Coal for metallurgical coke production: predictions of coke quality and future requirements for cokemaking

M.A. Díez*, R. Alvarez, C. Barriocanal

Instituto Nacional del Carbón (INCAR), CSIC, Apartado 73, 33080 Oviedo, Spain

Abstract

This paper reviews quality requirements of metallurgical coke for the blast furnace, coke structure, and relationships between structure and quality. Models of prediction of metallurgical coke quality parameters based on maceral composition and properties of coals being carbonized are summarized. Early prediction models of cold coke strength and the development of second-generation hot-strength prediction models based on parameters as coke reactivity index (CRI) and coke strength after reaction with carbon dioxide (CSR) are assessed. The review concludes with an assessment of current coke production and coal demand in the steelmaking industry, globally, followed by a preview of possible future alternative coking technologies. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Coking coal; Cokemaking; Coke quality; Prediction; Strength; Reactivity; New cokemaking technology

1. Coke for the blast furnace

Metallurgical coke is a macroporous carbon material of high strength produced by carbonization of coals of specific rank or of coal blends at temperatures up to 1400 K. About 90% of the coke produced from blends of coking coals is used to maintain the process of iron production in the blast furnace where it has three major roles:

1. as a fuel, it provides heat for the endothermic requirements of chemical reactions and the melting of slag and metal;

2. as a chemical reducing agent, it produces gases for the reduction of iron oxides; and

3. as a permeable support, it acts as the only solid material in the furnace that supports the iron-bearing burden and provides a permeable matrix necessary for slag and metal to pass down into the hearth and for hot gases to pass upwards into the stack.

Of these three roles, the first two can be substituted by oil, gas, plastics, and coal. These are injected at the tuyeres as generating energy and a carbon source. Such a substitution brings about a reduction in coke rates for the blast furnace (coke rate is the weight of coke required to produce 1 t of iron). However, there is no other satisfactory material available, which can replace, fully or partially, metallurgical coke as a permeable support of blast furnace charge.

---

* Corresponding author. Tel.: +34-985-2808-00; fax: +34-98-529-7662.

E-mail address: madiez@incar.csic.es (M.A. Díez).

Over the last few decades, due to ironmaking trends towards increased size and throughput of blast furnace performance together with a simultaneous reduction in coke rate by high levels of injection of carbon via the tuyeres, in particular pulverized coal injection (PCI), the role of coke as a permeable support became greater in importance and so further improvements in coke quality are required. In such conditions, a decrease in coke rate produces a decrease in the thickness of the coke layer in the stack and cohesive zone together with an increased residence time of coke in the lower part of the blast furnace. Coke residence times increase by more than 30% with variations in coal rate from 100 to 200 kg t^{-1} hot metal (Negro et al., 1996). This means that coke is subjected to longer periods under mechanical, thermal, and chemical stresses that further increase coke degradation in the blast furnace.

As a result, future research is focused to improve these critical quality parameters of coke to reach higher productivities when higher levels of fuel injectants are operative.

A realistic assessment of the likely performance of coke in the blast furnace operating with or without injection technology should include those properties of coke that reflect its resistance to degradation under the chemical and thermal environment of the blast furnace. Such properties, providing guidelines for coke use, relate to lump size, shape and size uniformity, chemical composition, mechanical strength, and thermal and chemical stabilities. Thus, coke for the blast furnace needs to be a successful compromise between structure and properties. To ensure good blast furnace performance, coke should be moderately large, with a narrow size range, and have a high mechanical strength in order to withstand the weakening reactions with carbon dioxide and alkali, abrasion, and thermal shock in the blast furnace. Because of the many unknown factors, it is not achievable to establish universal quality indices common to all blast furnaces, although typical specifications for metallurgical coke quality are available.

Impurities present in coke affect its performance in the blast furnace by decreasing its role as a fuel in terms of amounts of carbon available for direct and indirect reduction roles and also its role as a permeable support. Such impurities are moisture, volatile matter, ash, sulphur, phosphorous, and alkali contents. Their levels are kept as low as possible.

Moisture content is a direct consequence of the coke-quenching process with some dependence on size. High and variable moisture contents affect both the coke rate and the balances within the blast furnace, while high volatile matter contents cause operational problems in the cleaning of blast furnace gas. Coke moisture content ranges from 1 to 6 wt.% maximum and common values are in the range 3–4 wt.%.

Of other chemical properties, sulphur and ash (content and chemistry) are of particular importance because as they increase, coke productivity in the blast furnace decreases. The coke ash is a non-productive part of coke which influences slag volume and composition. Industrial experience indicates that a 1 wt.% increase of ash in the coke reduces metal production by 2 or 3 wt.%. Values higher than 10 wt.% can be satisfactory but only if the ash chemistry is acceptable.

Recent papers summarize coke quality requirements for some operating blast furnaces in Europe (Leonard et al., 1996; Großpietsch et al., 2000) with typical coke chemical properties being given in Table 1.

The importance of coke physical properties is linked to the need to support the ferrous burden and to give a permeable matrix through which reducing gases can flow and molten material can percolate in the lower blast furnace region. These physical properties are related to its size (mean and distribution) and its resistance to breakage and abrasion. Coke size is mostly controlled by screening. A large mean size with a narrow size distribution maintains adequate permeability. Most operators consider a mean optimum size to be in the range of 50–55 mm.

Empirical mechanical strength tests, commonly used to measure resistance to size degradation, involve

Table 1
Required chemical properties of blast furnace coke (Leonard et al., 1996)

Chemical property	European range
Moisture (wt.%)	1–6
Volatile matter (wt.% db)	< 1.0
Ash (wt.% db)	8–12
Sulphur (wt.% db)	0.5–0.9
Phosphorous (wt.% db)	0.02–0.06
Alkalies (wt.% db)	< 0.3

db = dry-based.

Table 2
Required physical properties of blast furnace coke in current operation

	European range[a]	Australian BHP Port Kembla[b]	American range[c]	Japan range[d]
Mean size (mm)	47–70	50	50	45–60
M_{40} (+60 mm)	>78–>88	85	n.a.	n.a.
M_{10} (+60 mm)	<5–<8	6.5	n.a.	n.a.
I_{40}	53–55	n.a.	n.a.	n.a.
I_{20}	>77.5	n.a.	n.a.	n.a.
DI150/15	n.a.	84.4	n.a.	83–85
ASTM stability	n.a.	63.6	60	n.a.
CSR	>60	74.1	61	50–65
CRI	20–30	17.7	23	n.a.

Data taken from: [a]Leonard et al., 1996 (also in Großpietsch et al., 2000);[b]Horrocks et al., 2000; [c]Poveromo, 1996; O'Donnell and Poveromo, 2000 (data presented from AISI coke quality survey) and [d]Nishioka, 2000.
n.a.: not available.

dynamic loading either in the form of shatter tests (ASTM D3038), where breakage occurs by impact; or revolving drum tests such as ASTM Tumbler (ASTM D3402), MICUM, half- and extended-MICUM, IRSID and JIS (JIS K2151) tests, where attrition takes place by a combination of breakage and abrasion. In Europe, the MICUM and IRSID (ISO 556 and ISO 1881) tests, which use the same equipment, are dominant. Details of these tests have been reported elsewhere (Patrick and Wilkinson, 1978; Ragan and Marsh, 1980; Loison et al., 1989; Alvarez and Díez, 2000).

Highly stabilized blast furnace coke needs a high abrasion resistance. The range of optimum values, however, is wide and strongly dependent upon the characteristics and operational conditions of the blast furnaces. As an example, typical coke size and strength values for European blast furnaces are given in Table 2 together with those reported for current operation in Australian BHP Port Kembla, American and Japanese blast furnaces.

Although coke mechanical strength indices provide blast furnace operators with a useful assessment of coke performance, they form only part of the overall picture. The tests used to assess coke mechanical strength are made at ambient temperature and hence fail to take into account the process conditions of the coke in the blast furnace.

The importance of high temperature properties of coke was established by the Japanese Steel Industry

from the dissection surveys of three blast furnaces that were quenched whilst operating (Nakamura et al., 1977).

As a result, a combined test for measuring the coke reactivity and post-reaction strength was introduced by the Nippon Steel Corporation (NSC) in the 1970s. The NSC method is described in a research report published by The British Carbonization Research Association (BCRA) (1980). Recently, it has been adopted as an ASTM standard procedure (ASTM D 5341-93a) and is currently being prepared by the International Organization for Standardization (ISO). The method, a major coke quality test which has achieved international routine use by industry, measures the solution loss reaction of coke by carbon dioxide at 1100 °C for 2 h (Coke Reactivity Index—CRI) under standardized gas flow rate conditions together with the coke mechanical strength after reaction with carbon dioxide (CSR index). For a good quality coke, the CRI should be low and the CSR index high. If the coke reacts excessively with the oxidizing gases of the blast furnace which contain increasing proportions of carbon dioxide, coke will weaken and will be degraded into smaller particles. Excessive coke degradation leads to permeability reduction, impaired efficiency of blast furnaces performance, and blockage of the tuyeres with coke residues. A high degree of correlation between the two indices, CRI and CSR, derived from the NSC method was found by various authors. Fig. 1 shows, as an example, the relationship between
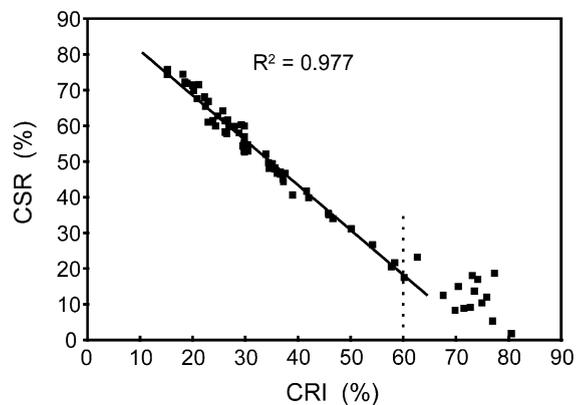

Fig. 1. Relationship between CRI and CSR indices (after Menéndez et al., 1999).

CRI and CSR index for a series of more than 60 cokes produced from single coals of different rank and geographical origin and from complex coal blends (Menéndez et al., 1999). The cokes were produced at the INCAR Experimental Coking Test Plant (Díez et al., 1991) using a coke oven of 6-t capacity in operation until July 1999. A correlation exists between CRI and CSR indices derived from the NSC test, but some points show scatter, being greater for high reactive cokes. Based on such a relationship, one parameter should be sufficient to classify a coke and, then, the CSR index is commonly used in the control of quality of blast furnace coke.

Other tests developed to evaluate the coke reactivity are summarized in a recent publication (Menéndez et al., 1999). These authors show a correlation between the ECE-INCAR reactivity and the CRI and CSR indices for 33 different cokes ($r^2 = 0.9463$ and 0.9339, respectively). The ECE-INCAR method measures metallurgical coke reactivity and is based on a test standardized by the United Nations Economic Commission for Europe (UNECE) (1965). In this test, a smaller amount of coke (7 g) with smaller particle size (1–3 mm) in comparison to the NSC test, which uses 200 g of coke of 19–21 mm size, is used.

Several authors have demonstrated the relevance of CSR in blast furnace operation to maintain permeability at constant and optimum values. For instance, it was found that when the CSR index decreases, the gas permeability resistance or impermeability index increases (Nakamura et al., 1977; Hatano et al., 1990; Colleta et al., 1990). From these relationships, different minimum values of CSR indices have been established to ensure stable operation, i.e., CSR values >48% and >55% for coke feed at NSC Oita and Kimitsu blast furnaces, respectively. A CSR index higher than 58%, needed to achieve an impermeability index lower than 6, was established at the Taranto works in Italy (Colleta et al., 1990). Moreover, due to a better burden permeability, an increase in CSR of 1% in the specific CSR range 45–55% accounted for 1.5 kg t^{-1} reduction in fuel rate in the NSC Hirohata works (Hara et al., 1980). For a Port Kembla blast furnace, an increase of 30 t day^{-1} in iron output and a fall of 0.4 kg t^{-1} in fuel rate for CSR, in the ranges 52–62% and 58–73%, respectively, is also reported by Rooney et al. (1987).

More so, the largest blast furnaces, operating with low coke rates (high PCI levels) and high productivity, also showed the importance of high CSR indices. Experience shows that with increasing CSR index of coke feed to the larger blast furnaces, the injected coal level can be increased and the low coke rates maintained. However, CSR indices below 60%, in most cases, are not acceptable because of resultant higher-pressure losses and reduced furnace permeability (Großpietsch et al., 2000). CRI indices should be in the range of 20–30%.

