 24, and 38 in. in diameter. The annular space between the shell and the outer stainless cylinder was packed with insulation.

The raw coal to be carbonized was fed from a hopper into the 2-in. space between the second and third stainless cylinders and descended by gravity. To assist in uniform descent of the coal without channeling, the 20-in. cylinder was slightly raised and dropped at short intervals by a mechanical lift. The coal descended through 34 in. of annular space before it passed into a funnel arrangement from which it was withdrawn by screw conveyors. Their speed could be varied to control the throughput of coal and the volatile matter of the char.

The volatile products were withdrawn from the retort through four vertical sets of louvres which bridged the outer heating annulus, and then were pulled by an exhauster through a heated manifold and a cyclone for dust removal. Tar, oils, and water were finally condensed by cooling the gas to a temperature of about 40°C.

![Figure 16](image)

**Figure 16**  Schematic diagram of pilot plant for Koal Krudes low-temperature carbonisation process
The heat for carbonization was generated by the combustion of fuel gas in a furnace below the retort, and the combustion gases were drawn up through the annular heating channels by a blower which recirculated them to the furnace. Flow of gases between the carbonizing annulus and those through which the combustion gases passed was prevented by water seals located at the upper and lower ends of the tube. The most important temperature was that of the char leaving the retort, which was held at from 700 to 880°C.

Following pilot-plant work, a single retort of similar design but of commercial size was constructed at Red Lodge, Montana. This retort was designed to carbonize washed Red Lodge slack coal at the rate of 40 tons/24hr. The outside dimensions of the retort were 24 ft in height and 9 ft in diameter. The char left the retort with a temperature of 590 to 650°C. About one-half ton of char and 18 to 20 gal of crude tar were produced per ton of coal. Because of financial difficulties, it is believed that work on this unit has been suspended.

4.10 Stansfield retort

The Stansfield retort [28] was under development for several years by the Research Council of Alberta to upgrade Alberta noncoking, subbituminous slack coal which is produced in that Canadian province. The objective was to carbonize the coal continuously at low temperatures with production of a char which, after briquetting with a suitable binder such as petroleum asphalt, would form a solid fuel possessing a reasonably high heating value and good handling, storage, and weathering properties. The use of a continuous vertical retort employing internal heating achieved by the combustion of the volatile matter distilled from the coal was proposed for this purpose. The slack coal contained 12 to 30% moisture and had a tendency to slake to fines. It was realized that operation with any large amount of fines or slack in a low-temperature carbonizer designed to operate with a deep bed on the principle of the gas producer could lead to irregular flow of gas and channeling. In turn, these conditions would result in uneven and incomplete carbonization of the coal, and in the production of gas of poor quality.
To avoid these difficulties, a pilot retort with a throughput of about 75 to 150 lb of coal per hour was designed on a different principle, Figure 17. The coal passed downward between two series of closely spaced baffles or refractory louvres in such a manner that it formed a continuous, unbroken thin layer alternately changing direction, but did not descend as a cascading stream as in cascade dryers. The retort had a rectangular cross section 16.75 by 24.25 in. o.d. and was 23 ft high. The baffle plates which were 9 by 6 by 0.75 in. were made of Carborundum, and were supported in slotted Carborundum plates at an angle of 60°. The total capacity of the retort from the top of the baffles to the outlet of the char cooler was 200 lb of coal. The retort consisted of three zones: an upper drying zone, a central carbonizing zone, and a lower equalizing zone, below which was a water-cooled discharge zone. The volatile matter distilled from the coal passed between the louvres to enter the flues which were located outside of the baffle system. Here it was burned with air which entered the flues through many small ports or tuyeres located at the level of the central carbonizing zone. With an effective carbonizing temperature of approximately 600°C in the carbonizing zone, a char with about 12% volatile matter was produced. The retort was operated at rates as high as 144 lb of coal per hour. At this rate, with an average flue-gas temperature of 500°C, the yield of char was 53%.
4.11 Archbald “Pipestem” retort

The Archbald “Pipestem” process [29] was reported to produce a hard, dense char containing 5 to 9% volatile matter, from a mixture of small sizes of bituminous and anthracite coals. A mixture of coking coal and inert material, such as anthracite, coke, or coal char, was wetted with tar, and the mixture was extruded at a pressure of less than 10 psi into vertical tubes installed in a furnace operating at temperatures between 650 and 760°C. Carbonization was continuous, and the time of passage of the mixture through the heated tube was reported to be between 13 and 15 min. The tubes were originally round, but in later work hexagonal tubes
with an inside diameter of 2.5 in. were used. The capacity of a tube was about 1500 lb of mixture per day.

The possible proportions of coking and inert coal varied widely with the volatile matter of coking coal. Two typical mixtures consisted of: one-third Pocahontas plus two-thirds anthracite; and 40% Clearfield coal containing 20% volatile matter plus 60% anthracite, respectively. The process thus resembles those in which an inert char is briquetted, and the briquettes carbonised. The coal was ground to pass a 10-mesh screen. The product from the Pipestem process had a density of 45 lb/cu ft, was nonfriable and free burning.

4.12 Geissen retort

In the Geissen process [2], brown coal was carbonized in a continuous, vertical retort containing a concentric revolving cylinder. Before carbonization, the brown coal was dried from its original moisture content of about 54% down to about 15%. The dried coal was then fed in at the top of the retort and carbonized as it descended between the inner revolving corrugated iron cylinder and an outer fixed wall consisting of a multiplicity of stationary annular collars. Revolution of the cylinder continually turned and mixed the coal.

The heat of carbonization was supplied by burning gas in a combustion chamber located inside the revolving cylinder, and then by conducting the products of combustion from the top of this cylinder down in contact with the corrugated wall. The volatile products of carbonization immediately escaped from the coal through openings between the annular collars and were withdrawn from the retort without the possibility of further cracking. The char was discharged continuously from the base of the retort.

The Geissen process was developed in Germany following World War I, and up to 1938 about thirty-five Geissen ovens had been installed, several of which were still operated during World War II. About 150 tons of brown coal with a moisture content of 54% could be carbonized per day in each retort.
4.13 Borsig-Geissen retort

The Borsig-Geissen retort [2], made by Borsig in 1935, was a modification of the Geissen retort. Its arrangement is illustrated in Figure 18. In place of the corrugated, revolving cylinder consisting of heavy iron castings, the Borsig-Geissen retort employed a stainless-steel tube for the rotating cylinder. The burner was situated at the top of this rotating cylinder, and the combustion gases simply flowed downward through it and then upward through a jacket surrounding the retort without contact with the coal. Partially dried brown coal introduced at the top of the retort passed downward between the rotating cylinder and the stationary collars, as in the Geissen oven. The process required that the coal charged should not be larger than 1/2in., with not over 10% of fines below 1/25-in.size. A portion of the make-gas was burned to supply the hot gases for carbonization.
4.14 Heliopore process

The Heliopore process [30] was designed to utilize the waste heat in the exhaust gas from an internal-combustion engine in carbonizing either noncoking coal or lignite. The gases distilled from the coal furnished the fuel for the internal-combustion engine, which in turn drove an electric generator. Thus the main products of a plant were expected to be electricity and char.

The principles of operation of the Heliopore process are illustrated by the flow diagram shown in Figure 19. The primary equipment consisted of a carbonizing retort, gas cooling
and scrubbing equipment, a gas storage holder, and an engine to burn the gaseous carbonization products. Coal was charged into the top of the retort while the hot exhaust gases from the engine traveled upward through special ducts in the retort walls countercurrent to the coal flow, but not in direct contact with the coal. Char was discharged at the bottom of the retort and the hot, volatile products at the top. The entire process was intended to be continuous and self-sustaining and to use coal as the only fuel. The process was tried out by the U.S. Bureau of Mines using a retort about 21 ft tall. The results showed that the engine exhaust gases were entirely inadequate as a source of carbonization heat. The tests of the Bureau of Mines showed that: even with the engine running at full load on natural gas, the temperature and volume of engine exhaust gases were such that the volume of carbonization gas produced from coal satisfied about 15 percent of the fuel requirements of the engine. In subsequent tests all the supplementary hot gas that could be produced with the available equipment and at the maximum temperature attainable were mixed with the engine exhaust gas, and the volume of carbonization gas produced was slightly less than enough to satisfy the fuel requirements of the engine at full load.

The maximum value that can be assigned to the heat available in the engine exhaust gas does not exceed 6 or 7 cents per hour. Consequently, the saving of this amount of heat could not justify any very appreciable investment in special equipment. Furthermore, the low-temperature of the engine exhaust gases (1300 to 1500 °F) militates against their efficient use, especially for carbonizing coal.
Figure 19  Flow diagram for Heliopore coal-carbonisation and power plant
5  Fluidised or entrained carbonisation

Carbonising fine coal while fluidised by or entrained in a stream of heated gas produces a finely divided char, which may be utilized for boiler firing. In addition, such chars are have found application in the preparation of metallurgical fuels-by replacement of low-volatile coal in the blends charged to coke ovens or by use in the production of briquettes which are subsequently carbonised at high temperatures.

A fluidised retort contains a bed of coal and char through which a gas is passed to maintain the solids in the fluidised state. Coal is added to the bed and char is withdrawn continuously. In an entrained process, on the other hand, the coal introduced into a stream of gas is carried through the retort into a separator.

Fluidised or entrained retorts are well adapted to the continuous processing of large tonnages of coal, and much useful experience on their design and operation is available to the industry from the large-scale applications of fluidised processing in other fields, such as in petroleum refining. For these reasons, fluidised processes for the carbonising of fine coals especially have attracted considerable interest, and much research is proceeding in laboratories throughout the world.

Fluidised and entrained processes are apparently applicable to practically all types of coals, ranging from the lignites or brown coals to bituminous coking coals. With the non-coking coals the process is relatively simple, involving carbonising the finely divided coal directly by contact with the heated gas. With coking coals, however, agglomeration occurs as the coal becomes plastic with increase in temperature, unless precautions are taken to avoid this difficulty. In most processes the coking property of such coals is destroyed by a preliminary oxidation of the coal, usually with heated air in another fluidised operation. However, other means have also been used to avoid agglomeration, including dilution of the coal mass with a large volume of inert, finished char before it is introduced into the carboniser.
The heat for carbonisation can be supplied in several ways. Although external heat has been used in several processes, internal heating in more is more common, either by use of preheated gases, by addition of hot char to the incoming coal, or by reaction of the oxygen in fluidising air with the carbonaceous solids or gaseous products in the retort. This last practice tends to dilute the gas produced with the inert products of combustion, thus materially lowering its heating value. Examples of fluidised processes are:

- United Engineers and Constructors process
- Singh process

### 5.1 United Engineers and Constructors process

The fluidized carbonization of coal for the production of a fine char suitable for boiler firing or other industrial use has been studied by United Engineers and Constructors in a pilot plant with a capacity of about 2 tons of coal per day over a period of several years. In this unit they reported that they successfully carbonised coals with a wide variety of characteristics, ranging from coking to noncoking. The pilot results led to the design of the process shown in the flow diagram, Figure 20.

This process consisted basically of three stages of coal treatment: a preoxidation, a carbonization, and a char-heating stage, each consisting of a fluidised bed. The coal, either coking or noncoking, was delivered into the plant, handled by conventional equipment, and crushed to pass a 1/8-in. mesh. The fine coal from the hammer mill was continuously fed into the oxidizer, which contained a fluidized bed of coal heated to about 200°C by means of hot, combustion products. This fluidizing gas contained some oxygen, the concentration so regulated that by a slight degree of oxidation of the coal its coking tendency was reduced sufficiently to prevent agglomeration in the subsequent carbonizing stage.

The hot coal was transported from the oxidizer through a pipeline at high velocity by means of recycled product gas from the carbonizer into this bed. Simultaneously into the same transport line was fed a controlled quantity of char which had been heated to about 650°C in
the char heater. About three parts of char were transported per volume of coal to secure the
desired carbonization temperature of 480°C.
Figure 20 Flow diagram of United Engineer's fluidized, low-temperature carbonization process.
Addition of this char also reduced any remaining tendency toward agglomeration of the coal in the carbonizer. From the carbonizer, char overflowed into the char heater where it was fluidized with preheated air. Approximately 3% of the char was burned, thus raising the temperature of the bed to about 650°C. The hot gases from the cyclone at the outlet of the char heater were heated further to about 980°C in a direct-fired heater and then introduced into the oxidizer. The carbonization products were withdrawn from the carbonizer, the make-char was withdrawn from the base through a cooler which reduced its temperature to about 200 to 260°C, and the volatile products-tar and gas-were withdrawn from the top, also through cyclones.

