

Automated Coke Petrography

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At ISCOR the quantitative determination of the structural properties of coke is done with a Leitz-TAS Image Analysis System. A reflected light microscopic image of a coke sample is converted into a binary image and stored in computer memory or on hard disc. By means of mathematical and stereological techniques the system can transform the images electronically. A lightpen can be used to determine the sequence of these operations. TASIC, which is based on BASIC language, is used as the program language on the computer. The majority of the programs are written by the user to ensure satisfactory results for specific studies.

The computer controls the whole system, including the movement of the microscope stage, focusing, transformation and measurement of the image and data processing. The computer is also applied for establishing statistical correlations between the structural and other coke and coal parameters as well as for graphics and predictions of the coke structure.

Factors affecting the structural properties of coke are discussed and conclusions are drawn.

Introduction

The carbonization of coal

Coke is produced by heating finely crushed coking coals in airtight gas heated coke ovens. The charge mostly consists of blends of different coking and in some cases also partially briquetted coal. After carbonization, which lasts approximately 18 hours, the coke is pushed from the ovens and quenched with water. This hard and porous carbonaceous product produces the heat, acts as the reductant for the reduction of iron ore and supplies a permeable support for the burden in the blast furnace. Although coke is expensive, it is still indispensable for the production of liquid iron in a blast furnace. In order to minimize costs cheaper coals are added to a blend, and special processes such as partial briquetting are introduced to obtain coke

of acceptable quality for blast furnace practice.

Factors affecting the quality of the coke

Since the quality of coke affects the performance of the blast furnace, factors affecting coke quality such as coal properties, coal screen size and carbonization conditions must be controlled. Coal properties include the petrographic properties such as maceral composition and rank; the rheological properties such as maximum plasticity, temperature range of plasticity, contraction, dilatation, free swelling and Roga index; the chemical properties such as moisture, ash, volatile and sulphur content; and the screen analysis of coal. The coke properties include the screen analysis; the cold and hot strength; the

reactivity with CO₂; and the petrographic properties such as structure and texture. Structural¹ properties include the porosity, cell wall and pore sizes while texture² includes the carbon forms in the coke. The carbonization conditions affect the rate of carbonization and can be controlled with oven temperature and soaking time.

The structure of the coke

In this paper the structural properties of the coke are examined in depth. The structure of the coke develops during the softening and resolidification stages. During the first phase of softening, pores develop in the vitrinite maceral (component of coal), the exinite maceral decomposes whereas the inertinite maceral still possesses a higher reflectance than that of the plastic vitrinite. Microscopical observations have shown that the temperature increases and the relief of the inertinite decreases until it is the same as that of vitrinite. Further devolatilization causes tension and contraction which results in the formation of cracks.

According to Stach,³ there are several factors affecting the formation of the structure of a coke prepared from a specific European coal, the first being the particle size of the coal. A large percentage of the plus 3 mm particles gives rise to foam-like structures whereas particles smaller than 0,2 mm tend to yield smaller and fewer pores. The second factor is the volatile matter. The width of the plastic zone varies in the coke oven, and the largest pores develop in coal with 23 to 32% volatile matter and abundant pores in coal with 15 to 20% volatiles. The third factor is the amount, size and distribution of the macerals. Pores only

develop in the vitrinite, reactive semi-fusinite and exinite. The distribution of the inertinite also has an effect on the swelling of the vitrinite.

Other factors that affect the formation of the structure are the variation in the coal properties from a specific coal source or from different coal sources, and the effect of individual coals on each other with blending. The use of additives such as pitch or the application of processes such as preheating and partial briquetting of the coal do affect the structure of the coke. Coking conditions such as the rate of carbonization and the position in which the coke was formed in the coke oven are also important.

The net effect of above-mentioned factors on the structural properties of the coke can only be quantified when statistical measurements of the properties are obtained. In this paper the effect of variations in coal composition, the use of partially briquetted coal and pitch additions to coal are examined in depth.