Despite the beneficial effects of a high CSR of coke on blast furnace operation and fuel consumption, some doubt has been raised about the validity of the experimental conditions related to the CRI and CSR measurements (Vogt, 2000). Some blast furnaces, for instance, operate stably with low CSR cokes. Coke in the blast furnace is subject to varying temperatures and gas compositions. Gasification occurs in the presence of many other reactions, some of them are interactive principally the reduction of the iron ore. However, no other test today has reached an industrial application. Assessment of coke quality using CRI and CSR indices, as defined by NSC, dominates in practice.

As with other coke quality parameters, there is no single optimum value of CSR and CRI. It may vary between blast furnaces having different furnace characterics and operational conditions but having the same coke rate. Coke qualities used by different European Companies are reported in Joint European Blast Furnace Committee papers presented at the two last European Cokemaking–Ironmaking Conferences (Leonard et al., 1996; Großpietsch et al., 2000). Typical coke quality values are in Table 2 together with those reported by non-European producers (Horrocks et al., 2000; Poveromo, 1996; O'Donnell and Poveromo, 2000; Nishioka, 2000).

An interesting aspect of the joint European Coke Committee paper presented at the 3rd International Cokemaking Congress is the estimation of the maximum attainable coke quality based on actual coking plant data (Vander et al., 1996). For a coke with a stabilization of a 25-m drop, the CRI and CSR estimated values are 24% and 70%, respectively, for a conventional coke oven of 450-mm width operating with a coking time of 18 h. These coke quality parameters can be achieved for coal blends develop-

ing a maximum gas pressure of about 7 kPa. Major limitations to obtain higher coke quality are considered to be coking pressure and the conventional coking process itself. Analysis of operating results by NSC in the 1970s suggested that CSR depends about 70% on the coal or coal blend and about 30% on coking conditions (Nakamura et al., 1977). The importance of the coal blend as a factor in coke gasification has been noted by other researchers (Bernard et al., 1985, 1986). Although coking conditions, for example, bulk density, coking time, preheating of the charge, and the incorporation of non-coal materials, do minimize some deficiencies in coal properties, the selection of an optimum coal or coal blend is a priority for best coke.

2. Coke structure and its effects on coke properties

Over the past 30 years, an extensive literature describing relationships between coke structure and properties has been generated. This paper does not review these papers but reports general guidelines.

There is a close relationship between the CSR index and the vitrinite reflectance of coal. The CSR index passes through a maximum in the region of prime coking coals with mean reflectance of 1.2% to 1.3% and falls towards coals of lower or higher rank (Nakamura et al., 1977). Fig. 2 shows the relationship of CSR index with coal rank. As coke reactivity to CO_2 is inversely related to CSR, there are minimum
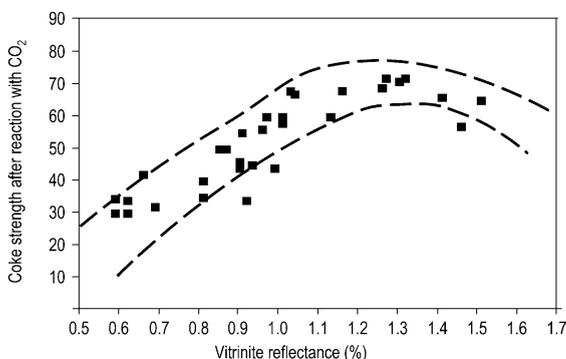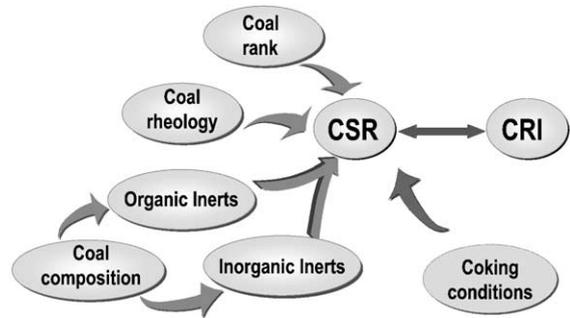

Fig. 3. Main factors influencing the coke strength after reaction with carbon dioxide at 1100 °C (CSR index).

values of reactivity to CO_2 in the region of prime coking coals, increasing for cokes from lower and higher rank coals (Hyslop, 1981; Patrick and Wilkinson, 1981). Such a behavior may be explained by the relative proportion of the different structural components within coke with strong dependence of coal rank. However, some cokes fall outside this close relationship (Hyslop, 1981), indicating that rank, although being a dominant factor, is not the only factor that influences CRI and CSR. Catalytic effects of the coke gasification and coking conditions also need to be considered (Fig. 3).

It is established that the rank and chemistry of the parent coal strongly influence the optical textures of cokes. Several studies show that the development of anisotropy (size, shape and intensity) during carbonization varies mainly with (a) coal rank; (b) petrographic composition of the coals; (c) plasticity of the parent coal; (d) carbonization conditions such as rate of heating, soak time and gas overpressure; as well as (e) the nature of additives used in the coal blends (Marsh, 1982, 1992; Marsh and Clarke, 1986).

In recent years, fundamental research has demonstrated that metallurgical coke should be considered as a composite material whose strength and reactivity depend on contents of isotropic carbon and inerts, the size and shape of the anisotropic carbon units, and the interfaces between textural components together with porosity and ash chemistry (Marsh and Clarke, 1986).

Coke strength involves fissuring and breakage along the boundaries of, or sometimes through, both macroscopic and microscopic coke components, with porosity being a dominant factor (Patrick and Walker, 1995). For cokes of comparable porosities, differences

Fig. 2. Relationship between vitrinite reflectance and coke strength after reaction with carbon dioxide (CSR index) (Nakamura et al., 1977).

in strength and related properties are dependent upon the constituents of pore-wall material. Marsh and Clarke (1986) summarized the following considerations about the strength of optical textural components and the strength of binding at the interface: (a) mosaics are interlocked with each other at the molecular level to form a firmly bonded interface; (b) fissure propagation across the randomly orientated mosaics is not facilitated by this type of structure; (c) inert materials in coke coming from both the parent coal or added breeze coke may either be encapsulated if they have smooth surfaces or may be interlocked over rough or porous surfaces; and (d) coke microstrength increases with coal rank to a maximum as the structure of the coke changes from isotropic, to mosaic, and to flow-type anisotropy.

To this end, a methodology has been developed to quantify the degree of bonding in the transition zones between the textural components of coke and the binding between textural components and inert material interfaces (Barriocanal et al., 1994). Coke properties and coking behavior of single coals and blends using petroleum coke (Menéndez et al., 1996, 1997) and waste materials (Barriocanal et al., 1998) have been related to extent of bonding at interfaces between optical textural components and inert material. Concerning coke reactivity, significant progress has been made in the understanding of those factors controlling internal bonding of cokes such as the coal rank, amounts of inerts and ash in coal, the metal oxide content of coke ash, and the optical textural components of the coke. Optical microscopic observations on non-gasified and gasified cokes established a relative reactivity scale for the different components in coke, inert>isotropic>fine mosaic>coarse mosaic> flow (Fujita et al., 1983).

3. Coke quality prediction

Currently, normal practice in cokemaking demands a coal blend that is low in cost, produces a high quality coke, and provides a safe oven pushing performance. Coal blending has been adopted by the industry partially because of the limited availability and high cost of prime coking coals and also because of the continued demand for better quality coke for the blast furnace. Coal blending varies in the number of coals used (four or more), the proportion, rank, coking properties, and geographical origin of coal components.

Coal selection and blend composition are major factors controlling coke properties (physical and chemical). As aids to coal selection and coke quality prediction, several mathematical models are available, divided into two groups according to the coke properties involved. The first-generation group of models focuses on the prediction of cold mechanical coke strength (i.e., ASTM stability and MICUM indices). The second-generation of models uses the CRI and CSR indices as coke quality parameters. As far as can be ascertained, no prediction model has reached universal application. Some coals or blends show significant deviations between values predicted by a model and values obtained experimentally. However, almost all coking plants have, for internal use, some form of a model based on coal rank, rheological properties, petrology, and ash chemistry.

3.1. Prediction models of coke strength from coal properties

Strength is the most important physical property of coke with considerable effort attempting correlations with coal rank and type in terms of total inert content, rheology as indicated by maximum fluidity, total dilatation, and parameters deduced from petrographic compositions of coal. Based on these considerations, some mathematical models use petrographic compositions in the evaluation of the coal coking potential and the prediction of coke strength. Thus, petrographic analyses are assuming a major role in coal blend assessments.

Petrographic analyses for the prediction of coking properties have classified macerals into reactives and inerts. The pioneering works of Stopes (1919) provided the basis for a definition of coal composition based on the optical properties and the ''maceral concept'' (Stopes, 1935). This represented a major advance into understanding of how an optimum ratio of reactive and inert components affects coal carbonization properties and behavior. The term ''reactives'' includes those macerals which soften on heating and bind ''inerts'' (those macerals that remain unaltered on heating) and then resolidify into a porous, fused, solid carbon material. Consequently, coke structure should

be considered as a composite material where most of the coke constituents came from the reactive materials (binder) and a much lower proportion from the material which acts as a filler (inerts) during the carbonization process. To obtain a good coke, a known proportion of reactive and inert material is required and the optimum amount of each one will vary with the type of reactive macerals. In general, reactive macerals include vitrinite, liptinite, and one-third of semifusinite, while inert coal constituents during carbonization are two-thirds of semifusinite, fusinite, macrinite, micrinite, inertodetrinite, sclerotinite, and mineral matter (Ammosov et al., 1957; Schapiro and Gray, 1960). However, such a classification of coal reactive and inert constituents is not equally applicable to all coals, consequently some laboratories do not follow it in detail. In particular, the division of the amount of semifusinite acting as reactive or inert is controversial. An explanation for this discrepancy is that composition and, hence, technological behavior of some macerals may differ from region to region because of different coalification and depositional conditions causing, e.g., for one coal to contain macerals that are mainly inert but which are reactive in another coal. Thus, Benedict et al. (1968a) considered a similar breakdown for semifusinite plus other inertinite macerals of low reflectance, while other authors used little semifusinite in their reactives (Brown et al., 1964); or relatively large and variable amounts (Steyn and Smith, 1977).

More details on the development on coal petrology (classifications and behavior of maceral groups and microlithotypes and its application to cokemaking) have been published in a number of excellent reviews (Stach et al., 1982; Bustin et al., 1985; Falcon and Snyman, 1986; Kaegi et al., 1988). As an introduction on coal petrology, a recent book chapter also summarizes the work carried out on this area (Suárez-Ruiz, 2000).

Based on the maceral behavior during carbonization (reactive and inert), a concept that is used today for explaining coal carbonization behavior, several mathematical models have been developed to predict coke strength. Each model seems to estimate with a high degree of accuracy when they are applied to coals which are more or less similar in petrographic composition to the coals for which the model was developed, and coke potential is being evaluated on coals which might be tested under the same experimental conditions used in the model.

Two models were developed in USA based on petrographic data for the prediction of coke strength and the formulation of coking blends, one developed by United States Steel and the other by Bethlehem Steel. A detailed history of the development of the two models has been recently reported (Dutcher and Crelling, 2000; Spackman, 2000; Thompson, 2000).

The US Steel model for ASTM coke stability was firstly designed by Schapiro et al. (1961) and based on the earlier Russian work of Ammosov et al. (1957). The model of Ammosov et al. (1957) relates to coke prepared in commercial coke oven batteries and predicts coke strength from the petrographic data of the parent coals. Schapiro et al. (1961) modified this model and applied it to coals used by US Steel for the prediction of the ASTM stability factor. Schapiro and Gray (1964), and later Harrison et al. (1964) and Moses (1976), further developed the models. The US Steel model is the basis for predicting coke quality from specific coals. The reactive coal components include all vitrinite and liptinite and one-third of the semifusinite, while the inerts include the remaining two-thirds of the semifusinite, together with the remaining inertinite and mineral matter. This model requires data from the maceral and reflectance analyses in which the reflectance of all the reactive and semi-inert macerals are measured, as well as the calculation of mineral matter of coal (MM) by the Parr formula (MM $= 1.08$ Ash $+ 0.55$ $S_{pyritic}$ (Loison et al., 1989, p. 29)). Reactive macerals are further subdivided into 21 ranges of 0.1% reflectance (vitrinoid type, V-type). From petrographic data and coke strength values, different curves can be plotted: (a) the optimum ratio of reactives to inerts (R/I) for each vitrinoid type; (b) the variation of the strength index (SI) with the quantity of inerts for various vitrinoid types; and (c) the variation of the strength index with the composition balance index (CBI) for various stability factors. For each vitrinoid type (V-type), there is an optimum ratio of reactive to inert components that influences maximum coke strength (optimum inert ratio). This optimum inert ratio was established by physically isolating each of the macerals and determining, by means of micro-coking tests, what ratio gave the maximum strength value. With higher or lower proportions of inerts than the opti-

mum, the coke strength decreases. The other indices defined by these authors are: a composition balance index (CBI) and a strength index (SI), the latter also being referred to as a rank index. CBI is the ratio of inert components in coal to the optimum ratio of reactive to inert that a coal of a given rank should have. When for a given coal the optimum CBI is equal to 1, the best coke is obtained. The strength index (SI) can be evaluated to determine the relative coke strength made from coals of different ranks and types. Thus, it is possible to predict ASTM stability factors from CBI (as a caking property parameter) and SI (as a rank parameter).