5.2 Singh process

The Singh process [31-33] also utilized three fluidized stages in series. There was an oxidation stage to reduce the agglomerating characteristics of the coal, followed by a carbonization stage. Heating of the charge in the carbonization vessel was accomplished by removal of a portion of the contents to a third bed wherein its temperature was raised by limited partial oxidation. The heated char was returned as a fluidized solid to heat the carbonization stage. A flowsheet of the process is shown in Figure 21. A pilot plant with a capacity of 2 tons of coal per hour was erected in Chicago. The char produced consisted of granular, hollow spheres, which, however, did not possess the rib and window construction of cenospheres. It was dustless and more easily pulverized than the original coal from which it was made.
An objective in the process was to provide a suitable substitute for low-volatile coal in the coal mixes for high-temperature coke ovens. Because serious segregation of char and coal in such a mix might result from differences in the bulk densities, an effort was made to produce char with a higher-bulk density in the pilot plant. Singh indicated that the bulk densities of the chars produced by various processes apparently averaged from about 25 to 28 lb/cu ft, but in tests with Elkhorn coal he increased the density to as much as 56.6 lb by addition of blast furnace flue dust to the mixture of coal and char charged to the retort. Typical process yields per ton of bituminous coal charged were given as 1320 lb char, 15 gal tar, 8200 cu ft gas (580 Btu/cu ft) and 35 lb solid sulfur. The heating value of the char, 12.180 Btu/lb, was higher than that of the raw coal, 11.460 Btu/lb.

Singh also patented a process for producing metallurgical coke from a coking coal, which incorporated a fluidised low-temperature carbonization step. The finely-divided coal was mixed with about 0.5 to 20.0% of iron oxide, and the mixture was carbonized by direct contact with a stream of hot gases, which raised the temperature to at least 450°C. An unagglomerated char was produced, but it contained entrapped particles of oxide. Additional amounts of a coking coal were then added, and the mixture was coked at high temperature.
5.3 Other fluidised processes

Several other fluidised bed processes have been developed [34-36]. The Southern Research Institute has pilot planted a process for carbonizing bituminous coal using a single fluidized reactor. Their research project began in 1952 when Alabama Power Company became interested in low-temperature carbonisation as a means for reducing their cost of fuel. The fluidized method that was developed reportedly handles satisfactorily coals with strongly coking or agglomerating properties.

In the SRI pilot plant (Figure 22), agglomeration of the raw coal was avoided by mixing it, before it entered the carbonizer, with at least 10 parts of hot char which had been recirculated from the top of the carboniser. The two materials were entrained together in a stream of high-velocity air which conveyed them upward into the carbonizer. The addition of this hot char was important because it acted as an inert diluent and prevented agglomeration.

![Figure 22](image)

**Figure 22** Southern Research Institute fluidised carbonisation process, pilot plant

The heat for carbonization was also produced in the same operation because the air burned a small portion of the hot char to raise the temperature of the mixture to the desired carbonization temperature—approximately 510°C. The velocity of the gases in the riserwhere
combustion took place was about 10 ft/sec. Temperature variations in the lines and carbonizer were at a minimum due to the high rate of recycle. Combustion was very rapid, taking place in less than a second while the mixture ascended to the carbonizer. The volume of air admitted was regulated to supply all the heat for carbonization. With America Seam coal, 400 Btu were required for carbonization, so that about 5 cu ft of air were fed to the carbonizer per pound of coal.

The superficial velocity of the gases in the carbonizer was approximately 1.1 ft/sec. Although tests indicated that a retention time of less than 10 min was sufficient for carbonizing the coal, the coal was retained in the carbonizer for approximately 20 min. no significant reduction in the retention time was possible without entraining the solids in the increased gas stream from the carbonizer. The unit was operated at a feed rate of 50 lb of coal per hour cubic foot of fluidized bed volume, with maximum sustained rate of 81 lb per hour per cubic foot of volume. A stream of finished char overflowed continuously from the top of the fluidized bed to a hopper. The volatile products were withdrawn from the top of the carbonizer and passed through two dust cyclones in series to an electrostatic precipitator. A pitch separator and the tar condensers followed.

Products from the America Seam coal were approximately 80% char, 10% tar, 5.3% aqueous condensate, and 4.7% gas. Because its volatile content was over 15% the char should form a satisfactory fuel for underfiring conventional boilers. The particles possessed a spherical, puffed shape, were friable, and the mesh size was slightly larger than that of the original coal. The gas amounted to 13,000 to 15,000 cu ft per ton of coal, with a low heating value only 120 to 140 Btu/cu ft. about 21.7 gal of tar were produced per ton of coal.

A fluidized process developed by Pittsburgh Consolidation Coal Company was disclosed in connection with the announcement of plans by Olin Mathieson to build an aluminum plant in the Ohio River valley. The power was to be generated by combustion of the char and gas which were to be produced in a low-temperature carbonization plant to be built by Pitt-Conso. The process comprised crushing the local coal to –10mesh and feeding it into a fluidized bed which was maintained at a temperature of 480 to 510°C in a reactor operated at
atmospheric pressure. The gaseous products were withdrawn from the top and the char near the bottom of the reactor. The gases were cooled and the greater portion was condensed as tar, but the balance, together with the char, were to be delivered to the boiler house. About 30% of the coal was volatilized. The plant was to have a capacity for carbonizing about 5000 tons of coal a day. This low-temperature carbonisation plant has not been installed. Pittsburgh Consolidation Coal Company also proposed to use the fluidized carbonization process as a primary step in a plant for the manufacture of synthetic fuels. By gasifying the char, Fisher-Tropsch gas could be produced which would be converted into the synthetic oils.

The three-stage process of Welinsky for carbonizing agglomeration coals included a preheating zone, a carbonizing zone, and combustion zone. The hot char from the carbonizer was partially recycled to the preheating zone to prevent agglomeration of the raw coal introduced there. Heat for the process was obtained by partial burning of another portion in the combustion zone and recycling this highly heated char to the carboniser. The temperature in the carbonizer was held below 760°C.

In an investigation of low-temperature carbonization in a fluidised bed, which was described by Peytavy and Foch, the primary aim was to manufacture a char from Lorraine coals to replace the coke breeze, which was normally added to the coal blends charged into tamped coke ovens in the Saar-Lorraine district. The retort was internally heated by partial combustion of the coal with the fluidizing air. By provision of a preheater to preheat both the coal and air, the rate of throughput of the unit was increased, the quality of the gas improved, and, in addition, the proportion of coal burned in the carbonizer was reduced to approximately 3%. The investigation was made in several pilot units capacities ranging up to 1 ton/hr. The fluidized process with air was selected as the most promising process after other methods of carbonization were studied. The char produced from the high-volatile, weakly-caking coals was made up of swollen particles which had been subjected to fusion. The char was relatively friable, but very uniform in quality for a given set of conditions. The furnace temperature determined its volatile matter content very accurately—within about 0.5%—as shown by Figure 23.
Chars prepared at temperatures of 500 to 800°C were so closely alike in appearance as to be indistinguishable. Char produced in a rotary furnace was very different, however, from that produced in the fluidized bed. In the former, the coal particles had maintained their shape and structure, but the color of the char was brown. It appeared that the rate of heating in the different retorts was more important than temperature in producing the differences in shape and structure. Carbonization time in the rotary retort had been about 30 min as compared to about 1 min in the fluidized bed.

Nathan and co-workers patented a fluidized process in which the finely divided coal was first dried and preheated in a dense-phase, fluidized dryer. The mixture passed downward as a confined stream through a carbonization zone into a preheating zone in which it was further preheated in a dense-phase fluidized bed by partial combustion with air or oxygen until the agglomerating tendency of the coal had been reduced. The preheated solids, together with the combustion gases, then ascended from the pretreater through a grid into the carbonization zone which contained a third dense-phase bed of char. It was maintained at a temperature above that in the pretreater by further partial combustion of the coal. Carbonization was completed in this third zone.
Coal was carbonised in a process in which hot char withdrawn from the carbonizer was added to a fluidised bed of fresh coal to dry it, according to a patent by Nelson. Not only was the coal dried, but its agglomerating tendency was eliminated by the addition of the char which entered at a temperature of 430 to 650°C. After from about 0.5 to 60 min in the fluidized bed, the mixture was transferred to the carbonizer. Nelson overcome the agglomerating tendency of certain coals during carbonization in a similar manner by injecting the particles into a turbulent suspension of hot coke or char. In another patent, Nelson and Martin supplied the heat for carbonization of the coal in a lower temperature range of about 430 to 480°C by partial combustion of the char from the carbonizer. The char was heated in a separate bed was returned to the carbonizer. Practically all the oxygen in the gas used to fluidize the char in the heating bed was consumed in the process.

To render the fresh particles of a caking coal noncaking without substantial carbonization, Garbo coated the tacky surfaces of the particles with soot while fluidizing them in a dense-phase bed at a temperature of at least 320°C.

According to Creelman, the char fines produced in fluidized carbonization may be completely burned, and the products of combustion may be returned to the fluidized bed as a source of heat for the low-temperature carbonization of the coal. The fines were recovered by separating them from the volatile products with which they left the retort. Creelman also patented a process in which a portion of the char product was burned, and the hot combustion gases supplied the heat for carbonization of the coal.

A low-temperature carbonization process involving what the inventor termed “thermal solution” was patented by Pevere and co-workers. The coal was mixed with a hydrocarbon solvent at 290 to 370°C under sufficient pressure to keep the solvent liquid. The mixture was then atomized into an inert gas stream and passed into a carbonising zone heated externally to 480 to 700°C where it remained until carbonisation was complete.

The use of atomic energy to supply the heat needed for low-temperature carbonization has been suggested. The purified gas from the retorts would be preheated by recycling it through a
nuclear reactor, and the heated gas would then be recirculated back through the fluidized carbonizer. Because the final product gas would be undiluted with products of combustion, its heating value would be high, about 800 Btu/cu ft.

5.4 Parry process

The major application of entrained carbonisation in the United States is the Parry process [16,34,37-39]. The Parry process has been the major application of entrained carbonization in the United States. Finely divided coal entrained in air was carbonized in an externally heated retort to produce a fine char, suitable for boiler firing. The process consisted essentially of moving the dried coal with air into the base of an externally heated vertical reactor. Heat was transferred from the wall of the reactor to the suspended mixture, but the major portion of the heat required for carbonization was produced by combustion of a portion of the coal with the air. All products of reaction, including char, tar vapors, and gas left the top of the reactor, and the separation of the solid and gaseous products was made in cyclones and the subsequent gas-processing equipment.

The Parry process was developed by the Bureau of Mines at Denver, in cooperation with the Texas Power and Light Company. As result of their work, a prototype carboniser for processing lignite was erected at Rockdale, Texas, in connection with the installation made by the Texas Power and Light Company to supply power to the aluminum pot lines, which the Aluminum Company of America was installing at the locality. The boiler installation there was designed for combustion of either dried lignite char; the latter to be utilized after the carbonisation process was perfected. The prototype carboniser began operation during 1953 and was operated to develop and improve the process and to produce crude tar for market evaluation. No additional carbonisers have been erected as yet.

The method of drying and carbonising the lignite has been described by Parry. The crushed raw lignite is moved from the storage silos by a closed loop conveyor which maintains an oversupply of lignite at the small hopper above the screw feed in the drier. Products of combustion at about 816°C produced by combustion of dried lignite dust with air and
recirculated gases enter the drier section at a pressure of about ¾ p.s.i.g. The wet lignite is suspended in this drying column in a turbulent state. The fuel to the furnace is regulated automatically to produce a given temperature at the exit of the drying section. Normally this temperature is about 149°C (300°F) when the stack temperature is 135°C (275°F). About 94% of the dried dust settles out in the primary separator, and the balance of the dust is separated in the secondary cyclone separator. The fine dust from the cyclones is returned to the furnace for combustion. The dried dust from the primary separator is moved with air to the boiler-plant storage silo or to the carbonizer.

The prototype carbonizing unit (shown in Figure 24) is designed to handle 31 tons of dried lignite per hour. This unit is designed for automatic control. The reactor is of alloy steel, 6 feet in diameter and 35 feet in height, mounted in a special furnace which is equipped with recuperators for efficient operation and preheating of process and combustion air. The process air is compressed to about 8 lbs. Per sq.in. gauge and passes through a recuperator where it is preheated to about 371°C (700°F). Part of the preheated air is diverted back to the drier where it picks up the dried lignite for transport to the reactor, and part of the air goes directly to the base of the reactor. The mixture of air and coal enters the base of the reactor, and the fine coal is moved upward through the reactor entrained in the products of reaction in a turbulent condition. The solids are not in a true fluidised state because of the high superficial velocity of 6 to 8 feet per second. The temperature of the products leaving the top of the reactor is controlled automatically by regulation of the combustion around the reactor or by regulation of the air-coal ratio similar to methods employed in the smaller pilot plants.
The combustion air is drawn into the system with a fan, which also circulates a portion of the products of combustion from the furnace. This mixture enters the fan at about 316°C (600°F) and is forced through the combustion air recuperator, where it is preheated to about 649°C (1200°F). From this recuperator the mixture of air and products of combustion passes to the furnace ports through a series of flues connecting the distributing bustle with each port. The
air-gas ratio is controlled to provide about 5 per cent excess air, and the recirculated products of combustion are regulated to secure a maximum flame theoretical temperature in the reactor of about 1482°C (2700F), but the developed temperature around the reactor is controlled to less than 1038°C (1900F) to protect the heat-resisting alloy. Under these conditions of combustion the rate of heat transfer to the reactor wall is maximum because of the high heat transfer by direct radiation from the gases. Rates of heat transfer exceeding 1.000 Btu per hr. persq.ft. have been attained in the pilot model at Denver, and it is expected that the rate will be in the range of 9.000 to 10.000 Btu per hr. per sq.ft. in the large unit when operating at rated capacity. The volume and heating surface of the reactor have been selected to permit carbonization of the coal at a rate of about 60 pounds per hr. per cu ft. when the superficial velocity of the gases is 6 to 8 ft. per sec.