Determination of the structural properties of coke

The determination of the structure of the coke quantitatively is a time-consuming task. The sample preparation involves the crushing and splitting of a large representative sample and the preparation of resin-impregnated polished sections. In the past the porosity was determined microscopically by means of point counting, and the cell wall and pore sizes by means of a calibrated eyepiece under reflected light. However, with the automation of the measurements with a flexible programmable software-controlled image analysis system, such as a Leitz-TAS presently used, more accurate statistical results are possible in a fraction of the time.

The principle of operation of the image analysis system

An optical image is obtained by a microscope (Leitz Orthoplan) from which a reflected image of a polished block is converted to a electronic image through a Plumbicon television camera. The amplitude of the electrical pulses of the electronic image is a function of the light intensities of grey levels, determined by the reflectance of the different phases in the optical image. An analogue of the electronic image is transmitted to a discriminator, which contains a densitometer where the image is divided into a matrix of approximately 400 000 picture points on a hexagonal point raster. Each of these points consists of an electrical signal. The densitometer divides the amplitudes of the electrical signal into intervals of grey level bands on a grey scale with 100 intervals. The grey spectrum is adjustable according to the brightness of the image. A phase (A) in the optical image can thus be extracted from the image by adjusting the threshold and band width of the grey scale so that it contains all the grey level intervals of that phase.⁴ With the use of the hit-or-miss transformation of the geometrical set theory, a value 1 can be assigned to every point (x) in the image that is located in the selected phase (A) by the selected grey level band, and a value of 0 if it does not belong to the selected phase (A^c). The image is thus divided into two sets of points which can mathematically be formulated as follows:

Let $K_A(x)$ be the indicator of set A

$$K_A(x) = 1 \text{ if } x \in A$$

$$K_A(x) = 0 \text{ if } x \in A^c$$

A binary image is thus created, which can be stored in the computer's memory.

Before the image is analysed the image is pretreated and modified by means of

transformations and interaction operations. Basic measurements such as area, perimeter, number of objects and projections through hard-wired functions as well as analysis according to Boolean logical functions such as intersections and unions can thus be done. Measurements of variable field data, in which the whole field is treated as a entity or individual particles, are done automatically or can be manually selected by the operator with a lightpen. The value obtained with field data is expressed as a total value for all the detected features in the field, and a mean value is determined for the field of view. The lightpen can also be used to modify the image before measurement and the determination of the sequence of the transformations. It can also be applied for basic hard-wired measurement by means of touching the sensfield next to the symbol as indicated around the screen of the monitor.

The users' programs are easily written and verified in the high-level programming language TASIC, which is similar to BASIC. It allows control over the entire system and data processing of the microcomputer. Operations such as stage movement, image focusing, phases selection, image pretreatment, basic and complex measurements, data processing, formatting and output can be programmed.

Basic measurement and transformations as used in image analysis of coke

Basic measurements

The area is defined as the number of picture points contained in the selected phase or measuring field.

Projection is defined as half the number of intersections between the selected phase and the line system in any one of the three major directions of the hexagonal raster.

The horizontal direction is used to measure the mean chordlength.

Elementary transformations

A structuring element B, which consists of a certain number of adjacent hexagonal picture points (a hexagon fills space in all directions when expanded) can be moved across the image by the setting of its center on all successive points of the hexagonal grid. If the centre of B is in any point x of the measuring raster, the structural element belonging to it is designated Bx. By means of elementary transformations (i.e. erosion and dilation) accurate information can therefore be generated regarding a selected phase A.

Erosion is the set of all image points on which B is centered for which the conditions $Bx \subset A$ holds (Bx contained in A), defines a new image of A, called the eroded image of A with respect to B, and is denoted as $A \ominus B$.

The net result is an overall shrinkage of the image. The images that cannot contain the structuring element are eliminated.

Dilation can be defined as $Bx \cap A \neq \emptyset$ (intersection between Bx and A is not empty) which gives the opposite effect of erosion, denoted as $A \oplus B$.