An advantage claimed for this model is that both indices, CBI and SI, are additive and it should be possible to use petrographic assessments without pilot coke oven tests. The US Steel model gave fully satisfactory results for the low semifusinite Appalachian coals and blends for which the system was developed. However, this model is based on correlations of test-coke data with the petrographic composition of the coals being carbonized, keeping the test conditions constant. The restrictions of this model are limited to a <2 wt.% of moisture content of coal, ash content no higher than 12 wt.%, tested coals crushed to 80 wt.% minus 1/8 in. in size, the bulk charge density of approximately 55 lbs/ft^3, and a coking rate of 1 in. per h (note: 1 in.=2.54 cm).

Various coke prediction models, in general, follow the same approach of Schapiro et al. (1961). The Bethlehem Steel model, first reported by Benedict et al. (1968a) and later by Thompson and Benedict (1976), was developed to accommodate the Eastern Kentucky coals used in coal blends of this company. Similarities are related to the use of a one-third–two-thirds breakdown of semifusinite and other semi-inerts, and the use of maximum reflectance as the parameter of rank determination. Differences with Schapiro et al. (1961) are mainly concerned with the carbonization behavior of that part of the vitrinite maceral group that remains unaltered in the coke. They called this material pseudovitrinite (Benedict et al., 1968b). The inertness of pseudovitrinite is determined by: (a) separate measurements of the maximum reflectance on pseudovitrinite (R_{maxPsv}) and normal vitrinite (R_{max}); (b) the reflectance spread ($R_{maxPsv} - R_{max}$); and (c) estimation of the pseudovitrinite amount that should be included in the effective inert content. The

remaining pseudovitrinite is considered to be a reactive component. The ASTM stability factor can be estimated from the effective inert content and the normal vitrinite reflectance.

This petrographic model has been adapted to select coals not having excessive coking pressures (Benedict and Thompson,1976). Some low-volatile coals, especially those with vitrinite reflectance >1.65% and of low inert content, produce excessively dangerous pressures during carbonization (Tucker and Everitt, 1992). In normal industrial practice, these low-volatile coals are never coked singly and are normally used as a 25–30 wt.% component in coal blends. The authors showed that both, vitrinite reflectance and maceral composition of coal, could be related to excessive pressures during carbonization. Coking pressure varied directly with reflectance and inversely with effective inert content of low-volatile coals. The model allows a classification of coals as safe, marginal and non-safe coals.

The coke strength prediction model of Brown et al. (1964) differs from those already described in the subdivision of semifusinite. They considered that "virtually all semifusinite remains unchanged during carbonization and the same applies to micrinite". The predominant inert character of semifusinite was first noted by Taylor (1957) who concluded, "semifusinite may be distorted and altered in its chemical nature, but that no appreciable amount fuses and enters the fused coke structure". Taylor et al. (1967), in an inter-laboratory study, provided evidence of the inert role of semifusinite using a range from low volatile to high volatile bituminous coals. They also suggested that transitional material between vitrinite and semifusinite became weakly plastic during heating and it should be included in the category of vitrinite. Consequently, they suggested that, as a rule, semifusinite with a reflectance higher than 0.1 to 0.2 compared with the mean reflectance of vitrinite in the same coal is inert. Other authors argued that the inert character of the semifusinite could be a result of the small laboratory-scale test oven, used in the carbonization tests. In coking conditions, the semifusinite could be much more interactive.

Brown et al. (1964) considered two other factors not already applied in the models previously described. One is related to the dimensions of the inertinite material in the coal. If the inertinites are

relatively large, they tend to form centers of weakness in the resulting coke and therefore have a deleterious effect on strength. The second factor concerns the relative association of reactive and inert materials. For these reasons, a prediction of coke quality (shatter index and ASTM hardness factor) using Australian coals was undertaken in terms of the microlithotypes, vitrite and clarite. The method can provide useful information on coke strength to assess the potential of a given coking coal.

On the other hand, when Cretaceous coals from Western Canada, with a higher proportion of semifusinite than Carboniferous coals were used, the model of Schapiro et al. (1961) predicted coke strength values lower than those measured experimentally. To get satisfactory coke strength prediction using such coals with a total semifusinite content greater than 20%, CANMET introduced a modification based on the assumption that semifusinite makes a greater contribution to the reactive components (Carr and Jorgensen, 1975). They decided that half of the semifusinite is effectively a reactive component. However, no evidence was found to support this assumption. The work of Nandi and Montgomery (1975) on the nature and thermal behavior of semifusinite in such coals using a hot-stage microscope provided useful data. They concluded that high-reflectance semifusinite is a totally inert component during carbonization (no signs of melting or softening were observed at 450 °C), while low-reflectance semifusinite was reactive, similar to vitrinite, and its melting point being about 85 °C lower than that of the corresponding vitrinite. The pure vitrinite was melted at a temperature of 420 °C.

Based on these conclusions, other models developed for CANMET (Pearson and Price, 1985) introduced the "cut-off" reflectance or reactive "cut-off" value ($R_{cut-off}$) that separates reactive and inert macerals in a random reflectogram of all coal macerals. A good correlation between maximum vitrinite reflectance and $R_{cut-off}$ for 76 coals ranging from 0.89% to 1.63% (R_{max}) was found ($r^2 = 0.92$). For a given coal, the new parameter can be deduced from the correlation between R_{max} and $R_{cut-off}$.

Recently, the proportion of semifusinite as a reactive component, in the range of 33–50%, has been determined by coke microscopy. The proportion varies from coal to coal and 50% of the reactive

semifusinite is recommended for Western Canadian coals when coal petrography is used to predict coke strength for blends. Another problem is to explain the low rheological values for Western Canadian coals. The rheological values cannot be compared to other similar rank coals (Australian or US Carboniferous coking coals) and, therefore, they should not be directly used in calculations of average fluidities of blends containing coals from many sources (Leeder et al., 1997). High quality coke can be produced from Canadian coals with maximum fluidities in the range of 3–10 ddpm. These values strongly differ from the minimum value of maximum fluidity (100–200 ddpm, see below) established in the MOF diagram (relation between Gieseler maximum fluidity and maximum reflectance of vitrinite for coal blending used in Japan). On the other hand, comparing dilatation values of Canadian and US coals of the same rank and inert content, Canadian coals have a total Ruhr dilatation of approximately three to four times lower. The authors suggested two approaches to assist with coal blend preparation. One is to use an "equivalent" maximum fluidity for blends containing low proportions of Canadian coals. However, it is concluded that the most satisfactory evaluation of the influence of such coals in blends made with coals from other sources is the use of pilot-scale carbonization tests.

Another prediction model is that developed by Steyn and Smith (1977) from ISCOR in South Africa. In this model, variable proportions of semifusinite as reactives are considered. Many South African coals have relatively low vitrinite contents and large amounts of semifusinite and of micrinite. With such coals, the coking coal blend is characterized by a similar amount of vitrinite and semifusinite plus micrinite. For instance, vitrinite contents are as low as 40% and semifusinite and micrinite contents are as high as 45%. Both semifusinite and micrinite should have a significant role during carbonization and for technological purposes need to be classified according to reflectance and structure into reactive and inert types. The authors indicated that "the classification proposed complicates maceral analysis but elucidates the coking behavior of low-vitrinite coals".

Other models to predict coke strength do not exclusively use coal petrography and utilize the dilatation characteristic of coals and blends using a Ruhr or Audibert-Arnu dilatometer. The model devised by

Mackowsky and Simonis (1969) in Germany provides not only a basis for prediction of the MICUM strength and abrasion indices (M_{40} and M_{10}) for cokes produced from a range of Ruhr coals and blends, but also yields of carbonization products (coke, gas, liquid by-products, and tar). In addition to coal composition parameters, the model takes into account coking conditions, that is, coal size of the charge, bulk density, coking rate, mean oven width, and coking time.

In this model, the MICUM M_{40} index (wt.% of coke, >40 mm after 100 revolutions) is expressed as:

$$M_{40} = aK + b + d\text{Ms}$$

Where K is a factor including the coking conditions as a function of the bulk density, oven width and coking time; Ms is the particle size content of the coal; b is a rank factor; and a and d are coefficients based on volatile matter and a parameter named G-factor.

The G-factor is usually obtained from parameters derived from the Ruhr dilatometer test, which is a modification of the Audibert-Arnu dilatometer test (the softening and resolidification temperatures and the percentage of coal contraction and dilatation). Although the G-factor is considered additive for coal blends, there is a limitation for blends composed of coals whose plastic range does not overlap sufficiently (Gibson, 1972). Experiments on the behavior of the different maceral groups in this dilatometer showed that it was also possible to calculate the G-factor for a given coal from its petrographic composition.

The prediction model for M_{40} and M_{10} indices (wt.% of coke >40 mm and <10 mm in size, respectively, after 100 revolutions) can only be applied to a certain range of coals characterized by volatile matter contents between 18 and 35 wt.% daf and inert contents below 20%, this being very restrictive.

A quite different approach to predict coke strength is based on the relationship between petrographic data (R_{max}) and rheological properties (Gieseler maximum fluidity) of the coals (MOF diagram). The fundamental concept for the blending target used is based on the assumption that coke strength is mainly governed by two main coal properties, i.e., the caking property and the rank of coal. Fig. 4 shows the dominant regions for each parameter.

The model developed by Nippon Kokan (NKK) in Japan is described as a coal property "window
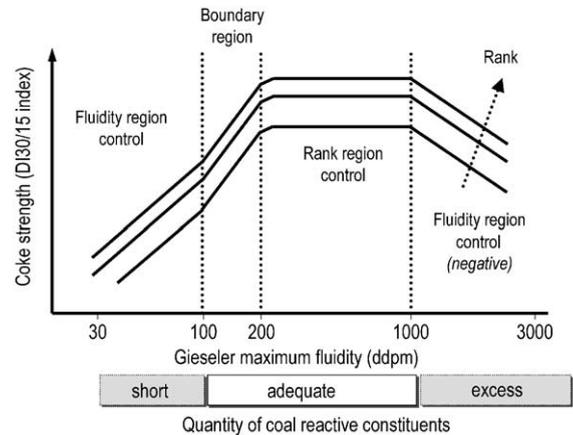

Fig. 4. Diagram showing the dominant regions of the fluidity and the rank of coal to control coke strength (Miura, 1978; Morishita et al., 1986).

model", where the blending ratio is determined such that the maximum vitrinite reflectance and maximum fluidity of the coal blend falls within an established optimum range ("window" or "rectangle"). If the coal blend characteristics fall inside this window, the resulting coke will be of acceptable quality for blast furnace use (Okuyama et al., 1970; Miyazu, 1974; Miura, 1978). Within the framework of this model, desirable properties of a coal blend range of 1.2–1.3% and a Gieseler maximum fluidity between 200 and 1000 ddpm (Fig. 5). The coal blends must meet these specifications if the target coke strength index of higher 92.0 (JIS DI30/15, wt.% of 15 mm coke after 30 revolutions) is to be obtained.

To determine the role of each coal in the blend, the MOF diagram (Fig. 5) is divided into four quadrants. Coals belonging to quadrants I and II provide adequate fluidity to the blend, while coals within quadrant IV are used to adjust the rank of the blended coals. However, coals with both low rank and low fluidities (in quadrant III) are used only as a source of carbon for blending.