Parry selected the entrained process because the fine coal was heated in a minimum time and the capacity of a reactor was relatively high. About 12 min were required for carbonisation, at the rate of 60 to 90 lb of lignite per cubic foot of reactor volume per hour, depending on the diameter and volume of the reactor. Of the heat required for carbonisation, 30 to 40% was transferred into the reactor through the wall, and 60 to 70% was derived from combustion of air with carbon inside the retort. Because approximately 80 Btu were transferred to the lignite per cubic foot of air introduced, 2 to 4 cu ft of air were required per pound of dry lignite carbonised. By regulating the amount of heat transferred through the wall and by changing the air-coal ratio, the extent of carbonisation could be controlled automatically.

### 5.5 Other entrained processes

An entrained carbonization process has been patented, wherein finely divided coal and highly heated gas were injected into an externally heated, cycloneshaped chamber in such a way that the mixture traveled in a helical path. The differences in speeds between the slower-traveling coal and the gas increased the rate of the heat transfer. By maintaining a suitable amount of oxygen in the gas mixture, the surfaces of the coal particles were rapidly oxidized, thus preventing sticking and agglomeration. An added precaution was the interposition of a smaller stream of the gas close to the wall of the cyclone. The larger particles of char were
collected in a water-cooled chamber at the outlet of the cyclone and the finer sizes were removed from the gases in a electrostatic precipitator.

A circulating bed was used by Taskaev and Kozhina [40] in experiments on the low-temperature carbonization of coals. Product gas preheated to from 600 to 700°C was recirculated to entrain the coal used in sizes up to 1/2 in. and to carry the particles through a vertical nozzle 1 in. in diameter and 20 in. long into a concentric tube 6 in. in diameter and 40 in. long. The coal or char particles fell through this space in a dense descending bed, and were then reentrained by the gas and recirculated until carbonization was complete. The yield of primary tar produced at a retort temperature of 450 to 500°C was about 80% of that obtained in the laboratory in an aluminum retort. The process was recommended by the authors for the production of char to be used in industrial boilers. The product gas possessed a high heating value, about 800 Btu/cu ft because it was undiluted with gaseous products of combustion.

In a process patented by Pettyjohn, a stream of 6-mesh coal granules was entrained in air at a temperature about 50°C below the softening point of the coal. Pettyjohn introduced the mixture, together with additional air at a temperature of at least 420°C, into a short, tubular vortex chamber. The temperature of the mass in the vortex was maintained at 650 to 1040°C solely by combustion of the volatile matter driven off from the coal. Devolatilization to thin, fragile granules occurred in less than 20 sec.

A free-falling process has been used by Schutte for carbonizing coal and oil together. The coal in particle form, liquid oil, and preheated coke particles were introduced independently but simultaneously into the free-falling zone under turbulent conditions. The coal was rapidly heated through its plastic state and the preheated mixture descended through a reaction zone as an unagitated, gravity-packed mass. The proportions of coal and oil and of the preheated coke particles were controlled to maintain in the reactor a temperature of 540 to 760°C until carbonization had been completed, without introduction of other source of heat. The char particles and the volatile products were withdrawn at the base of the reaction zone and separated.

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6 Horizontal retorts

Horizontal retorts consist essentially of a long, steel vessel, often cylindrical, through which the coal is moved as carbonisation proceeds. Horizontal retorts basically differ in the methods employed for transferring the heat of carbonisation to the coal charge. The most common heating medium has been hot combustion gases, which have been applied indirectly by external heating of the shell, directly by passing the hot gases through the retort in contact with the coal, and by a combination of both methods. To recover a rich product gas undiluted with combustion gases, the product gas has sometimes been reheated and recycled through the retort in direct contact with the coal. A further important method of heating is the use of preheated, solid bodies, including char, sand, or iron balls.

Cylindrical retorts may be either rotated or fixed. To keep the coal moving through the retort, the longitudinal axis of a rotated retort is frequently inclined a few degrees to the horizontal, with the coal fed in at the higher end and the char removed at the lower. Fixed horizontal retorts are provided with a screw or paddle device, which by rotation conveys the coal and char from the entrance to the outlet of the retort.

Rotating horizontal retorts are operated continuously and are essentially simple in design. Addition of the drive and conveying mechanism to a fixed retort increases its complexity. The capacities of horizontal retorts are often relatively limited, so that a greater number of individual units may be required for a given capacity than would be needed for fluidised or many vertical retorts.

Because of the direct contact, which necessarily exists between the heated coal and the walls or other parts of a retort, a caking coal will tend to stick and accumulate in large masses on these surfaces unless precautions are taken to prevent adhesion. As with other types of retorts a caking coal may be pre-treated to reduce its agglomerating tendency, or the coal may be diluted with an inert medium, such as char or sand, until sticking has been prevented.
6.1 Continuous horizontal retorts

Continuous horizontal retorts are divided in: indirectly heated and directly heated. Examples of indirectly heated retorts are:

- Disco process
- Hayes retort
- Mimura carboniser
- Wanishi carboniser
- Lesher processes

6.2 Disco process

The Disco process [16,31,41,42], the only commercially successful, low-temperature carbonisation process now utilized in the United States, employs rotating horizontal retorts. The process has been operated since the early 1930’s in a plant near Pittsburgh, which contains seven retorts with a total rated capacity of 800 tons of Disco char per day. The plant is located adjacent to a coal washery that belongs to the Consolidation Coal Company for the purpose of converting the fine sizes of cleaned bituminous coal produced in this washer into lump char suitable for domestic use. The cost of the present installation, which was completed in 1947, was reported to be $3,000,000.

The Disco process is unique in that it produces a lump char directly from the fine coal in the continuous rotating retorts, without the application of a briquetting step. This feature is secured by a preliminary oxidation on “roasting” of the coal in contact with air, thereby destroying a portion of its excessive caking characteristics. In the rotating retort, the oxidized coal, together with recycle char, rolls up into balls of char without adhering to the sides of the retort. The balls of char are screened and the sold as the smokeless domestic product.
In the Disco process, wet fine coal from the washery goes through five basic steps: (1) predrying, (2) “roasting”, (3) carbonising in the retorts, (4) cooling the char, and (5) screening and loading. The gas and tar are recovered as by-products.

The drying equipment lowers the moisture of the coal to 2 to 3%, and consists of three flash dryers heated by the hot flue gas from coal-fired furnaces located below the dryers. The dried coal is carried from cyclone collectors at the top of each dryer unit by a conveyer belt to the roasting step in the carboniser building.

In the roasting process, the coal is preheated and partially oxidized while hot by exposure to controlled quantities of air. By exposure to the air at an elevated temperature, some of the excessive agglutinating or caking property of the bituminous coal is destroyed, but enough is retained so that, under the conditions in the carboniser, the coal will roll up into the balls, which are the characteristic form of the finished char, but will not adhere to the retort walls. The oxidizing actions is closely controlled so that sticking of the coal is restricted to the extent of forming balls of char with the proper strength and reactivity but without the building up of adherent masses of coal or char on the walls of the retort. Roberts has shown that the process is governed by temperature and time factors, and that the proper conditions for a given coal are determined by experience. The oxidizing operation is carried out in a roaster consisting of six, long, rectangular hearts, located one above the other, through which the coal is continuously dragged in series by rabbles. The coal is heated indirectly by hot combustion products, which are circulated through flues built in the hearths. The lowest two passes differ from the four topmost ones in that some coal is recycled through them so that uniformity of quantity and quality of the feed from the roaster into the carbonising retort may be maintained exactly. Manual air control is maintained in the ducts of the roasters. The coal from the last pass is fed to the carboniser by a rotary-feed device at 320°C.

The carbonising retort proper is a rotating cylindrical 5/8 in steel shell approximately 9 ft in diameter and 126 ft in length, i.d. The retort is mounted inside an insulated, stationary steel cylinder. The inner shell is mounted almost horizontally, but with a slight slope so that it rotates like a typical cement or limekiln. Heating is external by means of flue gas circulated
at high speed through the annulus between the outer stationary shell and the revolving retort. The heating gas is retained at the two ends of the heating space by rubbing strips which are attached to the outer shell and which ride on the rotary shell to give a suitable gas-tight closure. The prepared coal drops directly from the roaster, which is located just above the retort, into a screw feeder which discharges at a point close to the upper end of the rotary shell. The coal at 320°C drops into the shell and is soon heated in to the plastic temperature at which the formation of lumps or balls begins. These balls roll around inside as the shell rotates, gradually working their way to the lower discharge end.

The gas generated during carbonisation passes out of the retort through a takeoff at the upper (cooler) end and thence through a wash box to a direct-type primary cooler. There, tar condensing and liquor scrubbing are accomplished. The liquor for spray operation is recycled through a cooling tower. After a final scrubbing, the gas is burned in the retort furnace to provide heat for carbonisation. Hot products of combustion are circulated from the furnace by a fan in a closed circuit through an annular space around the carboniser, through ducts in the roaster, and back to the furnace again. The maximum temperature in the heating space at the coke-discharge end of the carboniser is automatically held at 570°C.

The rate of throughput of a retort is determined by the rate of feed. After the balls are formed, they roll gradually down the slope and are discharged at the rate at which they are formed from the feed. The retort is largely free space except that, at intervals along its length, several 8-in baffle rings dam the flow of balls and cause some piling up. The latest retorts are each designed to process 150 tons of dry coal per 24-hour day (6.25 tons per hour). The yield of char has been approximately 75% of the weight of coal, thus insuring daily 750 tons or more of marketable balls from the seven retorts of the new installation.

As normally prepared, the solid product contains 15 to 17% by weight a residual volatile matter. This percentage can be varied within limits from 14 to 20%. The solid char is screened into several sizes. The smallest size is crushed to ¼ in, and added to the coal as the latter enters the feed hoppers above the roasters. A major control of the size and structure of the ball product is secured by varying the percentage of this undersize breeze in the feed.
Minchin described a somewhat similar process, which was operated experimentally at Marienau in the Saar, to produce a fine char suitable for use in the coal mix to coke ovens. The arrangement consisted of two horizontal, rotating cylinders, one above the other. Inside the upper cylinder was a second concentric drum, and both were provided with vanes attached to the inner surfaces of the walls for movement of the coal. Inside the drums the “flambants sec” coal was subjected to a degree of oxidation by contact with a counter current stream of air. In the lower cylinder, carbonisation was completed at 550°C. Passage of the coal through the entire unit took 1 ½ hr. The drums were heated externally by hot products of combustion. The loose, friable char from the retort, when added to a mixture of local, noncoking coals, lowered the volatile content of the mixture sufficiently so that a reasonably good coke could be produced in a coke oven. The mixture charged comprised char 20%, Saar coal (Gras A) 25%, and Gras B from Lorraine 55%. The final coke was hard and good enough for a blast furnace trial.

6.3 Hayes retort

The basic principles of the Hayes retort [16,23,43], which is shown in Figure 25, have been described by Price: A steel drum, 20 in diameter and approximately 25 ft long, is supported on rollers and enclosed within a brick furnace. This drum rotates at approximately 1 ½ to 3 rpm. Both ends of the drum are closed by stationary end covers. At the feed end, coal is fed into the inside of the drum by a variable-speed, sealed screw feeder. The charge is moved through the retort by means of a 16 in screw extending the full length of the retort and driven by a reversing motor. The charge is first moved forward (toward the discharge end) and then backward for a shorter distance within the retort. This reversing screw serves several purposes. The charge is kept well agitated within the retort; the length of time the charge is held within the retort can be varied by changing the period of forward and reverse motions of the screw; and the wiping action of the screw against the walls of the retort delays the accumulation of a hard carbon deposit on the inside of the retort.
Heat is supplied to the retort through a gas burner operated on premixed air and gas and provided with automatic temperature control. At the discharge end the hot char drops into an enclosed screw type-quenching conveyor, in which it is cooled with water sprays and discharged through a star discharge feeder.
The Hayes process was developed by the Allis-Chalmers Manufacturing Company. The first installation, erected at Moundsville, West Virginia, in 1928, contained five retorts and was operated until 1936 when they were damaged beyond repair by food. The char was rather small and suitable for boiler underfiring. In 1940, a pilot retort was installed adjacent to the coke plant of the Colorado Fuel and Iron Corporation at Pueblo, Colorado. The char was mixed with high-volatile coal to produce the charge for high-temperature coke ovens.

6.4 Mimura carboniser

The Mimura carboniser [44], a Japanese development at the time of World War II, consisted of an externally heated, rotating retort, 25 ½ ft long and 25 in i.d. The carboniser was fabricated from mild steel and provided with rabble arms connected to a concentric shaft. These arms possessed two motions, a slow rotation at intervals and a horizontal motion, lengthwise of the retort.