Sequential transformations

The application of opening is size analysis in which the size of the structuring element is the sieve size used at the time. Opening can also be applied in image cleaning to remove artifacts and bridges between grains.

Opening consist of erosion followed by dilation.⁴ The erosion of the image (selected phase A) with structuring element B gives an eroded image of A and removes all images smaller than the structuring element (B). The dilation of the eroded

image of A with the same structuring element B creates a new image of A, namely AB ($AB = (A \ominus B) \oplus B$). This image is very similar to the original image except that all the images smaller than the structuring element have been removed. A number of sequential erosions can be followed by the same number of dilations.

Reconstruction

Reconstruction is used to obtain only those particles marked with a lightpen or by means of erosion. It is a very useful transformation for preparation of the image, where artifacts can be removed without changing the major image.

Negation

Negation is given as the change of all points assigned with the value of 1 to 0 and those with 0 to 1.

Stereological principles used in image analysis of coke

In order to apply image analysis correctly a knowledge of stereology is necessary. Stereology is a mathematical technique which includes stereometry (measurement in three directions) and morphology (the study of shape). Image analysis of coke is carried out on a two-dimensional plane section, and measurements made on such an image are not necessarily directly related to the dimensions of the original three-dimensional object. Stereology can be used to deduce the quantitative information of a three-dimensional object from that of the two-dimensional section.

According to Underwood,⁵ the following stereological principles are used in the measurement of volume percent:

$V_V = A_A = P_P$
where: V_V m³/m³ volume fraction - volume of features per unit test volume,

A_A m^2/m^2 area fraction - area of intercepted features per unit test area,

P_p point fraction - number of points per total number of test points.

The relationship holds if the component is distributed randomly and the section can be considered as being representative. In the case of the image analyser the area is measured by counting the amount of picture points set to 1 (selected phase) per total number of points of the measuring field.

The projection measurement used in the mean chordlength can stereologically be expressed as:

N_L m^{-1} number of interceptions of features per unit length of test line.

Although mathematical and empirical methods do exist to convert particle size distributions observed in plane sections into three dimensions, the comparison of quantitative results in two dimensions can give valuable information of the size differences between samples.

Hardware and software facilities

The choice of hardware and software for the system was made to suit the requirements for coke analysis. The user programs supplied had to be altered and combined after commissioning of the instrument. The simplicity of the TASIC language in which the programs were written facilitated this task, but it was still a slow system and, together with the slow magnetic tape secondary storage medium, the measurement and data storage were unacceptably slow. After a number of years a faster RT 11 compiler was introduced to improve the software speed, and a Winchester hard disc (8,9 MBytes) and a double-sided floppy disc were obtained to improve the program and data storage as well as the recall speed.

A further improvement, namely a FORTRAN compiler for even faster software speed, will be commissioned shortly. A disadvantage of the FORTRAN programming is that it is not quite as simple as the TASIC programming.

Sample preparation

Coke is a rather heterogeneous substance owing to the different macerals present in the coal, differences between individual coals in a blend and varying carbonization conditions. A relatively large sample therefore is needed for the purpose of sampling, preparation and measurement. A representative sample of 50 kg coke is progressively crushed down to -15 +10 mm from which a representative sample of approximately 30 pieces is obtained.

Prior to the use of this method it was standard practice to analyse 6 half-oven width pieces of coke sampled at approximately the middle (horizontally and vertically) of the coke oven. Each piece was subdivided into smaller samples for micrometric analysis. Results on the half-oven width pieces were compared with those obtained on several sets of the -15 +10 mm pieces. It was found that the latter yields statistically more reproducible results.