The "window" of acceptable blends defined from the MOF diagram has been regarded as being too restrictive by the Centre de Recherches Metallurgiques (CRM) in Belgium. Here, blends with the optimum characteristics described in the MOF model (falling within the "window") gave excellent coke strength.
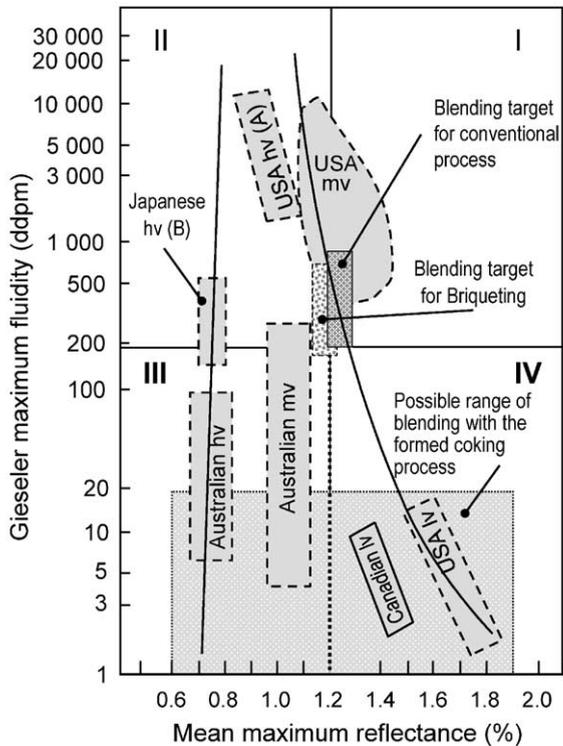
Fig. 5. Relationship between Gieseler maximum fluidity and maximum reflectance of vitrinite for coal blending target (MOF diagram) (Miyazu, 1974).

However, blends located outside of the optimum window also gave high quality coke indices (Poos, 1987). For the coal blends studied, other prediction models do not work satisfactorily. The CRM has developed a coke strength prediction model based on three parameters: (a) the inert content of coal, (b) the reactive caking index, and (c) the maximum fluidity (Munnix, 1984). The reactive caking index was defined as a function of vitrinite reflectance. Formulae have been developed for calculating each of these parameters for blends from the values of the individual coals carbonized in a 300-kg pilot oven.

Modifications to the Japanese MOF diagram were introduced by Spanish Steel, Ensidesa (now, Aceralia), to accommodate coal blends used for coke production (Sirgado and Verduras, 1978). For better correlation of coal rank with coke strength, they defined the mean effective reflectance of vitrinite as the mean random reflectance of vitrinite restricted to the range of V-steps 6 to 18 (both included).

The Coal Research Establishment in UK established a target specification for the blending of high- and low-volatile coals based on total dilatation and volatile matter content (Gibson, 1972; Gibson and Gregory, 1978). It is recognized that these two parameters, generally, are not sufficient in themselves. In addition, it is necessary to consider the size distribution of the mineral matter associated with the coal and the compatibility of blend components.

In summary, because of the fundamental publications by Ammosov et al. (1957) and Schapiro et al. (1961), the prediction models of coke strength have expanded considerably. Noting that coke quality parameters are strongly dependent on coal properties, efforts were focused on the use of petrographic and rheological properties. With regard to the single coal and coal blends used, one prediction model can be no better than any other model. That is, it is satisfactory if the model is confined to a particular coal and coal blend. Its application to coals of different geological histories is less successful.

3.2. Prediction models of CRI and CSR indices

In the first-generation group of models, coking coal properties were mostly established by petrographic and rheological properties in order to predict cold mechanical strength of blast furnace coke. But, many coal deposits having excellent coking ability from a traditional point of view, do not have an acceptable mineral composition. After introducing the parameters CRI/CSR to assess coke quality, the role of the ash composition has been given more consideration because certain minerals in coke have an accelerating or inhibiting effect on coke gasification because of catalytic reactions. From the almost linear correlation between CSR and CRI indices, it is deduced that the lower the reactivity the higher the coke strength (Fig. 1). Then, catalytically accelerated coke reactivity lead to lower CSR values even if the coking coal blend has excellent rheological properties. Fig. 3 summarizes the factors affecting the CSR and used in prediction models. Taking into account the above considerations, from the adoption of the NSC model for testing coke quality, different approaches have been developed for predicting the CRI and CSR indices and continue to be a research subject of considerable interest. They can be considered to be second-generation prediction models.

On this topic, excellent reviews exist in the literature (Goscinski et al., 1985; Goscinski and Patalsky, 1990; Valia, 1989; Gransden et al., 1991; Edwards, 1991).

The first attempt to predict CSR from coal properties was made from the mean vitrinite reflectance and the inert content (Miura, 1978). Following that, different prediction models from coal properties have been reported by several authors.

The model developed by NSC is based on the vitrinite reflectance and the inert content of coal, including inertinite plus two-thirds of semi-inertinite. Similar to the approach of Schapiro et al. (1961) for predicting ASTM stability factor, an optimum inert content for each vitrinite reflectance is necessary to obtain the best CSR value (Hara et al., 1980). They suggest that as CSR is controlled at least in part by the previous CO_2 gasification, an important role may be also expected of the ash content and composition.

Further studies on coals, prepared by 'specific gravity' separations, with the same mean vitrinite reflectance, but containing different amount of inertinite and ash conclude that inertinite, especially in coals with low reflectance, increases reactivity towards carbon dioxide even when the coke has a low ash content (Sakawa et al., 1982). A multi-parametric equation to predict reactivity towards carbon dioxide as a function of the mean reflectance of vitrinite, the inertinite content, and an alkali index, including quadratic and interaction terms, has also been reported. In this model, the rate of CO_2 gasification of coke is estimated as follows:

$$K = aR^2 + bI^2 + cB^2 + dRI + eBI + fBR + gR + hI + iB + j$$

where R, the mean reflectance of vitrinite; I, the inertinite content; B, the alkali index; and, a–j, numerical constants calculated from the least-squares method.

The alkali index (B_{ash}) defined in this model can be expressed as:

$$B_{ash} = \text{ash (wt.\%)}[(Fe_2O_3 + CaO + MgO + Na_2O + K_2O)/(SiO_2 + Al_2O_3)]$$

The NSC model, using a multi-parameter equation, has been used by Taranto works in Italy. For over 50 individual coals, a correlation coefficient of 0.91 was obtained for the relationship between the estimated values and experimental CSR values. The latter were determined from cokes produced from single coals placed in a box located in an industrial coke oven. A standard deviation of residuals of 1.7% was obtained (Colleta et al., 1990).

The model developed by Kobe Steel predicts coke strength after reaction (CSR) by means of the Reaction Strength Index (RSI), which is determined differently than the NSC model. The model, as reported in the review papers (Goscinski et al., 1985; Goscinski and Patalsky, 1990; Valia 1989; Edwards, 1991), is based on mean vitrinite reflectance (R_0), Gieseler maximum fluidity (MF) expressed as logarithm (base 10), and the ratio of principal basic ($Fe_2O_3 + CaO + K_2O + Na_2O$) and acidic ($SiO_2 + Al_2O_3$) components in the coal ash, as follows:

$$\begin{aligned} RSI = {}& 70.9 + R_0 + 7.8(\log MF) \\ & + 89[(Fe_2O_3 + CaO + Na_2O + K_2O) \\ & /SiO_2 + Al_2O_3)] - 32 \end{aligned}$$

where $CSR = RSI - 10$.

A similar model for RSI prediction, later reported by Kobe Steel (Oguri et al., 1987), differs mainly in the basicity or alkali indices which are used. The model takes into account previous findings which indicate how coke reactivity is affected by: (a) textural components in coke, which are mainly controlled by coal rank reflected as the mean vitrinite reflectance; (b) coke pore volume which is mainly controlled by the development of fluidity; and (c) ash components, especially Fe_2O_3 and K_2O. A new basicity index (B_{ash}) was defined as follows (Oguri et al., 1987):

$$B_{ash} = \text{ash} \times (Fe_2O_3 + CaO + Na_2O + K_2O)$$

However, using the new parameter, major differences between the estimated and experimental RSI values have been obtained, in some cases, values differing by 10 points.

The Iscor model is based on the relationship between CSR and a parameter F (Goscinski et al., 1985; Goscinski and Patalsky, 1990):

$$CSR = 2.56F - 15.52$$

where F is defined as a function of the maximum vitrinite reflectance (R_{max}), Gieseler maximum fluid-

ity in ddpm (MF), percentage of the inert content of organic nature, and ash components, as follows:

$$F = [R_{max} \times MF]/[(Na_2O + K_2O)(CaO + MgO + Fe_2O_3) \ (Organic \ inerts)]$$

A modified basicity index (MBI) has been also introduced by CANMET. The MBI is calculated from coal ash chemistry and proximate analysis (Price et al., 1988, 1992; Gransden et al., 1991):

$$MBI = [(100A)/(100 - VM)] \ [(Na_2O + K_2O + CaO + MgO + Fe_2O_3)/(SiO_2 + Al_2O_3)]$$

where A and VM are the ash and volatile matter contents in coal.

The MBI provides an estimation of the amount of the basic components in the coke. A good relationship between CSR and MBI was found, which can be improved by considering petrographic data (mean vitrinite reflectance) and dilatation parameters.

For 33 Cretaceous and 22 Carboniferous coals, a correlation coefficient of 0.94 and a standard deviation of 3.4 were obtained. When applied to the 33 Cretaceous coals and blends, the same correlation coefficient was obtained, and the standard deviation was 3.3. The model cannot be applied, however, to oxidized (weathered) coals and it is restricted to the coking conditions applied.

Recently, this prediction model has been modified by BHP works in two ways: (1) by introducing the coal dilatation temperature range as a new parameter in the equation; and (2) by substituting the MBI by $(MBI)^2$. With these modifications, the CSR can be predicted with a standard deviation of 2.72. However, the authors indicated that caution should be taken when applying the model to some of the coals tested (McKenzie et al., 1998).

Based on the most significant correlation found between CSR and the coal plastic range, (calculated as the difference between the solidification temperature and the softening temperature measured by Gieseler plastometer), compared to those found for other parameters such as maximum fluidity, total dilatation and free-swelling index, a prediction model has been developed at Inland Steel. The CSR prediction model,

first published in a brief paper (Valia, 1987) and later reported in detail (Valia, 1989), introduced the range of plasticity as a parameter reflecting rank and rheological properties of coal. The other term in the multiparameter equation is the catalytic index (CI), which takes account the combined effect of ash chemistry and sulfur. The CI was defined as:

$$CI = 9.64 \ alkali \ index + 14.04 \ sulphur \ content$$

The alkali index used is calculated as a product of ash content and the weight percent ratio of the basic $(Fe_2O_3 + CaO + MgO + Na_2O + K_2O)$ and acidic $(SiO_2 + Al_2O_3)$ components.

The equation used in the prediction model for CSR, equally applied for single coals and blends, is as follows:

$$CSR = 28.91 + 0.63PR - CI$$

where PR is the plastic range in °C; and CI is a catalytic index (defined above).

To justify the introduction of the above terms in the equation, the following considerations apply: (a) the increase in plastic range optimizes the extent and size of anisotropic carbon from mesophase which, in turn, decreases the reactivity to carbon dioxide (Valia, 1989); (b) the increase in alkalis, iron, and sulfur may have a double effect on CSR. On one side, creating functional groups that affect fluidity and inhibit growth of crystallites and on the other acting as a catalyst of gasification by carbon dioxide (Marsh and Walker, 1979).

The application of the Inland Steel prediction model is valid for the range of coals investigated in the study quoted: plastic range from 34 to 117.5 °C, catalytic index from 14.77 to 39.20, mean vitrinite maximum reflectance from 0.65% to 1.65%, total inerts from 3% to 30%, maximum fluidity from 2 to >30 000 ddpm, and the controlled conditions in the pilot oven (Valia, 1989).

In an attempt to apply the model to coals with high organic inert contents such as the Western Canadian and some Australian coals, where it seems that these organic inerts are not truly inerts, knowledge of inertinite behavior must be considered.

The CSR prediction models described are aided by the fact that CSR is additive. However, opposite views on this matter have been reported. Some authors

found that estimation of CSR by application of the additivity rule is not that accurate (Chiu, 1982; Bernard et al., 1986; Barsotti and Damiani, 1988; Ishikawa et al., 1990), while others claimed that CSR is an additive property (Caldeira and Da Silva, 1988; Morishita et al., 1986; Valia, 1989).

The above prediction models are based exclusively on different coal characteristics. Among the limitations of these models, most are not valid when coal was weathered (oxidized) during storage. Other prediction models of coke strength, CRI, and CSR using coke structure and properties (ash chemistry, porosity parameters deduced from image analysis, optical textural components) have been extensively reported, as well as comparisons between quality of cokes produced at pilot and industrial scales. The latter constitute the third-generation of prediction models that use statistical analyses of cokes produced in a sole-heated oven, small pilot, and pilot coke ovens.