On the forward stroke of the shaft, blades on the rabble arms pushed the charge ahead of them through the retort. The agitator was then rotated until the blades were out of the coal, and on the return stroke only pins were dragged backward through the plastic mass. The agitation, together with the rotation of the retort, was intended to prevent agglomeration of large masses of material and their adhesion to the wall of the retort. A diagram of a retort and agitator assembly is shown in Figure 26.

Operating difficulties encountered in the Nagoya plant appear typical of the possible problems, which could be encountered with this type of retort. The retorts were operated at a normal temperature of 500°C to carbonise a strongly caking coal; but this procedure resulted in masses of char, which adhered to the rabble arms of the internal agitator, and eventually froze the agitator in the retort.

If movement of the agitator was attempted under such a condition, the arms of the agitator bent and the shaft failed. Even with only moderately caking coals, char packed between the
blades of the agitator and prevented movement of the char. Attempts to eliminate the stoppages by a slight preoxidation of the coal during drying, by blending of coals, and by variation in speed of rotation of the agitator or in the carbonising temperature were of little benefit.

Trouble was also encountered when air leaked into the combustion space surrounding the retorts and disturbed the air: fuel ratio. Local overheating occurred, with the result that temperatures of the metal parts of the retorts sometimes exceeded 700°C.
Since all metal parts were of mild steel, the life of the retorts and agitators never exceeded 150 days, and failure sometimes occurred within 15 days of being placed in service. Warping of a retort induced by an uneven heating caused leaking of the product gas through the end seals. In the retort, which was operated under a static pressure of 1 in water gauge, this leaking was usually ignited, with resulting damage to the mechanical equipment driving the retort and agitator. In another retort operated at a negative pressure of 2 in water gauge, air leaked through the seals into the retort and burned a portion of the gas. Attempts to design these seals to prevent leakage were unsuccessful, the retort itself being insufficiently rigid to prevent warpage under the temperature conditions imposed.

The mechanical driving equipment was complicated; shutting down an entire bank of retorts to stop one for repairs was necessary and the hydraulic actuators for the horizontal movement of the internal agitators were inadequate. Because of jamming of the internal agitators, uncontrolled air leakage into the combustion chambers, warping of the retorts, leakage of rotating end seals, and general mechanical difficulties, not more than eighty retorts of the one hundred and twenty installed could be operated simultaneously. The retorts were designed for a daily capacity of 300 tons of coal, but under the best conditions only 82 tons/day could be carbonised in the plant.

6.5 Wanishi carboniser

The Wanishi carboniser [44], designed in Japan during 1936, employed an externally heated, rotating, horizontal, cylindrical retort without internal agitation. To move the charge through the retort, a continuous internal spiral flange was provided. Each Wanishi retort was 76 ft long and 7 ft 10 in i.d., was rotated at 0.28 rpm, and was mounted in an individual gas-fired furnace without recuperators. Four carbonisers were installed; but only three were operated at a time, one being retained as a spare. Each carboniser was designed to handle 100 tons of coal per day, and the normal capacity of the plant was 300 tons of coal per day. The gases from the retorts passed through a hot, cyclone type dust separator to remove tar mist and small amounts of dust. Because of the large size of a retort and its high thermal losses, the
quantity of gas burned for underfiring exceeded that produced in the process, and a
supplementary source of fuel gas was required. The temperature of a retort shell did not
exceed 600°C and the carbonising temperature was about 550°C. The coal was dried before
carbonisation in an inclined rotary drier, 73 ft long and 6 ft 4 in i.d., which was internally
heated by products of combustion leaving the main heating chamber. The drier was
connected to the carboniser by a closed hopper and a rotary feeder, which prevented loss of
volatile products. The carbonisers usually were stopped annually for cleaning. The shutdown
required about a month for removal of the accumulated tar and dust from the surfaces and for
general repairs.

The objective of the Wanishi process was to produce a char to replace low-volatile
bituminous coals in the coal blends to coke ovens. Only noncaking coals were carbonised at
Wanishi, because caking coals would have built up into masses of char, which clogged the
retort. Because the char was required in sizes smaller than 3 mm, it was necessary to use a
coal capable of producing lump char.

6.6 Lesher processes

In addition to the Disco process, Lesher has been responsible of the design of other retorts. In
one arrangement, he proposed, first, to preheat the coal by mixing it in a screw conveyor with
char heated to about 480 to 590°C, and then to devolatilise the mixture in an externally
heated, horizontal rotating retort. The temperature and proportion of recycled char were
regulated to raise the coal to a temperature just below its softening point. Recycling of the
char was employed to ensure uniformity of the finished product. Together with the control of
the speed of rotation of the retort, recycling permitted the production of a granular, almost
dustless char from a coking-coal feed.

A continuous, rotating horizontal reactor, which operated with a protective barrier of
preheated char in contact with the walls to prevent sticking of a highly caking coal, was also
patented by Lesher. The char discharged from the retort was maintained at a temperature
between 540 and 760°C, with a ratio of fresh coal to recycled char in the feed to the retort below 1 to 3.

Examples of directly heated horizontal retorts are:

- University of Kentucky process
- Humboldt sand carboniser

6.7 University of Kentucky process

Some experimental work has been done by the University of Kentucky on the production of char in a continuous rotary retort [45], which was operated at an inclination up to 7.5° to the horizontal. The retort was heated by direct contact with gases, which entered the lower end of the retort at about 540 to 760°C and flowed counter current to the coal. To overcome accumulation of char on the walls of the retort, a series of hammers was incorporated into the retort design to break off the accumulation by striking periodically against the shell of the retort.

6.8 Humboldt sand carboniser

In the Humboldt sand carboniser [6,46], the heat for carbonisation was supplied by addition of sand, which had been preheated to a temperature of about 750°C. This preheated sand, together with briquettes of the coal to be carbonised, were introduced at one end of the horizontal, cylindrical drum. The drum was divided into two semi cylindrical sections by means of a screen extending lengthwise through the center. Annular separating walls further divided the drum into a number of cells, and slanted conveying ledges were attached to the partition in such a way that, as the drum revolved, the briquettes were carried upward and transferred to the next cell, and the sand dropped freely from them through the screen onto briquettes below. In an experimental installation, which was tried in Essen toward the end of World War II, the briquettes were prepared from a mixture of coal ground less than 3mm and iron-ore fines held together by suitable binder. The briquettes were carbonised at 550 to
600°C for about an hour in a retort 10 m long with i.d. of 2 m, rotated at 1 rpm. The drum was surrounded with an insulating shell to retain heat. The vessel for heating the sand consisted of a refractory-lined housing fired by means of a gas burner with the sand passing counter current to the flame. In addition to the carbonised briquettes, about 220 lb of primary tar and 3178 cu ft of gas (heating value 843 Btu/cu ft) were recovered per ton of briquettes. In a final step of the Humboldt process, the carbonised briquettes were smelted with a proportion of lump ore in a low-shaft furnace employing cold blast. Although the combination of iron-ore fines with coke in briquettes fed to the blast furnace was not new, it was reported that the low-temperature char was advantageous because of its high reactivity. Successful results were claimed in operation of the experimental plant, but a full-scale test was needed to establish costs.

The use of hot sand in rotary retorts has been recommended by other workers because of the rapidity and ease with which heat is transmitted from the sand to the coal, a retort about 6 m long being adequate for such an operation. Cooling the briquettes slowly in the hot sand prevented the development of the stresses which might occur if the briquettes were quenched rapidly in water Jennings reported that approximately two-thirds of a ton of sand per ton of coal charged was sufficient to supply the heat of carbonisation, and that the conditions of carbonisation could readily be adjusted to suit requirements. Further advantages were the ease of handling and safety with sand and the production by the carbonisation process of an undiluted gas. No caking of briquettes containing pitch binder is possible because the pieces are separated by sand. Because of the higher temperature of the sand, any tar evolved is completely vaporized and does not adhere to the sand. Hot sand was also used by the British National Coal Board to supply the heat required for carbonisation. Operation of a pilot plant indicated that carbonisation with sand could be completed in about one-fourth of the time required in indirectly heated narrow ovens.

Korobochanski employed metal balls 50 mm in diameter containing a mixture of calcium chloride as the medium to supply the heat needed for low-temperature carbonisation of low-rank coals. Aspegren has also described a horizontal vessel in which heated balls could be used. Johansson has patented a process in which the coal is preheated and then carbonised in
direct contact with heat-carrying balls in a rotating horizontal drum. The balls removed from the carboniser were reheated by combustion of volatile products evolved from the coal.

The use of molten metals as a means of promoting heat transfer in carbonisation has also been proposed. Molten lead was used in the Piron-Caracristi process more than 30 years ago. More recently, molten aluminum has been proposed.

Use of a rotating horizontal kiln was proposed by Phinney for producing a char of uniform size and volatile-matter content. The heat for carbonisation was supplied by introducing into the kiln hot, recycled char of a size finer than that of the coal. The kiln products were discharged to a separator for gas and tar, and the solid material was then elutriated to separate the fine, recycled char from the larger lumps of freshly formed char. The finer sizes of char from the elutriator were reheated by partial combustion in a fluidised bed and recycled to the retort. Temperatures in the range 450 to 570°C were found suitable.

A flowable mass of solid particles made up of iron ore and char adhering together was produced by Lesher. He first mixed preheated iron ore and the coal under conditions such that the final temperature of the mixture did not exceed the softening point of the coal. The heated mixture was then heated and agitated in a rotating retort. Under these conditions, the coal softened and became sticky in contact with the ore, and the two materials were rolled into fine solid particles. Heating was continued until the particles had reached a temperature of 450 to 540°C.
7 Fixed horizontal retorts

Rabble arms, screws, or other devices are necessary to convey the coal through fixed horizontal retorts. Fixed horizontal retorts appear to have been used to a more limited extent than have rotating retorts. Some examples are:

- Shimomura retort
- Cotarco retort

7.1 Shimomura retort

The Shimomura process [44] was used in Japan in the 1920’s to produce a char suitable for use in the coal blends to coke ovens. Each stationary horizontal cylinder, constructed of cast iron, was externally heated, and was provided with rabble arms arranged on a central shaft to convey the coal through the retort. Two cylinders were combined to form a set, with one retort located above the other in the gas- or coke-fired refractory furnace. Each retort was usually 12 ft long and 2 ft in diameter, with a rated capacity per set of 8 tons of coal per day. The raw coal was fed into one end of the upper retort by a gas-tight screw conveyer. The rabble arms then propelled it along the retort to the other end were it dropped through a vertical pipe to the lower retort. The finished char left the lower retort through a screw conveyor and water seal. Volatile products were removed from the retorts at the junction pipe between them.

The rabbles were mounted on a water-cooled, concentrically located shaft, and were driven at the high speed of about 130 to 150 rpm. The functions of the rabbles were not only to propel the coal along the retort, but also to keep it from accumulating on the walls and to pulverize it. The char was produced in small, rounded particles most of which pass a 1/8 in screen. The maximum temperature in the carboniser was usually not over 500°C. During operation, carbon accumulated on the linings of these retorts, with a resultant decrease in the rate of heat transfer. When temperatures were increased to about 800°C to maintain output, the
cylinders warped and failed. The life of Shimomura retorts were strikingly similar in principle and operating features to the primary retorts erected in the 1920’s at the Clinchfield Carbocoal plant, Clinchfield, Virginia, to produce char for briquetting.

7.2 Cotarco retort

In the Cotarco retort the rate of heat transfer from the metal heating surface of the wall of the horizontal retort to the coal was materially accelerated by a use of a stirrer, resembling a set of ice cream mixer blades, which turned through the bed of coal at approximately 20,000 times/hr to keep the coal thoroughly mixed and to produce a finely divided char. In 1955, an installation of this process was projected in Colorado.
8 Conveyor retorts

Processes in which the coal is carbonised continuously during its passage through a fixed horizontal duct while spread out on a conveying surface, are a third type of horizontal retort. Conveying retorts have the limitations that the moving parts of the conveyor are exposed to the temperature conditions in the retorts, and the coal must be spread out in thin layer. Coal capacities therefore are relatively low, and a multiplicity of units would ordinarily be required for large tonnages of coal. Coking stoker units, however, are examples of conveying retorts, which give relatively high capacities and, hence, are receiving much attention for producing industrial chars and cokes. In these units the coal is simultaneously carbonised to produce a char or coke, and the heat of combustion of the volatile products, together with apportion of the coal itself, is utilized in boiler firing. Examples of processes taking place in conveyor retorts are:

- Coking stoker
- C.W. & F. process
- Flat bed retort
- Storrs process

8.1 Coking stoker

The coking stoker process [6,47,48] is unique in that the coal to be carbonised is fed onto the travelling grate, the regular heating operation has been carried out, and at the same time a char suitable for certain uses has been discharged from the grate. The process was developed by Shawinigan Falls, Quebec, in connection with calcium-carbide manufacture. Figure 27 shows the essential arrangement of the Shawinigan stoker as applied to a limekiln. Bituminous cola from an overhead bin was spread on the travelling grate 2 to 4 in thick. Air was blown through the grate from several compartments, so that combustion of the coal on the grate could be carefully controlled. As the grate advanced into the firebox, volatile matter was distilled from the coal and partially burned with the air above the grate to supply necessary heat for carbonisation. The temperature of these gases was about 1200°C, so
almost all the coal-tar products were cracked to carbon and the simpler gases, CO, H₂, CH₄, and the lime. The hot volatile gases then passed through a 4 ft diameter burner pipe to the kiln, and burning was completed with secondary air in the kiln itself to supply the heat for lime burning. The presence of free carbon in the gases gave the flame in the kiln good radiant qualities. At the same time, a char containing about 3% volatile matter, which was satisfactory for manufacture of the carbide, was discharged from the grate to a conveyor. The coking operation was accomplished with a small loss (about 1/12) of the original fixed carbon in the coal. The hot gases from the limekiln were further cooled in a waste-heat boiler before leaving the stack.