The -15 +10 mm pieces are mounted in white epoxy to eliminate internal reflections. The blocks are ground to obtain a flat cross-section of the coke pieces. The porous surfaces are then vacuum impregnated with white resin. Before setting, the excessive resin is pressed out on a flat plastic surface and left to harden. The sample is then ground down very carefully while great care is taken not to grind away too much of the impregnated surface. Removal of excessive impregnated material necessitates a

repetition of the impregnation procedure. Thereafter the samples are polished on a Texmet cloth and a slurry of water and Alpha alumina. After the polishing stage all the samples are microscopically scrutinized for signs of relief, scratches and broken cell walls. The occurrence of broken or discontinuous cell walls necessitates further grinding and impregnation. To ensure the best microscopic image for detection great care must be taken to obtain a well prepared surface.

Measuring procedure

A microscopic image of the polished surface is obtained with a 5X objective and a 4X condenser lens which give a 2,32 mm wide data field for measurement.

The intensity of illumination of the microscope is set according to the brightness and zero level balance which is calibrated in relation to the black-white level of the coke.

The scanning stage is programmed to cover the whole piece of coke in consecutive fields of 2,4 mm by 2,4 mm. About 17 fields per piece, depending on the size of the piece and a total of about 500 fields per 30 pieces (a single sample) are analysed.

The automatic focusing is programmed for the specific magnification being used, and manual focusing is only done on the first microscopic image.

The image consists mainly of two phases, namely lighter, yellowish carbonaceous and pyrite/pyrrhotite material, and the darker grey phase of the minerals, resin-filled pores and black unfilled pores. The mineral matter is usually finely distributed in the carbonaceous phase and can therefore be mostly grouped with the carbonaceous phase by means of electronic

transformations of the image (cleaning of image). In cases where the mineral matter is not finely distributed it is grouped together with the resin-filled and unfilled pores.

To obtain a binary image of the optical image a grey scale band level and band width is set to include all of the carbonaceous and pyrite/pyrrhotite phase.

Measurements on the computer-stored image can be done one by one while the operator takes care that the grey level is correctly set for each field and that each field is free from defects resulting from the preparation. Care must be taken that measurements are carried out on coke and not on the resin material binding two adjacent pieces of coke.

The two-dimensional size analysis, which applies the opening transformation, is however a time-consuming measurement. A fast hard-wired facility catering for this measurement is on the other hand too expensive. A better solution for the improvement of speed of analysis is to store the images initially on a large and fast Winchester secondary storage hard disc. One binary image requires approximately 8192 bytes. It takes the operator more or less 30 minutes to store all the data fields of 10 coke pieces in 10 data files. After storage of the images the measurement program is implemented and images are automatically recalled and measured. Measurements can therefore even be carried out after office hours without the assistance of an operator.

The first stage of the program modifies the image by means of negation, erosion and reconstruction, as well as negation and together with opening in order to close the small artifacts in the carbonaceous phase. These are caused by the cell structure of inertinite, finely disseminated minerals

and pitted polished surfaces. A more representative image is thus created for measurement.

The porosity of the coke is obtained by measuring of the mean area of the pores per test area, according to the formula:

$$\text{Porosity} = \frac{\sum \text{Area of pores}}{\sum \text{Area of data fields}} \times 100$$

The size analysis of the cell walls and pores can be done with two methods, namely the two-dimensional hexagonal structuring element method and the mean chordlength method. The structuring element method is programmed with the use of a loop, in which the area of the selected phase is measured and the values are stored in an array after nil, one, two, etc. openings. The loop continues until the structuring element thus formed is bigger than any structure in the image. All the area measurements for all data fields of a piece of coke are accumulated, mean values are calculated and the values are printed for control purposes. Subsequently all measured data are accumulated automatically for all the pieces of coke of a sample, and differential area distributions for nil, one, two, etc. openings (equivalent of 18,1 μm , 36,3 μm , etc. hexagonal structuring element) are plotted. A mean size can then be calculated from the distribution.

The mean chordlength method employs the projection measurement in the horizontal direction.