4. Coke production and demand

The blast furnace needs metallurgical coke and, therefore, the demand and market for both coal and coke will remain closely related to the steelmaking industry. In 1999, the world equivalent crude steel production accounted for 787 Mt with a hot metal production of about 540 Mt (Terjung, 2000; Terjung and Hermann, 2000; Wessiepe and Karsten, 2000). Consequently, the hot metal to crude steel ratio was approximately 700 kg t^{-1} crude steel. The growth of steel production is estimated for the next 15 years to be between an optimistic 975 Mt and a pessimistic 840 Mt. Increases in production could be especially high in the developing countries and could come from an expansion in electric furnace processes and a growth of hot metal production in existing blast furnaces (Derycke and Bonte, 2000). However, although the relative importance of electrical furnaces will increase during the next decades, especially in emerging countries, and will influence coal/coke demand, the blast furnace will continue to be the main process for crude steel production in the near future.

Coke quantity is the largest of the charge materials going into the blast furnace. In order to reduce costs and diminish operational and environmental problems, coke has been substituted in part by other fuels such as oil, granulated and pulverized coal (Cross, 1994), and more recently by plastic wastes (Janz and Weiss, 1996; Imai, 1999; Asanuma et al., 2000; Ohji, 2000). Following the use of PCI technology, the ratio coke/hot metal has changed from 1:1 (some 50 years ago) to 250 kg of both coal and coke per ton of hot
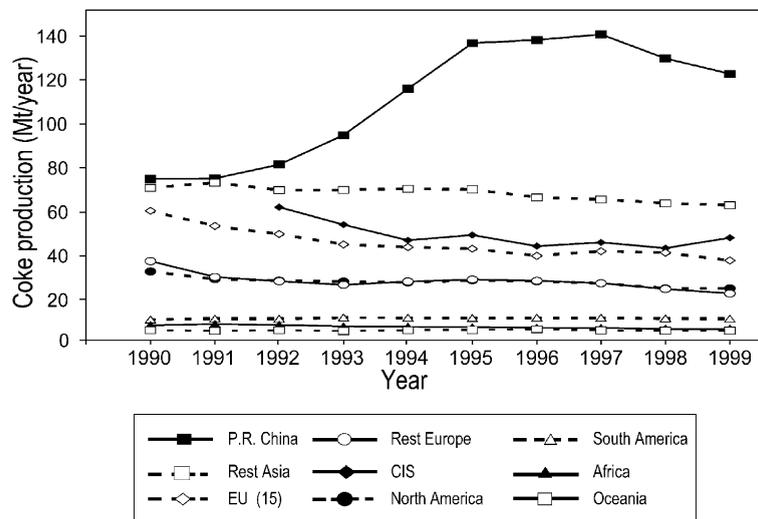

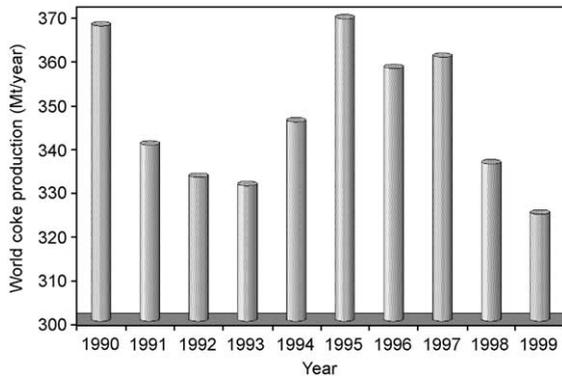Fig. 6. Evolution of coke production during the period 1990–1999.

Fig. 7. Worldwide coke production for the period 1990–1999.

metal. To achieve this PCI rate, especially high quality coke is needed together with advanced technologies both in coke production and blast furnace operation (Lüngen and Poos, 1996; Bertling, 1999; Terjung, 2000).

Demand for blast furnace coke in industrialized countries will drop due to higher PCI rates. Data taken from a world production review reported in the journal, Cokemaking International (2000), shows that coke production decreased in the early 1990s especially in Europe, North America, CIS, and Japan. This decline was compensated by Chinese coke production that has grown significantly from 43 Mt in 1980 to 139 Mt in 1997 (Fig. 6). From 1997, a decrease of about 18 Mt was reported in Chinese coke production. On a worldwide level, in 1997, there was a production of 360 Mt coke, falling to about 324 Mt in 1999 (Fig. 7). Fig. 8 illustrates the global distribution of coke production in 1999. Studies of the International Iron and Steel Institute (IISI) (1997) predict a shortage of coke by the year 2005, especially in Japan and USA (Hofherr, 2000). However, the world demand for the period 2000–2005 may be forecast at about 349 Mt of coke. A decline in coke production capacity relates to the age of installed cokemaking plants where huge investments would be needed to built new coke plants or renovate the old ones. The relative importance of an international coke trade is not that high; in 1999, it only accounted for about 5% of total coke production. The usual situation is that the coke is produced and consumed locally within integrated metallurgical works and steel mills. China during the last few years has become the biggest coke producer and exporter in the world, exporting to the USA, Brazil, and European countries. One of the problems of the use of imported coke is the lack of homogeneity and many consumers only use it as an admixing component.

Due to the double use of coal in the ironmaking industry, the situation of coal is quite different to that of the coke. On one side, amounts of coal for use in the blast furnace operating with PCI are expected to increase in the future (Trickett, 1999). No special
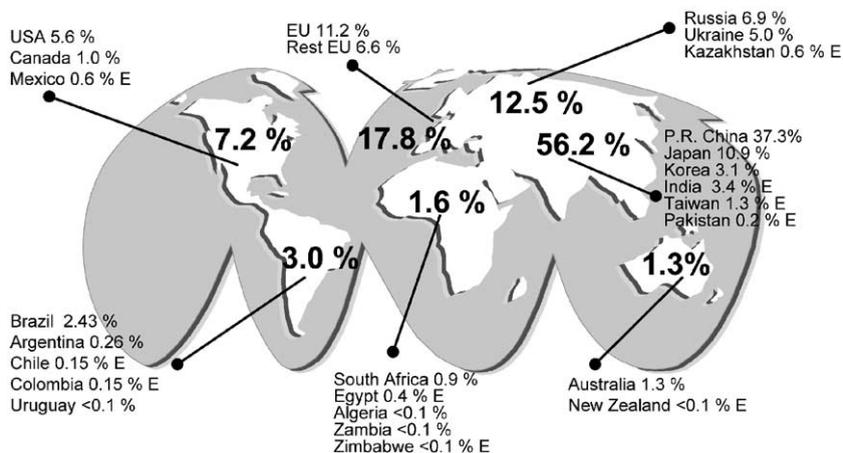

Fig. 8. Worldwide distribution of coke production on the basis of 324.4 Mt for 1999. E: estimated.

requirements are needed for such coals and they are readily available without problems. In general, carbon and hydrogen contents are key parameters for PCI as well as the absence of coking ability and a coal chemistry quite similar to that required for coking coals. On the other hand, coking coals are needed to produce metallurgical coke with very strict quality specifications. Generally speaking, coke is produced with a yield of 75 wt.%, so to produce 340 Mt within the 250 coking plants of the world, about 450 Mt of coking coal are needed. Excellent coking coals are produced in Australia, Canada, USA, and Poland. About 60% of coal mined is used in the country of origin and the rest is exported to Japan and Europe (Bertling, 1999). No shortage is expected in the next years, specially taking into account the high productivity of Australia and Canada. For the future, growth in consumption is only expected in countries such as China, Korea and Taiwan.

5. Coking processes of the future

On the industrial scale, present trends in coke manufacture are to use classical multi-chamber batteries with horizontal slot-like ovens and circular openings in the top for charging coal into the oven, removable doors through which the coke is discharged, laterally arranged heating walls in the upper oven section, regenerator heat exchangers in the lower section, and a substructure that includes waste heat ducts.

In conventional carbonisations, about 15 to 30 t of coal blend, crushed to 80% <3 mm particle size, is charged into the hot oven by gravity. The ovens are heated to a temperature of about 1200–1300 °C through the side-walls, which are kept at as uniform temperature as is possible. As the coal is gradually heated from both walls to the centre, a temperature gradient exists which decreases as the carbonization proceeds. This means that in the early stages of the carbonization process, the coal layer in contact with a side-wall is rapidly heated, while the centre of the coal charge remains at a relatively low temperature. When the temperature of the centre is 900 to 1000 °C, and after a period of soaking, the coking cycle is completed. A total period of 18–20 h is required for blast furnace coke production, the time depending on the oven dimensions. In the 1970, dimensions of by-product or slot-type coke ovens were commonly of 12–16 m length, 4–8 m height, and 400–450 mm width to ensure a good heat transmission from the oven wall to the centre. Very recent coke ovens in Germany have dimensions that exceed the recognized limits in all the three dimensions. Tables 3 and 4 summarize the state of the art in 1970s and 1980s, respectively, for a coke capacity of 2 Mt year^{-1} (Nashan, 1987). The gradual enlargement in chamber dimensions to near 8 m height, 16–18 m length, and 550–610 mm wide and in oven volume 70–79 m^3 reduces the number of ovens needed for the same production. Advantages claimed for these larger capacity ovens are: (a) low specific investment costs; (b) low heat losses; (c) small space requirements; and (d) improvements in environmental protection due to the decreased number of oven chamber openings as well as a shorter time period for charging. The Kaisertuhl III coking plant in Germany, with two batteries of 60 ovens each, represents the most advanced state of the development of the multi-chamber system incorporating the know-how and experience of more than 100 years. With this coking plant, inaugurated in December 1992, producing 2 Mt of coke per year, the development of multi-chamber systems reached the highest technological level in the last 100 years with successful results (Ameling et al., 1999).

Table 3
State of the art of coking plants with a capacity of 2 Mt coke per year in the 1970s (Nashan, 1987)

	Coking plant (size)		
	Small	Medium	Large
Height (m)	4.50	6.00	7.65
Length (m)	11.70	14.20	16.40
Width (mm)	450	450	435
Useful volume (m^3)	22.1	36.4	52.2
Productivity coke per oven (t)	22.1	36.4	52.2
Number of ovens	322	187	123
Total oven openings	2898	1496	984
Length of sealing faces (km)	10.5	6.9	5.1
Number of pushed ovens per day	430	257	171
Total opening cycles per day	3870	2056	1368
Length of sealing faces to be cleaned (km day^{-1})	14.0	9.5	7.2

Table 4
State of the art of coking plants with a capacity of 2 Mt coke per year in the 1980s (Nashan, 1987)

	Coking plant		
	Huckingen	Prosper	Kaiserstuhl III
Height (m)	7.85	7.10	7.63
Length (m)	17.20	15.90	18.00
Width (mm)	550	590	610
Useful volume (m^3)	70.0	62.3	78.9
Productivity coke per oven (t)	43.0	39.8	48.7
Number of ovens	120	142	120
Total oven openings	1080	1278	1080
Length of sealing faces (km)	6.0	6.2	5.5
Number of pushed ovens per day	128	138	115
Total opening cycles per day	1152	1242	1035
Length of sealing faces to be cleaned (km day^{-1})	5.6	6.0	5.3

Noting that future steel production will still be associated with conventional ironmaking via coke oven/blast furnace, the development of new coking systems is further accelerated by problems associated with the limitations reached in multi-chamber system; by current expenditures on pollution control being 25% and 30% of the specific investment and operating costs, respectively (Ameling et al., 1999); work place conditions to comply with strict environmental requirements; the maximum permissible wall pressure in multi-chamber systems which limits the design of coal blends; and finally the extreme age of existing coking batteries.

Concerning the age of coke oven batteries, they have a possible economical life of 20 to 30 years and an environmental life of 10 to 20 years. This means that an average service life is of about 25 years. The battery can operate for another 15 years, reaching a technical life of 30 to 40 years, but only with high cost maintenance. Because of the high investment cost of a new coke plant, the option of the extension of the lifetime is common practice. Improvements and developments in repair technology in the last decades have extended the life of coke batteries and provided optimum environmental behaviour. However, at present more than 50% of the installed capacity for coke manufacture is over 25 years of age (Nashan et

al., 2000a). But it has to be stressed that high quality coke will still be required by blast furnaces in sufficient quantity to ensure future iron and steel production. As a consequence, even with the use of PCI in blast furnaces, coke demand, in the short and medium term, cannot be met safely. Coke supplies from traditional exporting countries like China will also decline due to the closure of primitive and out-dated facilities. It is not surprising, then, that at the very beginning of the 21st century, several papers present opinions on cokemaking technology for the 21st century. The present status in individual countries and how to meet future demand have been analysed in the two volumes of the journal *Cokemaking International* (Blanco, 2000; Deshpande, 2000; Kiessling and Sundholm, 2000; Kovalev et al., 2000; Liu et al., 2000; Nashan et al., 2000b; Pensa, 2000; Rudyka, 2000; Sciazko, 2000).