Figure 27 Cross-sectional diagram of stoker carboniser

8.2 C.W. & F. process

A flat-bed, pan-pan process also known as the Wallace process [16], was developed by the Chicago, Wilmington, and Franklin Coal Company for the preparation of a smokeless domestic fuel from the coal fines produced in the operations of this company in the Southern Illinois coal fields. The coal was dropped into a pan conveyor, levelled, and then compressed by a second lid conveyor before entering the heating chamber. Because the heat was applied from heating flues to the top surfaces of the lids and bottom surfaces of the pans, a distinct zone of separation occurred in the charge. The volatile matter was reported to be reduced uniformly from 33% in the original coal to 14% in the char. The final char was produced in
blocks 6 by 12 by 2.5 to 3 in size. The cola company was reported to have installed a pilot plant with a capacity of about 10 tons of finished char per day at one of its mines in West Frankfort, Illinois.

8.3 Flat-bed retort

A continuously operated, flat-bed, low-temperature retort consisted of a flat plate [16,49], which was slightly inclined at an adjustable angle and was heated from below. Fine coal was fed onto the higher of the plate and was moved slowly to the lower end by means of vibrating mechanism, which actuated a series of scrapers, the faces of which were shaped to resemble a snow plow. The spacing and location of the scrapers was such that the entire surface of the plate was scraped. The slope of the plate and the number of strokes of the scraper controlled the rate of flow of coal. It was claimed that by use of a metal with high heat conductivity, such as copper for the plate, and by vigorous agitation, high throughputs were secured. A pilot-scale retort was reported to have been installed at the plant of Frank Lowe and Company, in St. Louis. Figure 28 indicates the general arrangement of the device.

![Schematic diagram for Poindexter-Lowe low-temperature carbonisation retort](image)

**Figure 28** Schematic diagram for Poindexter-Lowe low-temperature carbonisation retort
8.4 Storrs process

The Storrs, or “Coal Logs”, process [16,50-54], produced the char in the form of cylindrical sections, or “logs”, by a two-stage process, which included a flat, vibrating-bed carboniser followed by an extrusion unit. The carboniser consisted of a wiremesh belt conveyor onto which the sized coal was fed to a depth of about 0.5 to 1 in. About 1 to 2 in above the level of this belt was located a radiant heating surface. Below this surface the belt was moved at a speed generally in the range 0.75 to 1.5 ft/min and at the same time was vibrated through a vertical distance of about 0.5 in. The volatile and liquid products of carbonisation escaped downward through the wire mesh and were withdrawn from collector compartments located below the belt. The cola at about 400 to 500°C and in the proper condition of plasticity was discharged over the driving pulley into the extruder. The time of heating on the belt was about 1 to 3 min. The coal was compressed in the extruder into the logs (3 to 3.5 in. in diameter), which gave the process its name.

The radiant surface of the retort was heated by gas burners located at predetermined positions to provide the desired heat gradient. In a British demonstration plant, the heating facilities consisted of twelve independently adjusted burners, which were spaced along a combustion chamber 6 ft long, by 11 in. wide, and 4 ½ in. deep, extending the full length and width of the conveyor. This chamber was lined with refractories backed by insulating bricks. The bottom of the combustion chamber, which formed the radiant surface of the retort, was made of 3/8 in. “Staybrite” plate. The waste gases from the heating chamber could be used for heating and drying the coal or for steam generation in a waste-heat boiler.

The conveyor belt was fabricated of chrome molybdenum wire to withstand the temperature. The extruder consisted of a continuous, variable-pitch worm rotating at about 3 rpm inside a 3 ½ in. diameter tube through which the log was extruded. The tube was surrounded by a jacket, which was heated by gas burners to retain the plasticity of the coal during formation of the logs. As the coal was pushed through the tube, it was compacted by the pressure of the screw conveyor in combination with the resistance of the tube into a dense mass, which
solidified on cooling. As a result, the specific gravity of a log was not greatly different from that of the true specific gravity of the coal from which it was made. The carbonisation step could also be operated at a higher temperature to produce merely a small size of char for direct use as a boiler fuel or for manufacture of briquettes, which would then be carbonised at higher temperatures.

Figure 29  Longitudinal section through Storrs low-temperature carbonisation retort

The Coal Logs process was first developed by the Coal Logs Company in Salt Lake City, Utah, to produce a strong, smokeless, fuel from weakly caking Utah coal. Two installations, each of 100 tons/day feed capacity, were projected, one in Utah and the other at an undisclosed location. An installation was also reported at a plant belonging to the Appleby Frodingham Company in England. A flow sheet of the Storrs process is shown in Figure 29.

In the Utah plant, it was planned to remove the volatile matter as a series of separate fractions by withdrawing them independently from collector chambers spaced along the retort. This practice was expected to provide relatively pure materials with distinct physical and chemical properties, and to reduce the opportunity for cracking. No data on this arrangement have appeared. Pilot data from the retort indicated that a ton Utah bituminous coal could yield approximately 1400 lb char, 23 gal oil, and 1200 cu ft fuel gas with a heating value of 1000
Btu/cu ft. This gas was sufficient to supply the energy requirements for carbonisation. By further heating of the coal logs to 930 to 1090°C in a subsequent heater, a metallurgical coal was to be produced. The complete treatment was expected to furnish per ton of the Utah coal, 1200 lb coke, 29.5 gal volatile condensates, and 8000 cu ft of 600 Btu gas.

In a process resembling the Coal Log process, Reintjes coked a carbonaceous material, such as raw coal, by forcing it with or without admixing a fluid binder (depending on the thermoplastic nature of the material) under a slight or moderate pressure through a smooth walled, externally heated metal tube, from which oxygen was substantially excluded. The extent of carbonisation depended on the length of the tube, temperature therein, and the time of contact. The char was extruded from the end of the tube as a substantially solid cylinder, which was, however, cracked and fissured as a result of shrinkage and the escape of vapours. As it was extruded, the cylinder broke off into lumps. The temperature of treatment was at least 430°C, preferably in excess of 590°C. Calcining of the product at higher temperatures usually proved advantageous. The process was particularly useful for preparing superior cokes for electrode-forming masses.
9 Tray retorts

Retorts consisting of horizontal trays mounted in a vertical shell and resembling Nichols-Herreshoff furnaces are among the devices used for carbonising coals. Examples processes taking place in tray retorts are:

- Baumco process
- Buttner retort

9.1 Baumco process

In the Baumco process [5,51] for briquettes, the coal was subjected to a preliminary carbonisation at about 800°C in a tray retort, termed a “devolatiliser”, to produce a char without caking properties. This char was then mixed with a binder and with additional coal in proportions that would provide briquettes of the necessary strength. The briquettes were finally carbonised by contact with hot gases at approximately 1200°C. The process was recommended for carbonising coals of very low coking power (probably with a swelling number not over 1), noncoking coals, and lignites.

In the primary carbonisation, coal was used with a size range of up to 3 mm and containing not over 12 to 15% moisture. In the Herreshoff type of furnace, the coal was raked slowly in thin layers over the series of horizontal, circular trays, descending successively from one tray to the next. Temperatures not in excess of about 700°C were attained by direct contact with the hot gases from the second carbonisation stage, which were passed upward through the trays of the retort counter current to the flow of coal.

The char from the devolatiliser was then mixed with the binder, which consisted of up to 10% of coking coal and up to 10% pitch. The mixture was briquette, and the briquettes were carbonised at the elevated temperature in a continuous, vertical-shaft retort. This retort was of a type similar to ones described earlier in this chapter and was heated both directly and
indirectly, with heating gases flowing through flues around the retort and upward through the shaft in direct contact with briquettes. The lower section of the retort was used as a quenching chamber to cool the briquettes by the injection of product gas, which had been cleaned and cooled in the tar-recovery system. Passage of the coal through the devolatiliser and the briquettes through the vertical retort required about an hour each. Because of dilution of the make-gas with the products of combustion, the volume was large, with a heating value of only about 140 Btu/cu ft. However, by preheating the combustion gases, it was reported that product gas with a heating value as high as 500 Btu/cu ft could be obtained. The finished briquettes were strong, dense and uniform in size. By adjustment of conditions in the second carbonisation stage, their volatile content and other properties could be varied. They appeared suitable for domestic, industrial, or metallurgical use.

9.2 Buttner retort

The Buttner retort [2,19] was developed in Germany before World War II for the low-temperature carbonisation of oil shale or noncaking coal fines, such as floated coal or slimes. Figure 30 shows two cross sections of a Buttner retort. It consisted of a stationary cylindrical housing, insulated against heat losses. The housing enclosed a structure consisting of circular shelves, which were revolved slowly by an electrical motor. A vertical shaft in the center of the shelf structure supported several circulating fans, the blades of which were sloped like those of a turbine. These fans were rotated in a direction of rotation of the shelves. Hot waste gases or hot combustion gases produced by burning a fuel in a furnace were introduced into the cylindrical chamber, and then were drawn between the shelves by the turbine fans. From the top of the retort, a portion of the exit gases were recirculated back to the furnace and the remainder were released to the atmosphere. The coal to be carbonised was admitted uniformly at the top and distributed evenly on the top shelf. After the coal had completed one revolution on the top shelf, it was forced by means of a stationary adjustable scraper down through a slot onto the next lower-rotating shelf. After each revolution, the process was separated until char was discharged at the bottom onto a belt, which transferred it to the screening plant. The coal was not agitated violently while passing through the unit, and dust
formation was said to be small. No data on its performance as a drier has been found. However, it is similar in many respects to the Wyssmont turbo drier.

**Figure 30** Buttner coal drier adapted to low-temperature carbonisation of fine coal
10 Typical formed-coke processes

Carbonisation of coal is basically accomplished by heating it to a temperature at which the coal is broken down chemically and/or physically to simpler components, thereby converting the coal to a tar, and yielding gas, water, and a residual solid char. The most representative low-temperature carbonisation methods are:

10.1 The FMC process

The FMC coke process can produce a metallurgical grade of coke in a continuous system from a wide range of coking and non-coking coals [55,56]. The process is based on three-stage, fluidised bed carbonisation of coal to produce a solid calcinate (char) and tar, which are recombined, blended, and briquetted into a desired shape and size.

The first fluidised stage is called the catalyser. It operates at 149-260°C, depending on the coal. In this stage the coal is dried and mildly preoxidised to destroy its caking property. Heat is supplied by a submerged steam-heated coil, and oxygen is supplied at low concentrations in the fluidising gases. This step also tends to densify the calcinate and improve the quality of the briquettes. The coal from the catalyser is transferred to the second stage, called the carboniser and operates at 482°C, where it is fluidised by air. Tar and low Btu fuel gas are taken overhead, the tar recovered by condensing, and the gas used for plant fuel. The char from the carboniser contains about one-half the original coal volatile matter. This char is transferred to the third stage, called the calciner and operates at 816°C, where it is fluidised by additional air. The particles lose most of the remaining volatiles, and they shrink. This step is necessary for the formation of strong briquettes. The exiting calcinate is transferred to a fluidised-bed cooler, where the solids temperature is reduced to allow safe downstream processing of the calcinate. The calcinate is fluidised with recycled inert gas, and heat is removed by a submerged heat exchanger than can generate either hot water or stream.
10.2 The COED process

The COED process uses low-pressure, multistage, fluidised-bed carbonisation of coal. The hot fluidising gases flow counter-currently to the net forward flow of solids (coal char), thereby heating the solids to successively higher temperatures. Heat and fluidising gases are simultaneously generated for the process by the combustion of part the residual char with oxygen in the last (hottest) fluidised stage. The basic approach of this process is to minimise solids agglomeration while maintaining maximum inherent liquid yield from the coal by controlling the temperature in each of the successive carbonisation reactors at a little below the fusion point of the coal (or char). After the solids are allowed to reside at this temperature for a period of time, while the fusion point temperature is increased significantly. The solids can then be safely forwarded to the next reactor operating above the fusion temperature of the original feed coal. As the caking tendency of the coal increases, the number of such carbonising stages also increases. When lignite or sub-bituminous coal is used a minimum of two carbonising stages are required.