The values obtained are calculated as follows:

$$\text{Mean chordlength} = \frac{\sum \left(\frac{\text{area}^*}{\text{projection}^*} \right)}{(\sum \text{data fields}) (\text{width of data field}^+)}$$

* Already defined

+ Depend on magnification

No size distribution is possible with this method, and orientation of the structures can play a role in the values obtained. For coke the structures are randomly orientated, and the mean values obtained do not differ significantly for different orientations.

For the above-mentioned measurements the number of fields per piece of coke are taken in consideration and weighed mean values are calculated. Standard deviations are obtained for each value. Standard error of mean, 95% confidence limit and relative accuracy are calculated on the porosity values and standard deviation, number of classes, class width and percent variation are calculated on the hexagonal structuring element size analyses.

All the analytical values obtained in addition to the blend compositions are stored in datafiles to form a databank. In addition, other information such as coal and other coke properties is also stored in the databank. Linear and curve correlations and graphics can thus be obtained from the stored data with the aid of a self-compiled TASIC program. Choices are made with respect to specific properties or parts of the data and type of graphics required during the use of this program. Since the image memory is used to create the graphs, good accuracy of the plotted data can be obtained. With the aid of the KED Editor, which can be used as a word processor, subscripts are added to the printed graphs.

Predictions, in which extensive data and complex statistical techniques are used, are done on a mainframe computer.

Reproducibility

Reproducibility of results is tested by means of the z-test:

$$z = \frac{\bar{x}_1 - \bar{x}_2}{\sqrt{\left(\frac{s_1}{n_1}\right)^2 + \left(\frac{s_2}{n_2}\right)^2}}$$

where \bar{x}_n = mean value of one sample,

s_n = standard deviation of one sample,

n_n = number of values used to obtain mean value.

If $-1,96 \leq z \leq +1,96$ a meaningful difference, between the mean values exist, at a 95% confidence limit.

Good reproducibilities were obtained for results on a single sample, different samples taken from the same coke oven and mean values of +5 samples of coke produced from the same coal blend over a period of time in the same and different coke ovens of the same battery. However, differences are common in the results of one or more structural parameters between samples of coke produced from the same blend over a period of time.

Typical results obtained on the structural analysis of coke

Relationships

During extensive pilot plant trials, ten different coals were carbonized individually and 48 blends each consisting of two of the coals with blend compositions of 33/67% or 67/33%. Good linear relationships were found between the results obtained with the two different methods of measurement of cell wall and pore size. The porosity of the coke also correlates well linearly with the mean cell wall size.

It was furthermore found that the maceral composition of a coal shows the best correlation with the porosity and the mean cell wall size of a coke (Figure 1). Other coal properties, however, show poor or no correlation with these structural properties. The maceral composition therefore plays a most important role in