On the basis of the above considerations, a stage may be reached at which coke producers will make decisions on the modernization and repairing of existing coking plants, on the construction of new batteries, or to access the restricted coke market. Starting at the end of the 20th century, the development of 21st century technology appears to be the reaction towards increasing future supplies. The new cokemaking technologies address the development of an environmentally friendly technology with high productivity and a more flexible utilization of weakly and non-coking coals in the blends carbonized.

Different approaches at different stages of development consider two-product technologies, non-recovery ovens, and single chamber system (SCS). They offer an interesting alternative to conventional coke ovens where the demand is only for coke and not for gas and other by-products such as benzol, tar, and ammonia. Other technologies being developed keep the conventional multi-chamber system with by-products recovery from coke-oven gas and waste water in additional installations, but introduce coal pre-treatment and coke post-treatment in special design installations (SCOPE 21).

5.1. The non-recovery coke oven

This new approach in cokemaking technology resembles the early beehive ovens that carbonized coal in horizontal layers, being of simple design,

and operation. In the Jewell–Thompson non-recovery ovens, the volatile products produced during coal carbonization are not recovered. The evolved gas, including benzol and tar, is burnt directly in the oven space above the coal, thus generating the heat needed for the process. The mixture of crude and waste gases is led through vertical ducts in the side-walls to the heating flue system under the oven sole. There the combustion is completed by the staged supply of air so that the coal layer is evenly heated from the top and bottom. The hot waste gas is cooled in waste heat boilers to raise steam, which can either be fed into the steam grid or used for generating electricity. As an estimation, about 75–95 MW electric power can be generated for a coke production of 1 Mt year^{-1}, depending on the volatile matter content of the coal and the composition of the coal blend charged. The exploitation of the waste gas is a major improvement over old beehive ovens of the 19th century (Ellis and Schuett, 1999; Walker, 1999; Westbrook and Schuett, 2000). The dimensions of the non-recovery ovens are about 13.5 m long, 4.6 m wide, and 2.4 m high at the crown of the arch, the thick coal layer being of approximately 1 m. The coal charged into the oven accounts weighs about 36 to 41.5 t (Ellis and Schuett, 1999) with a coking time of 48 h. Depending of the volatile matter of the blend carbonized, the charge weight, coke yield, and coking time will vary.

In USA, the Sun Coke non-recovery or heat recovery technology was first developed at the Jewell coke plant in Vansant, Virginia, producing 635,000 t of coke. Recently, the first high capacity plant (Indiana Harbor Coke facility) with four batteries of 67 heat-recovery ovens each, representing the advanced developed of the Jewell–Thompson non-recovery ovens, has been put into operation at Inland Steel Chicago. It was designed to produce about 1.2 Mt of screened coke per year and 94 MW of power generation. Fig. 9 displays a schematic diagram of this plant. Technical details are reported after 1 year (Ellis and Schuett, 1999) and 2 years (Westbrook and Schuett, 2000) of operation. This technology has emerged as a virtually clean technology. As the ovens operate under suction, no emissions during the coking process and wastes derived from by-products facilities are generated.

On the other hand, lateral coking pressure of a coal blend is much less important than in conventional coke ovens. This means that the use of coals or coal blends, which develop a higher coking pressure, is not a limitation and they can be carbonized without any danger for the non-recovery ovens. From 1998, different coal blends containing from three to six coals have been used at Indiana Harbor facility. Two types of coal blends were used, a relatively high-volatile blend of 28.6 wt.% dry basis and a low-volatile blend of 24.5 wt.% dry basis. As regards coke quality, CSR ranged from 67% to 72% and stability from 60% to 64%.

With this new technology, prediction models have been also developed for such coke properties, stability
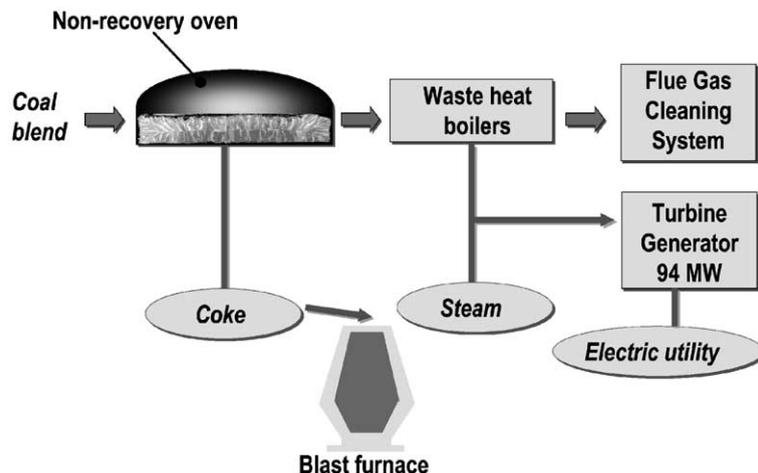

Fig. 9. Schematic diagram of the non-recovery oven system.

and CSR. The models are based on a so-called "fishbone" diagram, which includes the factors affecting coke quality grouped in different categories and subcategories.

5.2. Single chamber system (SCS)

In September 1987, during the first International Cokemaking Congress held in Essen, Germany, the single chamber or coking reactor system with two basic proposals for the design of the so-called Jumbo Coking Reactor—JCR (with underneath- and side-arranged regenerators) was presented (Nashan, 1987). To explore the possibilities of the JCR as a new cokemaking technology for the 21st Century, the European Community accepted a European project in 1990. For the EUREKA project development, the European Cokemaking Technology Centre (ECTC) was created with the leadership of the German coking industry and 11 companies representing seven European countries.

The SCS combines coal preheating to temperatures of 180 to 200 °C with dry coke quenching. It abandons the multi-chamber system in favour of a single and independent reactor and avoids the recovery of by-products from coke-oven gas, producing only two products—coke and hydrogen gas-rich product (Nashan, 1987, 1990, 1992). The recovered crude gas can be used as traditional energy source in steel mills, for steam and electric power generation, or as a reducing gas in iron ore reduction process (Ameling et al., 1999; Nashan et al., 2000c). Fig. 10 shows the concept of the SCS technology. In the 1990s, different stages of the construction and progress of the demonstration plant built in Prosper-Germany and results obtained were reported (Bertling and Rohde, 1994, 1995, 1996, 1997; Ameling et al., 1999; Baer, 2000; Nashan et al., 2000a, 2000c).

The single coking reactor built in 1992 and operated by ECTC, having dimensions of 10 m high, 10 m long, and 0.85 m wide, was charged with preheated coal and produce about 50 t coke per charging cycle. During the trial period, evaluations of coking conditions (bulk density, coking rate and time, and final coke temperature), emission control, coal blend characteristics (volatile matter content between 25 and 31 wt.%), and coke quality in terms of CRI and CSR parameters were made. As regards to coking pressure, the SCS is able to withstand high generated coking pressures due to the rigidity of the heating wall (>300 vs. 100 mbar in conventional multi-chamber system) (Nashan et al., 2000a, 2000c). Using high-volatile coals, the coke obtained was less porous as a result of the preheated coal charges, less reactive, and highly resistant to mechanical stresses (Bertling and Rohde, 1994). For comparison purposes with the SCS technology, the coal blends were also carbonized in multichamber system and the coke quality evaluated. Results using single coals, two- and four-component blends with amounts ranging from 30% to 80% of
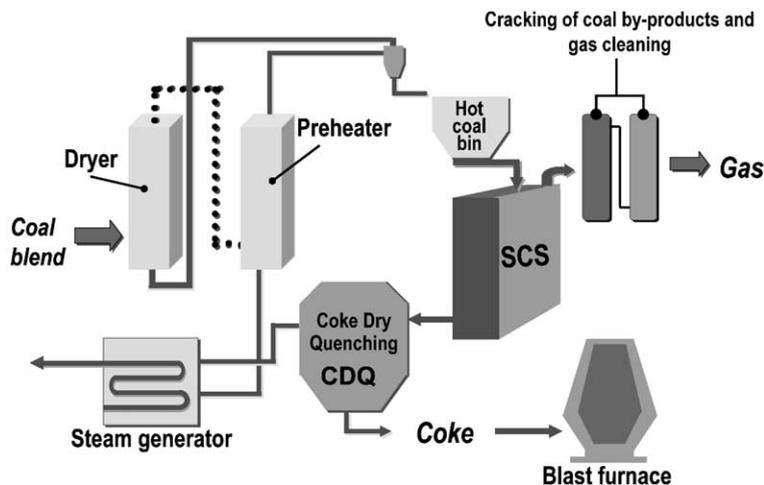

Fig. 10. Schematic diagram of the single chamber system (SCS).

poor-coking coals and 10% to 50% of non-coking coals (Bertling and Rohde, 1996) and multi-component blends, at the final stage of demonstration, showed the flexibility of the SCS in the use of a great proportion of cheaper poor- and non-coking coals in the blends and the improvement in coke quality, in particular the CSR values (Nashan et al., 2000a).

At present, the SCS, initially known as the Jumbo Coking Reactor, is proposed to be ready for an industrial scale application. A new stimulus from the cokemaking industry, at the beginning of the 21st century, is needed.

5.3. SCOPE 21

As an alternative to conventional coking process, the Japan Iron and Steel Federation is promoting the development of a new system under a project called the "Super Coke Oven for Productivity and Environment enhancement in the 21st century" (SCOPE 21). It is an 8-year project started in 1994 (Nishioka, 1996; Kubo, 1996; Sasaki et al., 1998; Nakashima, 1999; Taketomi et al., 2000).

In this new coking design, the conventional coking process is divided into three separated stages: the pre-treatment of coal blend, the medium-temperature coking process, and the upgrading of coke followed by coke dry quenching. The initial step consists of the separation of fine coal particles < 0.3 mm and coarse coal particles > 0.3 mm and the separated rapid heating to near to the temperature of thermal decomposition of coal (350–400 °C). The fine coal is agglomerated by

hot briquetting and charged into a conventional coke oven together with the preheated coarse coal. The next stage is a coking process at a much lower temperature (750–850 °C) than in conventional process. The coke is finally discharged and subjected to a further heat treatment up to 1000 °C in the upper part of the CDQ facility. Fig. 11 shows a schematic diagram of the SCOPE21 process.

A bench scale test plant with 0.6 t h^{-1} coal processing capacity was built at NSC Nagoya works. Test runs investigated the dry classification of coal, with rapid heating and compaction of fine coal. The results revealed that the combination of high bulk density, rapid heating and carbonization gave an acceptable coke quality even when using up to 50% poor- or non-coking coals. After considering the benefits of the new coking design in a blench scale, a pilot plant test is planned to be performed in 2001.

5.4. Calderon coking reactor

Another cokemaking approach in development is the Calderon Coking Reactor which is based on continuous coking of coal in a heated tubular reactor. The continuous process is designed as a two-step process, including carbonization at low temperature followed by calcination of the coke in a separate device with complementary hot gas cleanup system. In 1997, Bethlehem and US Steel joined with Calderon Energy to further develop the process and to check coal blends for the pilot scale plant (Strauss, 1999). The construction of a full size reactor is
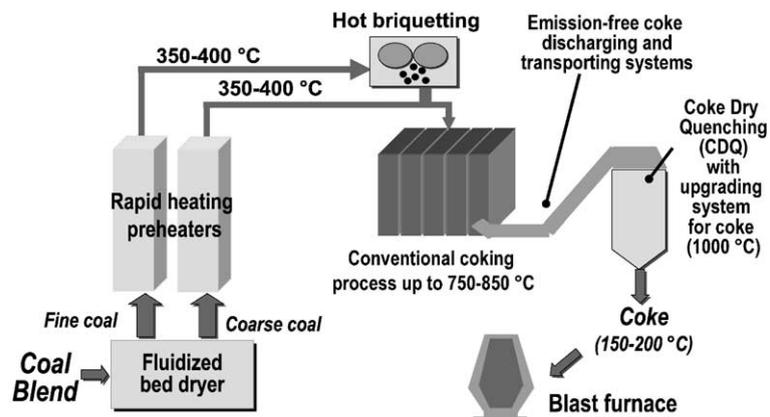

Fig. 11. Schematic diagram of the SCOPE21 cokemaking system.

underway and it is planned to test the coke in a commercial blast furnace.