In a multiple stages COED process, the coal is superficially dried and milled. Then it is fed to the first stage where it is heated to about 316°C by hot, oxygen free fluidising gases entering at about 482°C. The balance of the free moisture, most of the inherent moisture in the coal plus some gas, and up to 10% of the tar yield, are driven off from the coal. The tar is recovered by condensation, and the non-condensables are reheated and recycled to stage one. The fusion temperature of the solids is increased, so that they can be transferred from the first stage to the second stage, which may operate at about 454°C. Heat is supplied to the second stage by hot gases from the third stage plus recycled char from one of the downstream stages. Most of the tar and some of the pyrolysis gases are distilled from the coal in the second stage. The overhead gases from this stage will have substantial tar content. This precludes using these gases to supply the heat for the first stage, since considerable tar would be condensed from them at the lower first-stage operating temperature. Char from the second stage is forwarded to the third stage operating at about 538°C. When operating with bituminous coal, the third stage temperature is selected more to minimize vapour phase coking of the distilled tars than to avoid agglomeration of the solids. Heat is supplied to the third stage by hot gases.
plus hot recycled char from the fourth stage. The balance of the tar and much of the pyrolysis gases are distilled from char in the third stage.

The last stage of the process generates the necessary heat and fluidising gases to pyrolyse the coal. Char from the previous stage is forwarded to the last stage, where air or steam-oxygen mixture fluidises the solids and provides the oxidant for combustion of char. Ideally the last stage is maintained at the highest temperature possible without ash clinkering. Temperatures as high as 871°C have been used in the past. To avoid clinkering, it was found that helpful to increase the gas velocity at the point where the air or steam-oxygen contacts the char. Only about 5% of the coal is required to supply the heat for the carbonisation process. The rest of the char produced, which may represent upward of 60% of the dry coal, can be used in any number of ways [57].

The overhead gases from the second stage are treated (two alternatives exist) to recover the product char. In the first system the gases are quenched by a re-circulating system of water in a venturi scrubber, with the resulting tar and water separated. A substantial amount of tar fog is formed in this quenching operation, and recovered by a second venturi scrubber, followed possibly by an electrostatic precipitator. In the second approach the tar is removed from the pyrolysis gases by a staged oil scrubbing. A bottom, hot stage is used to knock down entrained solids and to condense the heavy ends of the tar. The resulting heavy tar-solids slurry could be recovered for hydro-treating operations, noted below. Alternatively, this slurry could be pumped back to the third carbonisation stage to crack these heavy ends, thereby attaining a lighter overall tar product. The lighter tar fraction, not recovered in the bottom section, is recovered by in the lower temperature, top section.

The recovered tar is filtered using a pre-coated rotary pressurized filter, to remove entrained solids carried over by the gases. The filtered oil or the top oil from the adsorber is forwarded to the hydro-treating step, in which the tar is reacted catalytically with hydrogen to remove the heteroatoms (O, N, S) to add hydrogen to the tar molecule and to produce a distillable fuel oil or synthetic crude oil for refinery operation.
10.3 The U.S. Steel Clean Coke process

The Clean Coke process, based on concepts developed by U.S. Steel, is designed to employ both a carbonisation and a hydrogenation step, operating in parallel, to produce metallurgical coke and also tars, oils, organic liquids, and gases [22-24, 58].

Carbonisation takes place in a two-stage vertical fluidised bed. After beneficiation, a portion of the coal is carbonised in a fluidised bed in the presence of a recycle stream of gas, rich in hydrogen and essentially free of sulphur. Most of the sulphur is removed from the coal. The char is palletised, using a tar binder derived from the process. The pellets are cured and then calcined, producing hard, low sulphur metallurgical coke and a hydrogen-rich gas.

The remaining portion of the coal is slurried with a process-derived carrier oil and then hydrogenated non-catalytically at 3000-4000 psia and 482°C. Liquids and gases are removed from the residual solids and are processed into liquid fuels, chemical feed-stocks, and oil fractions, which are recycled to the process.

10.4 The Occidental Pyrolysis process

The Occidental Pyrolysis process is designed to produce liquid the gaseous hydrocarbons and a char suitable for use as a power plant fuel. The process is based on rapid low-temperature carbonisation of coal with short residence times, and it achieves higher liquid yields than any of the other carbonisation processes.

Carbonisation takes place in an entrained bed, in which minus 200-mesh coal is added to a stream of hot char. The operating pressure is 50 psig. Pyrolysis occurs in fractions of a second. Because the residence time in the reactor is short (less than 2 s), secondary decomposition of the volatilised products is minimized, thereby maximizing the yield of liquids. The non-condensable gases are recycled through a compressor for use as the carrier gas for conveying the feed coal to the pyrolysis reactor [59].
A portion of the char collected in the cyclone downstream from the reactor is heated by direct heat exchange with char combustion gases. The contact time between the recycling char and the combustion gases is short, thereby minimizing the formation of CO (from CO$_2$ and carbon). This reduces heat losses to the combustion gases and significantly improves the process heat balance [59]. The reheated char is returned to the reactor and supplies the heat for carbonisation. The quality of the gas produced can be increased by raising the pyrolysis temperature and also by using a lower rank coal.

The marketability of the char that is produced, and the feasibility of using it as a power plant fuel, would be enhanced significantly if its sulphur content were reduced. Laboratory tests showed that if the pyrolysis char were subjected to a mildly acid aqueous leach to remove iron and calcium compounds in the ash, the effectiveness of the hydro-desulphurisation reaction would be improved markedly. The sulphur concentration of the resultant char is a function of the H$_2$S concentration in the H$_2$ used for hydro-desulphurisation, the residence time of the leached char, the type of coal from which the char has been produced, and the reaction temperature.

The tars that are produced by coal pyrolysis can be upgraded by hydrogenation. However these chars contain organometallic compounds, whose composition is a function of the coal from which the tar had been produced. Organometallic compounds are potential hydrogenation catalyst poisons. This factor will have to be considered when selecting the proper catalytic hydro-treating process.

### 10.5 The Lurgi-Ruhrgas process

The Lurgi-Ruhrgas process is very similar to the Occidental process. In this process the coal is carbonised by heating it rapidly with a circulating stream of hot char in a mechanically agitated mixer. The mixer has two shafts with radially attached arms, which rotate in the same direction to prevent the collection of unwanted deposits of coal and char dust and tars. This mechanical system was replaced by a single high-pressure steam jet coal feeder.
new mixer led to a coarse char size consist, which created char circulation control problems and cyclone furnace combustion difficulties.

After carbonisation is complete, the char mix is split into two streams. One stream contains the product char, which consists of the finer size fraction of the total solids stream after carbonisation. It can be forwarded hot as the fuel to cyclone furnaces, which provide heat to the utility boiler. The other stream is conveyed to a lift line and is pneumatically raised to the top of the line. At the bottom of the lift the char is picked up by air, which rapidly oxidizes a portion of the char, thereby releasing heat of reheating to a higher temperature the char leaving the carboniser. The resulting combustion gases and elevated hot char are separated in a series of cyclone separators. The hot char is returned to the mixer to pyrolyse the feed coal. The combustion gases, containing about 6% CO+H₂, are used for lift-line air preheating and coal drying. The CO and H₂ in these gases can be burned with air, introduced into the lift-line gases after the removal of the char.

Product gases and vapours driven off from the feed coal in the carboniser are passed to cyclone separators to remove as much of the entrained solids as possible. Then they are cooled and scrubbed in a three-stage, gas-liquid contact system. Tars, oils, and water are condensed and collected separately. The uncondensed portion is a fuel gas having a higher heating value of about 550-1090 Btu/scf. The heating value increases with decreasing carbonisation temperature and the quality of fuel gas increases with increasing carbonisation temperature.

10.6 Consolidation Coal Studies

Consolidation Coal studied the effect of sweep gas flow on the tar yields obtained by low-temperature carbonisation of highly caking coals. To simulate large-scale operating conditions in a small-scale unit, they equipped their 6-in diameter fluidised reactor with a stirrer. This device provided the necessary added mixing in the small reactor to assist in achieving rapid heat transfer and good temperature control, even when operating at low fluidising velocities. In this way it is possible to use the same sweep gas rates in the small
unit as in a large one, and still maintain the same residence times without having to use the same bed depths.

Tests were made on high-volatile bituminous coals. The fluidising gas was cleaned recycle gas. The carbonisation bed was operated at 496°C and 10 psig. From the experiments was found that the sweep gas rate is defined as the number of cubic feet of gas entering the carboniser bed (at reaction conditions) per pound of maf coal fed. The amount of pre-oxidation of the feed coal is defined as the weight percent of oxygen consumed, based on maf coal.

Tar yield decrease with decreasing sweep gas rate. It was also found that the total tar yield for untreated coal decreased from 23-25 wt % maf coal at a sweep gas rate above 0.94 m³/kg maf coal to about 19 wt % at 0.31 m³/Kg. During these tests the solids residence time was 45-120 min. The amount of pre-oxidation required to avoid dogging of a fluidised carboniser varies greatly with the feed coal. For the highly caking coals used in these experiments, approximately 4% of oxygen addition is required for operation in an unstirred fluidised carboniser. The loss in tar yield due to pre-oxidation is about one pound of tar per pound of oxygen consumed. This is similar to the yield loss obtained in direct carbonisation of untreated coal when oxygen is injected with the coal. The loss of tar is less when oxygen is injected below the coal feed point, indicating that oxygen preferentially reacts with char instead of with tar vapours when both are present.

Increasing the solids residence time from 45 to 120 min appeared to have no effect on tar yields at 496°C. Increasing the vapour residence time from 22 to 84 s in the stirred carboniser appears to reduce tar yield by about 2 yield-percent. This phenomenon, however, may be confused by the decreased sweep gas flow associated with the increased vapour residence time.

The advantage of this process lies in its ability to handle non-pre-oxidized, caking, high-volatile coals under non-agglomerating conditions thereby achieving the inherently high
liquid yields of these coals. The scale up to commercial size of the stirrer used in this process does not appear to be feasible.

10.7 CSIRO Studies

CSIRO has studied fluidised carbonisation in a pilot plant, with particular reference to the production of char for use in the coal mixes to coke ovens. The coal was usually carbonised at 399-599°C, using air or gas recycled from the process as the fluidising medium. When the coal was fluidised with air, the heat for carbonisation was largely derived from the combustion of a portion of the char product, although electrical heaters were employed when starting. When reduction of the caking tendency was necessary, the coal was first oxidized at 300-350°C in a similar but separate bed. The tests showed that when air was use for fluidising, the yields of char and tar from a medium-volatile, non-agglomerating coal were lower than with recycle gas. On the other hand, the gas yield was increased, although it possessed a low-heating value. During carbonisation the coal particles swelled considerably, and a large proportion of the finer particles was carried over with the gases, so that efficient dust removal equipment was necessary.

Newer experiments were concentrated on the development of a process that would maximize the yield of liquids [60]. Laboratory experiments were conducted using a very small flash pyrolysis reactor operating at atmospheric pressure and about 510°C. The coal feed was finely ground. It was mixed with sand, and a stream of nitrogen fluidised the entire bed.

Evolved vapours and gases were flushed away by a secondary stream of nitrogen introduced above the bed. The gas residence time was about 0.5s. Under these conditions, the tar yield was 20 wt % (maf) for brown (lignite) coal feed and 30 wt % for a sub bituminous coal feed. When the tar produced from the subbituminous coal was submitted to catalytic hydrogenolysis, 63% was converted into a clear, volatile, easily ignited liquid and 27% was converted to into gas and water.
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OVERVIEW of LOW TEMPERATURE CARBONISATION

Present Status – Properties, Yields and Utilisation of LTC chars – Survey of
Various Methods – Pre-treatment Conditions & Effects – Advantages, Economic &
Technological Development

Part III.

Thermal Pretreatment: Conditions and Effects
Assessment of LTC Advantages, Economic and Technological Development

Report prepared for

OPET
Network

ISFTA – May 2004
1. **Thermal Pretreatment: Conditions and Effects**

1.1 **Introduction**

Pyrolysis is one of the most important carbon processes, since it is involved in all the main carbon processes such as, combustion, carbonisation, liquefaction and gasification. The name pyrolysis stems from Greece: Πυρ means fire and Αφεω means loose, untie. Thus, pyrolysis means decomposition due to exposure to high temperature. Pyrolysis takes place in inert atmosphere, specially $N_2$ or He, in order to avoid heterogeneous reactions between the solid and the gas medium. During pyrolysis, coal structure crashes causing the volatile matter release.

1.2 **Chemistry of pyrolysis**

Although a detailed analysis is not possible, a rough interpretation of the chemical reactions involved in pyrolysis can be given. At the lowest temperatures desorption takes place of molecules, held by physi- or chemi-sorption forces. At higher temperature pores are formed and, consequently trapped molecules can escape to the gas phase. These processes are more or less of a physical origin, except that the porosity at higher temperature is generated by chemical reactions. Therefore, it has to be expected that trapped molecules can be evolved up to relatively high temperature. At about 700K (heating rate <1000K/s), in the case of lignite, $CO_2$ is observed. This is associated with decomposition of carboxylic groups. In agreement with this, at these temperatures $CO_2$ is not observed for bituminous coals, obviously because they do not contain significant amounts of carboxylic groups. At slightly higher temperatures for lignites $H_2O$ evolution dominates. The major reaction probably is dehydration of OH-groups, leading to ether bridges.