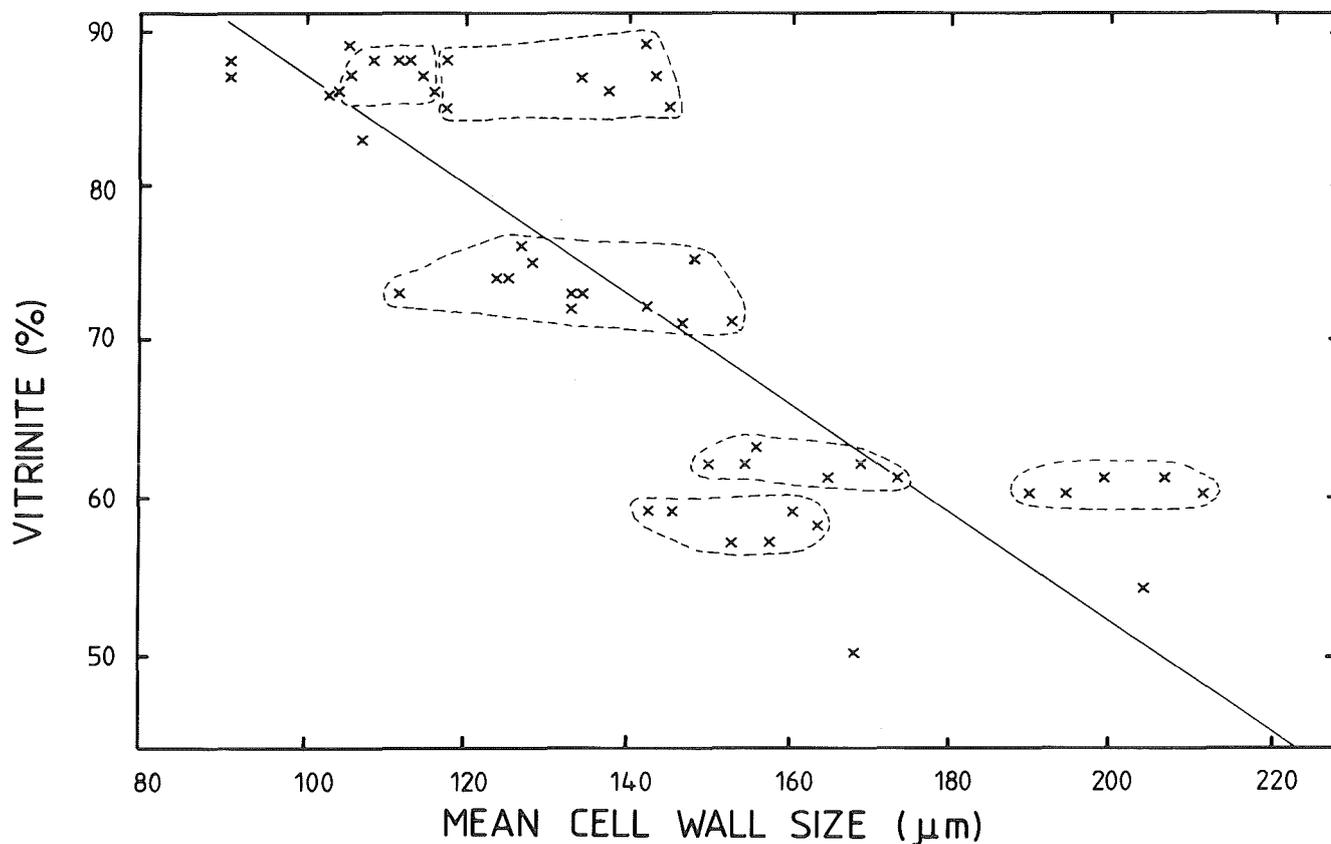


FIGURE 1. Relation between vitrinite content of single coals and their blends and mean cell wall size (thickness) of corresponding coke

the formation of the structure of a coke when the screen size of the coal and carbonization conditions are kept more or less constant. The combined effect of the other coal parameters on the structure of coke such as rank, maximum plasticity, temperature range of plasticity and free swelling index is also important. For example, a blend comprised of 3,5 to 6,5 ratio of two different coals formed a denser structure than is the case with coke prepared from the two individual coals, coked on their own. Both coals are vitrinite-rich. The former component, having a low plasticity and a limited range in plasticity temperatures, was enveloped by the latter component which is characterized by a high plasticity and broader range of plasticity temperatures (Figure 2).

No correlations could be found between the coke properties which are normally

tested and the structure of the coke. The structural properties thus provide additional information on the coke and can be applicable in the understanding of blast furnace behaviour.

The fact must be stressed that the given results can only be applicable to blends of coals with these specific coal properties. Coal from different sources and different blends may act differently owing to the complex interactions between the coal properties during carbonization.

Predictions of the structural properties from coal properties are possible. The testing of the validity of these predictions for a wider range of coal composition and blends is still in progress.