At the very beginning of the 21st century, coal will continue to be a dual source in blast furnace technology with pulverized coal injected at the tuyeres and coke as a permeable support. Extensive R&D in coal blending and coke quality will be needed to meet the blast furnace requirements. With a short- and medium-time horizon, a new impulse is expected for clean and cost-effective cokemaking technologies at the actual stage of pilot scale trials and those at a more advanced state together with improvements in conventional cokemaking technology.

Acknowledgements

References

Alvarez, R., Díez, M.A., 2000. Chemistry of production of metallurgical coke. In: Marsh, H., Rodríguez-Reinoso, F. (Eds.), Sciences of Carbon Materials, Chap. 18. Secretariado de Publicaciones, The University of Alicante, Spain, pp. 595–635.

Ameling, D., Baer, H., Bertling, H., Lüngen, H.B., 1999. Cokemaking technology 2000. State-of-the-art and new structures. Cokemak. Int. 11 (1), 32–39 (also in: Technical Exchange Session. Int. Iron and Steel Institute (IISI), Committee on Technology, Brussels, 1999, pp. 30–45).

Ammosov, I.I., Eremin, I.V., Sukhenko, S.F., Oshurkova, L.S., 1957. Calculation of coking changes on basis of petrographic characteristics of coals. Coke Chem. U.S.S.R. 2, 9–12.

Asanuma, M., Ariyama, T., Sato, M., Murai, R., Nonaka, T., Okochi, I., Tsukiji, H., Nemoto, K., 2000. Development of waste plastics injection process in blast furnace. ISIJ Int. 40, 244–251.

Baer, H., 2000. Cokemaking with the single chamber coking system. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 659–663.

Barsotti, A., Damiani, R., 1988. Coal properties evaluation and blending philosophy at Italsider as mean for predetermining coke characteristics according blast furnace requirements. Proc. 47th Ironmaking Conf., ISS-AIME, pp. 163–171.

Barriocanal, C., Hanson, S., Patrick, J.W., Walker, A., 1994. The characterization of interfaces between textural components in metallurgical cokes. Fuel 73, 1842–1847.

Barriocanal, C., Alvarez, R., Canga, C.S., Díez, M.A., 1998. On the possibility of using coking plant waste materials as additives for coke production. Energy Fuels 12, 981–989.

Benedict, L.G., Thompson, R.R., 1976. Selection of coals and coal mixes to avoid excessive coking pressure. Proc. 35th Ironmaking Conf., ISS-AIME, pp. 276–286.

Benedict, L.G., Thompson, R.R., Wenger, R.O., 1968a. Relationship between coal petrographic composition and coke stability. Blast Furn. Steel Plant 56 (3), 217–224.

Benedict, L.G., Thompson, R.R., Shigo, J., Aikinan, R., 1968b. Pseudovitrinite in Appalachian coking coals. Fuel 47, 125–143.

Bernard, A., Duchene, J.M., Isler, D., 1985. Etude de la gazeification du coke. Rev. Met. CIT 82, 849–860.

Bernard, A., Duchene, J.M., Vogt, D., Jeulin, D., Steiler, J.M., Bourrat, X., Rouzaud, J.N., 1986. Effect of rank and of interactions of coals on experimental coke properties. Proc. 45th Ironmaking Conf., ISS-AIME, pp. 211–219.

Bertling, H., 1999. Coal and coke for blast furnace. ISIJ Int. 39 (7), 617–624.

Bertling, H., Rohde, W., 1994. Jumbo Coking Reactor. First operational experience with a new cokemaking technology. Cokemak. Int. 6 (1), 8–11.

Bertling, H., Rohde, W., 1995. Jumbo Coking Reactor. Progress report. Cokemak. Int. 7 (2), 20–25.

Bertling, H., Rohde, W., 1996. Jumbo Coking Reactor (JCR) and two product coking process. Proc. 3rd European Cokemaking Cong., CRM-VDEh, Gent, Belgium, pp. 273–278.

Bertling, H., Rohde, W., 1997. Coke quality improvements by SCS-Technology. Cokemak. Int. 9 (1), 31–34.

Blanco, R., 2000. The coal and coke industries in Colombia. Cokemak. Int. 12 (2), 56–57.

Brown, H.R., Taylor, G.H., Cook, A.C., 1964. Prediction of coke strength from the rank and petrographic composition of Australian coals. Fuel 43, 43–54.

Bustin, R.M., Cameron, A.R., Grieve, D.A., Kalkreuth, W.D., 1985. Coal petrology. Its principles, methods and applications, 2nd revised ed. Geological Assoc. of Canada, Short Course Notes, Victoria, British Columbia, vol. 3, pp. 167–203.

Caldeira, J.S., Da Silva, P.H.L., 1988. Microscopy as a means to assess coke mechanism and behaviour under reactive conditions. Proc. 47th Ironmaking Conf., ISS-AIME, pp. 357–360.

Carr, A.J., Jorgensen, J.G., 1975. An estimate of the effective reactive/inert ratio of semifusinite in Western Canadian coals. CANMET Rep. 75-141, 12 pp. (ref. taken from Gransden, J.F. et al., 1991).

Chiu, Y.F., 1982. Study of coke petrography and factors affecting coke reactivity. Ironmak. Steelmak. 9 (5), 193–199.

Cokemaking International, 2000. World production review. Cokemak. Int. 12 (2), 72–78.

Colleta, A., Barnaba, P., Federico, G., Masella, D.C., 1990. Influence of coal properties on high coke quality for blast furnace. Proc. 49th Ironmaking Conf., ISS-AIME, pp. 243–252.

Cross, W.B., 1994. The future and the European steel industry and its demand for coal. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, pp. 97–108.

Derycke, J., Bonte, L., 2000. Ironmaking perspectives for the early 21st Century. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 693–702.

Deshpande, D.P., 2000. Cokemaking technology in India. Cokemak. Int. 12 (2), 60–62.

Díez, M.A., Alvarez, R., Sirgado, M., Marsh, H., 1991. Preheating

techniques to manufacture metallurgical coke. ISIJ Int. 31, 449–457.

Dutcher, L.A.F., Crelling, J.C., 2000. History of applied coal petrology in the United States: I. Early history of the application of coal petrology in the steel industry. Int. J. Coal Geol. 42, 93–101.

Edwards, D.G. (Ed.), 1991. The strength of coke after partial gasification. BCRA Quarterly, Chesterfield, UK, No. 30, pp. 37–59; 53–76.

Ellis, A.R., Schuett, J., 1999. Heat recovery cokemaking at Indiana Harbor coke company—one year of operation. Cokemak. Int. 11 (2), 40–49.

Falcon, R.M.S., Snyman, C.P., 1986. An introduction to Coal Petrography: Atlas of Petrographic Constituents in the Bituminous Coals of Southern Africa. The Geology Society of South Africa, Johannesburg, 27 pp.

Fujita, H., Hijiriyama, M., Nishida, S., 1983. Gasification reactivities of optical textures of metallurgical cokes. Fuel 62, 875–879.

Gibson, J., 1972. Dilatometry and the prediction of coke quality. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, pp. 182–205.

Gibson, J., Gregory, D.H., 1978. Selection of coals and blend preparation for optimum coke quality. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, pp. 159–182.

Goscinski, J.S., Patalsky, R., 1990. CSR control. A coal producers point of view. Proc. 49th Ironmaking Conf., ISS-AIME, pp. 53–74.

Goscinski, J.S., Gray, R.J., Robinson, J.W., 1985. A review of American coal quality and its effect on coke reactivity and after reaction strength of cokes. J. Coal Qual. 4 (2), 21–29.

Gransden, J.F., Jorgensen, J.G., Manery, N., Price, J.T., Ramey, N.J., 1991. Applications of microscopy to cokemaking. Int. J. Coal Geol. 19, 77–107.

Großpietsch, K.H., Lüngen, H.B., Dauwels, G., Ferstl, A., Karjalahti, T., Negro, P., van der Velden, B., Willmers, R., 2000. Coke quality requirements by European blast furnace operators on the turn of the millennium. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 1, pp. 2–11.

Hara, Y., Mikuni, O., Yamamoto, H., Yamanaka, Y., 1980. The assessment of coke quality with particular emphasis on sampling technique. In: Lu, W.K. (Ed.), McMaster Symposium on Iron and Steelmaking. McMaster Symposium, McMaster University, vol. 8, pp. 4.1–4.38.

Harrison, J.A., Jackman, H.W., Simon, J.A., 1964. Predicting coke stability from petrographic analysis of Illinois coals. Illinois State Geological Survey, Circular, vol. 366, 20 pp.

Hatano, M., Miyazaki, T., Iwanaga, Y., 1990. Analysis of blast furnace operation based on the mathematical model for evaluation of coke properties. Coke Reactivity and its Effects on Blast Furnace Operation. The Iron and Steel Society, pp. 21–34.

Hofherr, K., 2000. State of the art in cokemaking and future development. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 686–692.

Horrocks, K.R.S., Cunningham, R.B., Ellison, J.F., Nightingale, R.J., 2000. Coke quality at BHP Steel Port Kembla. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 1, pp. 167–173.

Hyslop, W., 1981. Carbon solution and coke quality. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, pp. 131–182.

Imai, T., 1999. Production and technology of iron and steel in Japan during 1998. ISIJ Int. 39, 509–523.

International Iron and Steel Institute (IISI), 1997. Coke and its Alternatives. Committee on Raw Materials, Brussels, 162 pp.

Ishikawa, I., Case, M., Abe, Y., Ono, K., Sugata, M., Nishi, T., 1990. Influence of post-reaction strength of coke on blast furnace operation. Coke Reactivity and its Effects on Blast Furnace Operation. The Iron and Steel Society, pp. 37–48.

Janz, J., Weiss, W., 1996. Injection of waste plastics into the blast furnace of Stahlwerke Bremen. Proc. 3rd European Cokemaking Cong., CRM-VDEh, Gent, Belgium, pp. 114–119.

Kaegi, D.D., Valia, H.S., Harrison, Ch.H., 1988. Maceral behaviour and coal carbonisation. Proc. 47th Ironmaking Conf., ISS-AIME, pp. 339–349.

Kiessling, F., Sundholm, J., 2000. Cokemaking in the United States. Cokemak. Int. 12 (2), 67–68.

Kovalev, E.T., Sklyar, M.G., Shulga, I.V., Starovoit, A.G., 2000. Main trends of development of cokemaking facilities in Ukraine. The view and assessment of UKhIN and UKRKOKS. Cokemak. Int. 12 (2), 64–65.

Kubo, S., 1996. Recent trends in the Japanese steel and coke industries and the concepts of a new cokemaking process. Proc. 3rd European Cokemaking Cong., CRM-VDEh, Gent, Belgium, pp. 279–284.

Leeder, W.R., Price, J.T., Gransden, J.F., 1997. Western Canadian coals. Thermal and rheology and coking quality. Proc. 56th Ironmaking Conf., ISS-AIME, pp. 37–44.

Leonard, D.C., Bonte, L., Dufour, A., Ferstl, A., Raipala, K., Scmole, P., Schoone, P., Verduras, J.L., Willmers, R.R., 1996. Coke quality requirements of European blast furnace engineers (joint EBFC-Paper). Proc. 3rd European Cokemaking Cong., CRM-VDEh, Gent, Belgium, pp. 1–10 (also in: The Coke Oven Managers' Association (COMA) Year-Book, Rotherham, 1997, pp. 279–303).

Liu, H., Cai, Ch., Zheng, W., 2000. China's coking industry stepping into the new century. Cokemak. Int. 12 (1), 48–53.

Loison, R., Foch, P., Boyer, A., 1989. Coke. Quality and Production. Butterworth, London, pp. 178–189.

Lüngen, H.B., Poos, A., 1996. Injection of coal into the blast furnace. Cokemak. Int. 9 (2), 14–31.

Mackowsky, M.Th., Simonis, W., 1969. Die Keenzeichnung von Kokskohlen fur die mathematische Beschreibung der Hochtemperaturverkokung in Horizontalkammeröfen bei Schuttebetrieb durch Ergebnisse mikroskopischer Analysen. Gulckauf-Forschungsheft 30, 25–37.

Marsh, H., 1982. Metallurgical coke: formation, structure and properties. Proc. 41st Ironmaking Conf., ISS-AIME, pp. 2–11.

Marsh, H., 1992. The coke making process. Proc. 51st Ironmaking Conf., ISS-AIME, pp. 569–580.

Marsh, H., Clarke, D.E., 1986. Mechanisms of formation of structure within metallurgical coke and its effects on coke properties. Erdöl Kohle Erdgas Petrochem. 39, 113–122.

Marsh, H., Walker Jr., P.L., 1979. The effects of impregnation of

coal by alkali salts upon carbonisation properties. Fuel Process. Technol. 2, 61–75.