Whereas for lignites O-containing products dominate, for bituminous coals tar and hydrocarbons are the major species observed. This is hardly surprising, considering the fact that bituminous coals contain much less oxygen than lignites. In bituminous coals the initiation reactions probably are mainly C-C bond breaking reactions because: (1) model
compounds, containing C-C groups decompose at about 900K, the temperature above which extensive tar evolution occurs, (2) bituminous coals contain many C-C bonds, Figure 1.

![Figure 1. Model of the chemical structure of a high-volatile bituminous coal](image)

At very high temperature reactions take place forming H₂ and products containing N and S. This can be well understood from the model compounds studies. The aromatic ring clusters containing these heteroatoms are very stable and decompose only at temperature, giving probably highly aromatic structures the poly aromatic hydrocarbons type. Furans have the lowest stability and consequently react most extensively. N- and S-aromatics are more stable. Therefore, it can be explained that chars heated to high temperature do not contain O, but do contain considerable amounts of N and S. It is noteworthy that the C/H ratio of chars produced at high temperature is in the order of 8, suggesting a higher degree of condensation than poly aromatic hydrocarbons. In this interpretation the initiation part of the radical chains is emphasized. Of course, also secondary reactions have to be considered. It is well known that in using packed-beds or applying the captive sample technique, volatile yields are lower for larger sample sizes. The reason is that secondary reactions are favoured in the case of large samples; the heating time is larger and residence time in the sample bed is larger. In the same way it can be understood why large particles give lower volatile yields than small particles.
1.3 Product distribution in coal pyrolysis

Product distribution is important for two reasons. First the economics of commercial processes strongly depend on the type and amount of products. Secondly, product distributions give fundamental information on the processes occurring during pyrolysis; in fact, they also considerably help in better understanding the structure of coals. A typical plot of the product distribution as function of the temperature is given in Figure 2, for a lignite at high heating rate. Up to about 700K, only water is produced. Subsequently the evolution of CO\textsubscript{2} and tar starts. At slightly higher temperatures CO, hydrocarbons and H\textsubscript{2} are observed. The hydrocarbons mainly consist of methane and ethylene. It is interesting that the production curves exhibit a two-step behaviour: CO\textsubscript{2} evolution increases enormously at about 700K, while subsequently, it levels off to an asymptotic value, but at 1050K suddenly an increased production takes place. CO and hydrocarbons show analogous behaviour. From a detailed analysis it appeared that this does not apply to H\textsubscript{2}: evolution starts at 800K and it increases continuously up to about 1150K.

![Figure 2](image-url)

**Figure 2.** Pyrolysis product distribution from Montana lignite heated to different peak temperatures [1] (heating rate >1000K/s).
Figure 3 gives the product distributions for a bituminous coal. Figure 2 and 3 give results, which can be compared directly with each other. It is obvious that the bituminous coal behaves quite different than the lignite. Whereas in the former case the product distribution is dominated by H$_2$O, CO and CO$_2$, in the case of the bituminous coal tar is by far the most important product. Figure 3 shows that tar production takes place over the whole temperature range. H$_2$O is formed at relatively low temperature, below 700K, whereas at higher temperatures no additional water is formed, in sharp contrast to the case of lignite.

**Figure 3**  Pyrolysis product distribution from Pittsburgh Seam bituminous coal heated to different peak temperatures (heating rate >1000 K/s).

Analysis of the chars produced in pyrolysis gives information, complementary to chemical analysis of the gaseous products. Figures 4 and 5 give results for lignite and a bituminous coal. In these figures the retention in the char is shown for C, H, N, S and O. It is clear that the char becomes relatively enriched in C: even at the highest temperature only about 20% of the original carbon is lost. The reason, of course, is that the volatile contain large amounts of
H₂O and H₂, while also the evolution of molecules like CH₄ and CO₂ lead to enrichment of C. The decrease of H compares well with the foregoing. At low temperature H₂O, which is adsorbed or trapped in the coal, is evolved, whereas at higher temperatures defunctionalisation takes place. N-loss does not start before 1000K and even at the highest temperature this does not exceed 30%. S-loss takes place over the whole temperature range. Compared to N less S remains behind in the char. O-volatilisation exhibits two stages: at 700K already about 40% of the O is lost, obviously due to H₂O and CO₂ evolution. At higher temperature oxygen volatilisation continuously takes place, while at about 900K a sudden increase in O-loss occurs. At 1300K virtually no oxygen is retained in the char.

![Figure 4. Elemental composition of lignite char for different peak temperatures (heating rate >1000 K/s)](image)

Figure 4. Elemental composition of lignite char for different peak temperatures (heating rate >1000 K/s)

Figure 5 gives analogous information for a bituminous coal. Comparing these results with the previous ones, it is clear that especially the C-profile is fundamentally different. Up to 50% of the carbon, originally present, is volatilised eventually. It is interesting that upon keeping the sample at 1275K, additional H-loss takes place, whereas the C-content of the char does not increase further. This indicates that more extended poly-aromatic hydrocarbon structure
is formed. Over the whole temperature range the carbon content decreases continuously. The H-profile shows an analogous pattern, which is in good agreement with the predominant evolution of tar, Figure 3.

**Figure 5.** Elemental compositions of bituminous coal char for different peak temperature (heating rate 1000 K/s); the dashed lines represent conditions at 1273 K during 2-10s

### 1.4 Effect of temperature

Most of the available data on coal pyrolysis were obtained at temperatures below about 1000°C, although some measurements at higher temperatures have been reported. Figure 6 illustrates how pyrolysis yields depend on temperature in the range up to 1000°C. The data of each investigator have been normalized to a yield of 100% at 1000°C. This rather arbitrary basis is convenient because the yields observed in this temperature range approach a higher temperature asymptote, which is closely approximated by the yield observed at 1000°C. Although Figure 6 supports the concept that the yield of pyrolysis products, for
reasonably long experimental times, becomes independent of temperature above about 1000°C, recent data obtained at higher temperatures, presented in Figure 7, suggest that additional devolatilisation in relatively short times may be observed by heating the coal to temperatures well beyond 1000°C. This suggestion seems reasonable because the absolute time required for a given degree of completion of devolatilisation is much less for the higher temperatures. For example, the heating of coal from 1000 to 1800°C might produce in fractions of a second an additional weight loss that at 1000°C could be produced only in times too long to be available in laboratory experiments. Furthermore the slower rate of escape of volatiles from the particles at a lower temperature might present a larger opportunity for secondary cracking reactions and carbon deposition, which could eliminate the possibility of ever attaining the weight loss achievable at a higher temperature. On the other hand, an equally plausible argument might be that an increased rate of cracking and carbon deposition at the higher temperatures might reduce the net weight loss to a value less than that achievable at lower temperatures, given enough time.

**Figure 6.** Effect of temperature on pyrolysis weight loss (yield) at different residence times. Curves: solid, Dryden [11]; short dashes, Shapatina et al.; long dashes, Anthony et al.; dots and dashes, Wiser et al.
The effect of time on the variation of weight loss with temperature, shown in Figure 6, can also be seen by the data of Kobayashi et al. [1], shown in Figure 7 (Pittsburg No.8, Montana lignite). To achieve coal residence times longer than those available in the laminar flow reactor, the same reactor was operated in a free-fall mode by allowing the coal particles to fall at their terminal velocities into either a cooled or an uncooled collector at the bottom of the furnace. The data in Figure 7 for a reaction time of 10 min represent a fall time on the order of 1 s and a soak period of about 10 min in an uncooled alumina collector. As in the case of the lower temperature data in Figure 6, the increase of weight loss with temperature shown in Figure 7 is much stronger for shorter reaction times. Thus as time increases, the effect of temperature on pyrolysis rate becomes less apparent, since the decomposition eventually approaches completion at any of the temperatures. Given enough time and the absence of secondary cracking and carbon deposition, the weight loss versus temperature curve would presumably be the horizontal line approximated in practice by the high temperature asymptote, which is defined above as the ultimate yield $V^*$. It is clear from Figure 7 that $V^*$ for a given coal unfortunately remains poorly established despite the availability of these high-temperature data.
Figure 7. Effect of higher temperatures on pyrolysis weight loss at different residence times. A, Blair et al. [7]; B, Kimber and Gray [4]; Kobayashi et al. [1]; D, Ubhayaker et al. [6] Residence times (where available) and coal seams indicated on curves

1.5 Effect of heating rate

The fact that rapid heating techniques in coal pyrolysis give substantially larger yields of volatiles than are obtained by the slow heating of coal in conventional packed-bed carbonisation retorts or standard volatile matter crucibles is sometimes attributed to an effect of the heating rate itself. The rationale is not proved by data, since the techniques used to achieve faster heating, such as small particles well dispersed in a gas or thinly spread within a wire mesh heater, promote larger volatiles yields by avoiding some of the cracking reactions and carbon deposition associated with the conventional methods. In a critical test of the rationale, Anthony et al. [2] varied only the heating rate, in the range 650-10,000°C/s, while holding all other variables constant in a pyrolysis experiment on both a caking and a non caking coal. The yield of volatiles did not change significantly; thereby indicating that heating rate is not important with respect to volatiles yield under the conditions studied. The
data of Suuberg [3] for the same two coals confirm this picture and extend it to include heating rates down to 270°C/s, and volatiles composition as well as yield. Thus yield increases sometimes attributed to heating rate probably result primarily from the associated experimental conditions employed to achieve the faster heating.

The data of Kimber and Gray [4,5], Ubhayaker et al. [6], and to a smaller extent Kobayashi et al. [1] indicate that increased volatile yields may result from extensions of heating rate and final temperature to values above 100,000°C/s and to the range 1800-2000°C, respectively. Since heating rate and final temperature were not varied independently in these studies, the results do not resolve the separate effects of these two variables. However, the same results in combination with the high-temperature but lower heating rate data of Blair et al. [7], shown in Figure 7, support the view that temperature, not heating rate, is primarily responsible for the higher yields.

Heating rate does have a clear effect on the temperature-time history of pyrolysis. Thermogravimetric measurements of Van Krevelen et al [8] showed that as coal is heated, the temperature at which the rate of devolatilisation assumes its maximum value increase with increasing heating rate. Furthermore the maximum rate of devolatilisation was found to increase approximately linearly with heating rate.

Figure 8 shows the effect of heating rate on the temperature dependence of the extent of devolatilisation. It can been seen, for example, that the temperature attained before devolatilisation reaches 10% completion is 400°C at a heating rate of 1°C/s and 840°C at 10^5°C/s. For 90% completion the coal reaches 860°C at 1°C/s and 1700°C at 10^5°C/s. Figure 9 shows how the rate of devolatilisation depends on heating rate and temperature.
Figure 8. Effect of heating rate on weight loss at different temperatures.

The maximum rate increases somewhat less than linearly with heating rate. As the heating rate is increased, not only does the temperature of the maximum rate shift to higher values, but also a given fraction of the devolatilisation occurs over a wider temperature range. Thus, the temperature range traversed by the coal in going from 10 to 90% completion of devolatilisation increases from 400-840°C at 1°C/s to 860-1700°C at 10^5 °C/s. This behaviour reflects the dependence of the amount pyrolysis occurring at a given temperature on the time spent at that temperature, hence its dependence on heating rate. For the same reason, heating rates affects the cumulative amount of devolatilisation occurring before a given temperature is attained, which in turn affects the instantaneous devolatilisation rate at that temperature, since the rate is first order in the amount of hydrolysable material remaining. These effects of heating rate on the temperature –time history of pyrolysis can clearly influence the fate of the pyrolysis products in situations where secondary reactions are important. Furthermore, heating rate, through its effects on rate of volatiles evolution, would presumably affect the
partial pressure of the ambient gas at or within a particle undergoing devolatilisation. Such behaviour has practical implications regarding the achievement of intimate contact between paralysing coal particles and reactants in the ambient gas.

![Figure 9](image)

**Figure 9.** Effect of heating rate on rate of devolatilisation at different temperatures.

### 1.6 Effect of pressure

It has been known [9] that the pyrolysis of coal under reduced pressure results in larger weight losses, the main part of the increase being in the tar fraction. The behaviour id generally attributed to secondary reactions, including cracking and carbon deposition, which can convert some of the tars to both heavier and lighter species. These reactions are less favoured at lower pressures owing to the smaller resistance to volatiles flow out of the coal particles.

Data from Pittsburgh Seam bituminous coal heated at different rates to 1000°C are given in Figure 10. A certain amount of carbon deposition from cracked volatiles occurred on the
stainless steel screen sample holder, particularly at the lower pressures. Since weight loss was determined by weighting the screens before and after the run, the deposit reduced the observed weight loss by an error that was measured by weighting the screen without the coal residue. The corrected weight loss is represented by the dash curve.

Figure 10. Effect of pressure on weight loss in the pyrolysis of Pittsburgh Seam bituminous (Ireland Mine) coal. Final (holding) temperature, 1000°C; experimental time, 5-20s; mean particle diameter, 70µm; atmosphere, helium; nominal heating rate (°C/s), 10,000 (−), 3000 (4), 650-750 (4). Curves: solid, uncorrected for cracking deposition on sample holder; dashed, corrected for cracking deposition on sample holder.