Partial briquetting and pitch additions

It was found that coke prepared from either partially briquetted coal (Figure 3) or

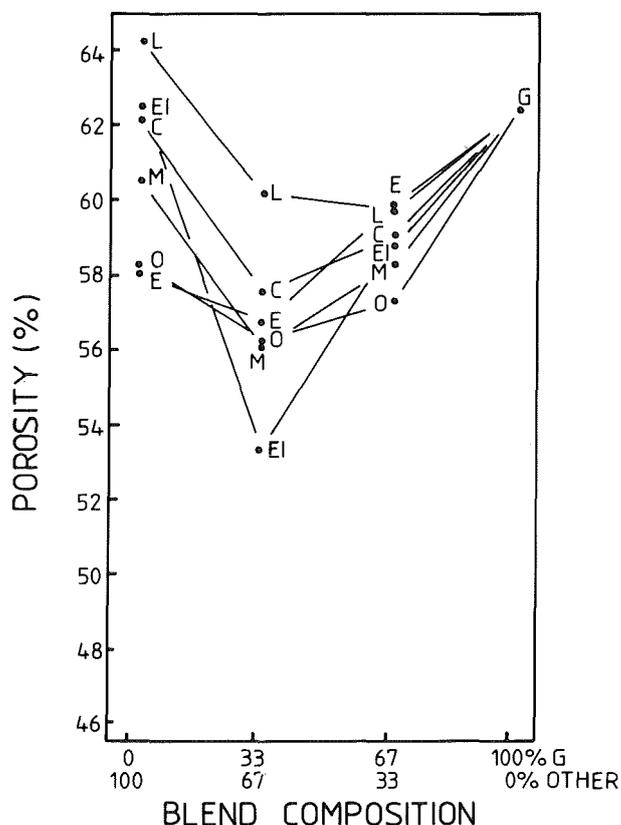


FIGURE 2. Effect on coke porosity by blending coal G with other vitrinite-rich coals (L, El, C, M, O and E)

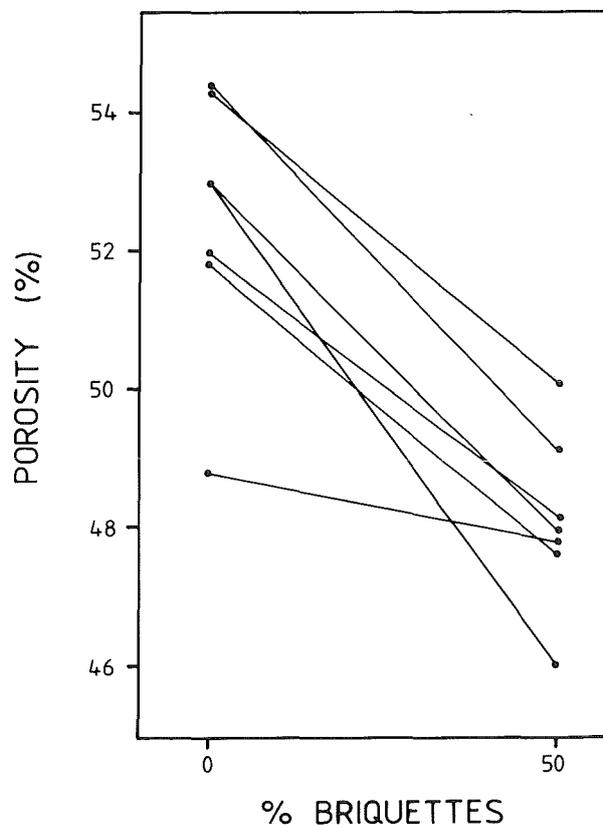


FIGURE 3. Effect of partial briquetting of coal blends on coke porosity

from coal with pitch additions has lower porosities, thicker cell walls and usually larger pore size than the coke prepared from loose coal. The volatiles from the binding material in the briquettes or the pitch thus play an important role in the formation of the structure of the coke. This denser coke contributes toward improved blast furnace behaviour.

Conclusion

The use of an automated image analyser, which is a highly sophisticated computer-based instrument, can give good statistical results on the determination of the structural properties of coke. The information thus obtained can effectively be used in the determination of coke quality.

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