Weight loss is seen to decrease monotonically with increasing pressure, presumably reflecting an increasing extent of cracking and carbon deposition within the particles. The weight loss under atmospheric pressure is 50% of the as-received coal (ash=12.2%; moisture=1.65%), but the loss is 57.2% under vacuum and 37.2% at 100 atm. Furthermore, the trend suggests asymptotic behaviour at the pressure extremes, but the scatter of the data precludes a firm location on the asymptotes. Such behaviour would be expected theoretically. The pressure inside the particles during devolatilisation probably becomes independent of the external pressure as the latter is reduced below a certain value, hence the low-pressure asymptote. The approach to a limiting yield at high pressures probably reflects the depletion of species that are susceptible to the secondary reactions. Accordingly, the weight loss
achieved at the high-pressure extreme is the yield of nonreactive volatiles, and the additional weight loss available by reducing the pressure to vacuum represents reactive volatiles.

Figure 11. Effect of temperature on weight loss from Pittsburgh Seam bituminous coal heated in inert atmospheres at different pressures. Coal: VM=39.8%, moisture=1.6%; nominal heating rates, 65-750°C/s; mean particle diameter, 70 µm; residence times, 5-20s. Heated in helium (X) and nitrogen (8), 1 atm; helium (–), 69 atm; two-step heating helium (2), 69 atm.

The variation of the pressure effect with temperature is shown in Figure 11 for the same coal as above [10]. Two points are obvious: the effect of pressure appears only above a certain temperature, here about 600°C. The high-pressure yield becomes roughly independent of temperature above the same temperature. In view of the foregoing interpretation of the pressure effect, these points imply that volatiles formed below a certain temperature are effectively inert to secondary reactions under the temperature, time, and other conditions prevailing in the particle, whereas most of the additional volatiles formed at the higher temperature are reactive. Although this picture is quantitatively useful, it is clearly an approximation from a mechanistic point of view. Furthermore, the critical temperature here found to be 600°C is a function of both heating rate and time spent at the final temperature.
Similar experiments were performed with Montana lignite. The effect of pressure on weight loss was too small to be interesting in comparison with the foregoing behaviour.

1.7 Effect on particle size

Although particle size has been varied in several studies, the results are often inconclusive because of failure to use the same temperature, or temperature time history if time is small, for all particle sizes. Even small temperature differences associated with different particle sizes can cause larger changes in volatiles yield that would result from the particle size change at constant temperature. The problem is especially difficult because the effect of increasing particle size is a small to negligible decrease in volatiles yield, whereas this direction of particle size change usually results in lower temperature, owing to the slower heat up of larger particles, and hence lower yields.

![Figure 12](image)

**Figure 12.** Effect of particle size on weight loss from Pittsburgh Seam bituminous coal heated in helium. Ash=12.2%; moisture=1.6%, 1000°C final (holding) temperature, 5-20s residence time, 650 –750°C/s nominal heating rate, 1 atm helium.

Anthony systematically varied particle size over the range 53-1000 µm in pyrolysis experiments with the wire mesh heater and an atmosphere of helium [10]. The effect of increasing the particle size of Pittsburgh Seam coal over this range was a decrease in weight
loss of about 3 wt % (as received) of the original coal as shown in Figure 12. This behaviour apparently reflects an increased extent of secondary reactions and carbon deposition for larger particles, which is consistent with the view that larger particles offer more resistance to the escape of volatiles. In contrast to the bituminous coal, Montana lignite exhibited no significant effect of particle size within the stated range.

The rapid heating rate, high-temperature study of Ubhayaker et al [6] involved the use of two size fractions, +200 mesh and –200mesh of pulverized Pittsburgh Seam bituminous coal. The mass mean diameters of the two fractions were 100 and 20µm, respectively. The yield of volatiles from the fine fraction exceeded that from the coarse fraction by an amount that decreased with increasing residence time. When the particles were heated in a gas that was fed at a temperature of 1700°C, the difference in volatiles yield was 14 wt% (maf) of the coal at 7 ms, 7% at 25ms, and 5% at 70ms. This trend indicates the existence of a heating effect whereby the difference in yields at a given residence time within the range above is due at least in part to the requirement for longer heating times for the larger particles. Consequently the effect of particle size per se must have been less than 5%, as found for the longest residence time. In fact, extrapolation of this trend indicates that the difference would become negligible compared with the data scatter if residence time were increased to about 100ms. The occurrence of no significant particle size effect in the range 20-100µm would be consistent with the data in Figure 12.

1.8 Effect of coal type

Only fragmentally information is available on the effect of coal type on weight loss in pyrolysis. According to the Dryden correlation [11], Figure 6, the temperature at which a given fraction of the final weight loss is achieved decrease by about 50°C for each 10% increase in volatile matter content. Although this picture does not appear to have been tested critically in literature, it is consistent with the data of Anthony [10]. Approximately the same relationship between the fractional attainment of the final (1000°C) weight loss and temperature was observed for a bituminous coal and lignite having essentially the same volatile matter contents.
One of the most striking differences in the weight loss behaviour of different coals is the potential for volatile yields to exceed the proximate volatile matter content when conditions are employed that avoid extensive tar cracking and carbon deposition of particles. This potential correlates with tar yield, is small to negligible for both low and high rank coals and strong for coals in the bituminous range. The effects of pressure and particle size discussed above reveal another, but probably related, difference in behaviour. Although weight loss from Pittsburgh Seam bituminous coal decreases with increase, in pressure or particle size, weight loss from Montana lignite is relatively insensitive to both these variables. Therefore, according to the interpretations given above, the extent of secondary reactions in the lignite seems to be relatively independent of changes in the resistance to volatiles escape from the particles. Unfortunately additional weight loss data don’t indicate whether the primary lignite volatiles are: (1) extremely reactive (thereby permitting the completion of secondary reactions even for the lowest pressures and smallest particles studied) or (2) so inert to secondary reactions that almost no carbon deposition occurs, even for the highest pressures and largest particles studied.

Jones [12] discusses the concept that certain reactive aromatic units formed in pyrolysis can either take up hydrogen and be evolved as tar or recondense to form semicoke. Hydroxyl or other reactive oxygen groups can act to favour the latter route by consuming hydrogen that would otherwise promote tar evolution, or they can increase coke formation directly through cross-bonding. In view of the much larger oxygen content of lignite, such behaviour would appear to be more prevalent for lignite that for bituminous coal. If the secondary reactions thus indicated are fast on the time scale of pyrolysis experiments, the extent of coke or semicoke formation associated with their occurrence would not change within the range of experimental conditions. Since the reactions of the oxygenated groups may indeed be fast, alternative 1 stated above seems reasonable.

Another measure of the difference between the weight loss behaviour of Montana lignite and Pittsburgh Seam bituminous coal comes from a comparison of the augmentation in volatile yields observed by increasing the temperature or decreasing the pressure from a base case of
1000°C and 1 atm [2]. Whereas for the bituminous coal the increase of volatiles yield affected by increasing the temperature to 1800°C [1], is approximately the same as that achieved by lowering the pressure to 0.001 atm, the increase of yield from the lignite was relatively insignificant when the low pressure was employed but was comparable to that of the bituminous coal when the high temperature was used. In terms of the foregoing rationalization based on the supposed role of oxygenated groups, these observation support the view that the char promoting reactions of the abundant oxygenated groups of lignite at the higher temperatures may not be as dominant over aromatic stabilization by hydrogen as they seem to be at 1000°C or less. Accordingly, pyrolysis of the lignite at the higher temperature may avoid some of the char-forming secondary reactions that are relatively unaffected by physical augmentation of volatiles escape, such as reduction in pressure or particle size, at lower temperatures. However an alternative explanation that is also consistent with the data is that the effect of the higher temperatures on weight loss from the lignite reflects a larger extent of primary decomposition that is not observed at the lower temperatures because of insufficient residence times. Although this explanation cannot be ruled out for the lignite, it is in conflict with the bituminous data, since the high-temperature weight losses are not significantly larger than those available at lower temperatures through the use of reduced pressure.
2. Assessment of LTC Advantages, Economic and Technological Development

Economic and market prospects of Low Temperature Carbonisation technology can be identified by evaluating markets and market developments in detail. Results are quite promising, since such fuel pre-treatment technology fits into the known and predicted European, North American and global market developments, fulfils technical and cost efficiency demands, and meets International Protocols and Agreements.

From 2000 to 2020, high- and medium-sulphur coal production is projected to decline by 0.2% per year, and low-sulphur coal production is projected to rise by 2.0% per year. Over 90% of world coal reserves are classified as high sulphur coal, while the low sulphur coal mines are located outside of Europe. This means an economic dependence of the low sulphur coal, while Kyoto Protocol provides clear GHG emission goals to reach. As a result of the competition between low-sulphur coal and “end-of-pipe” solution (post-combustion sulphur removal), it is expected that the cost for both low sulphur coal and post sulphur and HAPs removal treatment will significantly increase. Changes in industrial energy intensity (consumption per unit of output) can result in either underlying increased demand in equipment and production efficiency, such as LTC pretreatment or from structural changes in the composition of manufacturing output, such as “end-of-pipe” solution. Since 1970, the use of more energy-efficient technologies, combined with relatively low growth in the energy-intensive industries, has dampened growth in industrial energy consumption. In any case, the demand for environmental protection issues, especially for control of hazardous air pollutants and GHGs, and reuse of residual materials has been strongly increased during the past decade, resulting in increased need of high quality clean energy production.

Among key-points of the economic development and scientific/technological prospects of the Low Temperature Carbonisation is that the market tendency is the expansion of small- and medium-sized power generation plants, i.e. less than 150 MW capacity, which are estimated to be at least 35% of the total solid fuel energy generation market. Traditional plants can
easily be retrofitted by LTC add-on module, whereas old plants can cost efficiently upgraded and still meet the new environmental norms and regulations.

Potential LTC project development offers economical benefits, such as:

- Implementation of LTC technologies can result in total cost (including amortisation and O&M cost) decrease of the end-product Clean Coal by at least 10% compared to presently best offer of low-sulphur content coal
- Cost savings may be realised from environmental control at the “end-of-pipe”
- Efficiency improvement will enable profit increase by at least 3%
- Renewable utilisation, such as agricultural wastes, may decrease raw material cost by even more than 10%.

Major technical and scientific benefits of LTC technologies employment include:

- Preventive environmental protection
- Improved boiler efficiency
- Total cost savings that may exceed 10%
- Reutilisation of industrial residuals since flexible multi fuel operation is possible
- Enhancement of fuel diversity and security
- Elimination of environmental and health care risks
- Reduced boiler corrosion, resulting in maintenance cost savings

Most of the operational problems with alternative fuels arise from the lower fuel quality, such as (a) high temperature corrosion due to high Cl-S content and (b) slagging due to high alkali content (Na, Ca, K). These fuel constituents reduce the ash softening point and cause slagging in the combustion chamber and fouling in heat exchangers. Preventive improvement of solid fuel quality and production of cleaner blends could be an alternative to utilise alternative raw material and residual streams. Therefore, one of the major LTC advantages is fuel availability, since it offers potential utilisation of alternative biomass and food industry wastes, whose disposal may present today high cost and technical deficiency, such as:

- Waste vegetable oils (co-combustion)
• Non usable – non recyclable residuals from bio-diesel production
• Biomass residues, such as straw pellets and husk
• Energy crops, such as short rotation wood (willow) and sawdust.
• Biomass waste, such as demolition wood
• Meat meals from animal by-product processing industry, etc.

Among known limitations that LTC technologies present are (a) reduced efficiency in treatment of feed streams with very high moisture content and (b) removal of all heavy metals from low grade fuels.

Development of R&D projects and future implementations may be beneficial for most countries:

• **EU Accession Countries:** the technically and cost efficient Clean Coal management will be a critical issue for the EU Accession Countries, particularly for countries with large coal reserves and significant renewable potential (such as biomass potential)

• **EU Countries:** Many countries will face increased problems to fulfil Kyoto Protocol targets, while the application of new EU environmental directives are imposing more strict restrictions concerning the environmental performance of the power generation. Development of such technology will improve the raw material quality by preventive means, offering an increased and direct protection of the environment, water resources and the human health. It may also result in promotion of International Emissions Trading (IET) to allow emissions trading between developed countries and of Joint Implementation (JI) to allow emissions trading by projects between developed countries.

• **East European and Asian Countries:** LTC is a technology with good industrial export potential. Reduction of hazardous air pollutants in power generation will further lead to the improvement of the global climate. Such results are promoting the application of the Clean Development Mechanism (CDM) that allows developing countries without reduction targets to benefit from emission reduction projects, while developed countries may obtain emission reduction credits by participating in such projects.
In conclusion, adoption of LTC technologies offers significant advantages, among which are:

(1) It is a preventive option that compared to “end-of-pipe” solutions results in:
   a. Guaranteed and competitive long-term clean energy supply
   b. Providing compatibility between development of open – liberalised energy market and the objectives of the sustainable environmental protection
   c. Providing overall cost savings for sustainable clean energy production

(2) Improved overall safety and recycle-reuse of industrial material streams

(3) Flexible feed choice application from regionally available feed supply by converting trash (low grade fuels) into cash (clean energy)

(4) Improving overall cost efficiency for sustainable green energy production

(5) Supporting schemes for reduction of greenhouse gases as per Kyoto Protocol and European Commission commitments
References