McKenzie, F., Hockings, K., Livingstone, P., 1998. Modelling of coal blending to predict coke quality. Proc. AIE 8th Australian Coal Science Conf., Sydney, pp. 101–106.

Menéndez, J.A., Pis, J.J., Alvarez, R., Barriocanal, C., Fuente, E., Díez, M.A., 1996. Characterization of petroleum coke as an additive in metallurgical cokemaking. Modification of thermoplastic properties of coal. Energy Fuels 10, 1262–1268.

Menéndez, J.A., Pis, J.J., Alvarez, R., Barriocanal, C., Canga, C.S., Díez, M.A., 1997. Characterization of petroleum coke as an additive in metallurgical cokemaking. Influence on metallurgical coke quality. Energy Fuels 11, 379–384.

Menéndez, J.A., Alvarez, R., Pis, J.J., 1999. Determination of metallurgical coke reactivity at INCAR: NSC and ECE-INCAR reactivity tests. Ironmak. Steelmak. 26, 117–121.

Miura, Y., 1978. The science of cokemaking technology and its development in Japan. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, pp. 292–311.

Miyazu, T., 1974. The evaluation and design of blends using many kinds of coals for cokemaking. Int. Iron Steel Cong., Dusseldorf, Pap. 1.2.2.1.

Morishita, N., Tsukada, K., Suzuki, N., Nemoto, K., 1986. Development of automatic coal/coke microscopic analyzer and its application to cokemaking. Proc. 45th Ironmaking Conf., ISS-AIME, pp. 203–209.

Moses, R., 1976. Use of Petrographic Analysis for the Prediction of Coke Stability Report Bituminous Coal Research, Monroeville, PA, 19 pp.

Munnix, R., 1984. Coke strength prediction based on coal petrography and fluidity measurements. Proc. 43rd Ironmaking Conf., ISS-AIME, pp. 19–27.

Nakamura, N., Togino, Y., Tateoka, T., 1977. Behaviour of coke in large blast furnace. Coal, Coke and Blast Furnace. The Metals Society, London, pp. 1–18.

Nakashima, Y., 1999. Scope 21 cokemaking process. Technical Exchange Session. Int. Iron and Steel Institute (IISI), Committee on Technology, Brussels, pp. 16–29.

Nandi, B.N., Montgomery, D.S., 1975. Nature and thermal behaviour of semi-fusinite in Cretaceous coal from Western Canada. Fuel 54, 193–196.

Nashan, G., 1987. The production of blast-furnace coke can be secured in the long term ecologically beneficial and cost-efficiently. On the way to a new coking system. Preprints 1st Int. Cokemaking Congr., vol. 1, A2. Verlag Glückauk, Essen, 51 pp.

Nashan, G., 1990. European development centre for cokemaking technology—tasks and targets. Cokemak. Int. 2 (2), 19–29.

Nashan, G., 1992. Economic fundaments for new cokemaking system and a new process structure of a coke plant—Jumbo Coking Reactor. Proc. 51st Ironmaking Conf., ISS-AIME, pp. 349–358.

Nashan, G., Rohde, W., Wessiepe, K., Winzer, G., 2000a. Modular and 2-product technology. The cokemaking process for the future. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 646–653.

Nashan, G., Rohde, W., Wessiepe, K., 2000b. Long term perspectives for cokemaking technology from individual country's view. Cokemak. Int. 12 (1), 44–47.

Nashan, G., Rohde, W., Wessiepe, K., 2000c. Some figures and facts on the present status and new proposals for a future-oriented cokemaking technology. Cokemak. Int. 12 (2), 50–55.

Negro, P., Steiler, J.M., Beppler, E., Jahusen, U., Bennington, C.R., Willmers, R.R., 1996. Assessment of coke degradation in the blast furnace from tuyères probing investigations. Proc. 3rd European Ironmaking Cong., CRM-VDEh, Gent, Belgium, pp. 20–27 (also in: The Coke Oven Managers' Association (COMA) Year-Book, Rotherham, UK, 1997, pp. 256–278).

Nishioka, K., 1996. Challenge for innovative cokemaking process in Japan. Proc. 3rd European Cokemaking Cong., CRM-VDEh, Gent, Belgium, pp. 285–290.

Nishioka, K., 2000. History and future of the Japanese cokemaking technology. J. Jpn. Inst. Energy 79 (876), 284–298 (in Japanese).

O'Donnell, E.M., Poveromo, J.J., 2000. Coke quality requirements from a North American perspective. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 1, pp. 12–19.

Oguri, K., Kitahara, A., Hijiriyama, M., Nishida, S., Aketa, K., 1987. Evaluation of coking coal. Preprints 1st Int. Cokemaking Cong., vol. 1, E1. Verlag Glückauk, Essen, 12 pp.

Ohji, M., 2000. Production and technology of iron and steel in Japan during 1999. ISIJ Int. 40, 529–543.

Okuyama, Y., Miyazu, T., Sugimura, H., Kumagai, M., 1970. Prediction of the coking property of coal by microscopic analysis. J. Fuel Soc. Jpn. 49 (522), 736–743.

Patrick, J.W., Walker, A., 1995. Porosity and strength of carbon materials. In: Patrick, J.W. (Ed.), Porosity in Carbons: Characterization and Applications, Chap. 7. Edward Arnold, UK, pp. 195–208.

Patrick, J.W., Wilkinson, H.C., 1978. Analysis of metallurgical cokes. In: Karr Jr., C. (Ed.), Analytical Methods for Coal and Coal Products, vol. II. Academic Press, New York, pp. 339–370.

Patrick, J.W., Wilkinson, H.C., 1981. Coke reactivity. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, UK, pp. 191–220.

Pearson, D.E., Price, J.T., 1985. Reactivity of inertinite (coal-typing) of Western Canadian coking coal. Proc. Int. Conf. on Coal Science. Pergamon Press, Sydney, pp. 907–908.

Pensa, R., 2000. Future perspectives for cokemaking technology: current situation in Italy. Cokemak. Int. 12 (2), 58–59.

Poos, A., 1987. Constitution and monitoring of coking coal blends. The Coke Oven Managers' Association (COMA) Year-Book, Mexborough, UK, pp. 223–253.

Poveromo, J.J., 1996. Operational considerations for high-level blast furnace fuel injection. Proc. 55th Ironmaking Conf., ISS-AIME, pp. 79–92.

Price, J.T., Gransden, J.F., Khan, M.A., 1988. Effect of the properties of Western Canadian coal on their coking behaviour. Proc. 47th Ironmaking Conference, ISS-AIME, Toronto, pp. 39–55.

Price, J.T., Gransden, J.F., Khan, M.A., Ryan, B.D., 1992. Effects of selected minerals on high temperature properties of coke. Proc. 2nd Int. Cokemaking Cong., The Institute of Materials, London, vol. 1, pp. 287–292.

Ragan, S., Marsh, H., 1980. A critique of industrial methods of measurement of strength of metallurgical coke. J. Phys. D: Appl. Phys. 13, 983–993.

Rooney, K.A., Gregory, O.R., Nightingale, R.J., Price, K., 1987. Developments in coke quality and the improved performance of a 300 cubic meter blast furnace. Preprints 1st Int. Cokemaking Cong., Essen, vol. 1, C3, 17 pp.

Rudyka, V., 2000. Main trends of development of cokemaking facilities in Ukraine. The view and assessment of Giprokoks. Cokemak. Int. 12 (2), 66–67.

Sakawa, M., Sakurai, Y., Hara, Y., 1982. Influence of coal characteristics on CO_2 gasification. Fuel 61, 717–720.

Sasaki, M., Komaki, I., Matsuura, M., Saito, K., Fukada, K., 1998. A new carbonization process with high temperature preheating. Proc. 57th Ironmaking Conf., ISS-AIME, pp. 803–809.

Schapiro, N., Gray, R.J., 1960. Petrographic classification applicable to coals of all ranks. Proc. Illinois Min. Inst., pp. 83–90.

Schapiro, N., Gray, R.J., 1964. The use of coal petrography in cokemaking. J. Inst. Fuel 43, 234–242.

Schapiro, N., Gray, R.J., Eusner, G.R., 1961. Recent developments in coal petrography. Blast Furnace, Coke Oven and Raw Materials Committee, Proceedings, ISS-AIME, vol. 20, pp. 89–112.

Sciazko, M., 2000. Future of coke production in Poland. Cokemak. Int. 12 (2), 63–64.

Sirgado, M., Verduras, J.L., 1978. Coking of complex coal mixtures, pre-estimating of ironmaking coke quality and development of an economical optimization pattern for coking mixtures. Proc. Int. Iron and Steel Cong., Chicago, 404–420.

Spackman, W., 2000. History of applied coal petrology in the United States: II. A personalized history of the origin and development of applied coal petrology at the Pennsylvania State University. Int. J. Coal Geol. 42, 103–114.

Stach, E., Mackowski, M.Th., Teichmüller, M., Taylor, G.H., Chandra, D., Teichmüller, R., 1982. Stach's Textbook of Coal Petrology, 3rd ed. Gebrüder Borntraeger, Berlin.

Steyn, J.G.D., Smith, C.H., 1977. Coal petrography in the evaluation of South African coals. Coal, Gold and Base Minerals of Southern Africa, pp. 107–117.

Stopes, M.C., 1919. On the four visible ingredients of banded bituminous coals. Proc. R. Soc. B 90, 470–487.

Stopes, M.C., 1935. On the petrology of banded bituminous coals. Fuel 14, 4–13.

Strauss, A.D., 1999. Evaluation of metallurgical coal blends in the Calderon coking reactor. Cokemaking Technology. Technical Exchange Session. Int. Iron and Steel Institute (IISI), Committee on Technology, Brussels, pp. 9–15.

Suárez-Ruiz, I., 2000. Coal petrology and reflectance microscopy. In: Marsh, H., Rodríguez-Reinoso, F. (Eds.), Sciences of Carbon Materials. Secretariado de Publicaciones, Chap. 16. The University of Alicante, Spain, pp. 533–569.

Taketomi, H., Nishioka, K., NaKashima, Y., Suyama, S., Matsuura, M., 2000. Research on coal pre-treatment process of Scope 21. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 640–645.

Taylor, G.H., 1957. The behaviour of petrological components of coal on carbonization. Fuel 36, 221–235.

Taylor, G.H., Mackowsky, M.Th., Alpern, B., 1967. The behaviour of inertinite during carbonisation. Fuel 46, 431–440.

Terjung, J., 2000. World coal and coke market presence and future. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 20–29.

Terjung, J., Hermann, W., 2000. Present tendencies on the world coke market. Cokemak. Int. 12 (1), 32–37.

The British Carbonization Resarch Association (BCRA), 1980. The evaluation of the Nippon Steel Corporation reactivity and post-reaction strength test for coke. Carbonization Research Report 91, Chesterfield, UK, 50 pp.

Thompson, R.R., 2000. History of applied coal petrology in the United States: III. Contributions to applied coal and coke petrology at the Bethlehem Steel Corporation. Int. J. Coal Geol. 42, 115–128.

Thompson, R.R., Benedict, L.G., 1976. Coal composition and its influence on cokemaking. Iron Steelmak. 3 (2), 21–31.

Trickett, A., 1999. The outlook for coke supply and demand. Cokemak. Int. 11 (1), 28–31.

Tucker, J., Everitt, G., 1992. Coking pressure—its causes, measurement and control. Proc. 2nd Int. Cokemaking Cong., vol. 2. The Institute of Materials, London, pp. 40–61.

United Nations Economic Commission for Europe (UNECE), 1965. Method of measuring reactivity of metallurgical coke. ST-ECE-COAL-12, United Nations New York, 17 pp.

Valia, H.S., 1987. Predicting coke strength after reaction with CO_2 from coal analysis. Proc. 46th Ironmaking Conf., ISS-AIME, p. 78.

Valia, H.S., 1989. Prediction of coke strength after reaction with CO_2 from coal analyses at Inland Steel Company. Iron Steelmak., 77–87 (also in: Coke Reactivity and its Effect on Blast Furnace Operation, ISS-AIME, 1990, pp. 109–119).

Vander, T., Alvarez, R., Ferraro, M., Fohl, J., Hofherr, K., Huart, J.M., Mattila, E., Propson, R., Willmers, R.R., van der Velden, B., 1996. Coke quality improvement. Possibilities and limitations. Proc. 3rd European Ironmaking Cong., CRM-VDEh, Gent, Belgium, pp. 28–37 (also in: The Coke Oven Managers' Association (COMA) Year-Book, Rotherham, UK, 1997, pp. 304–328).

Vogt, D., 2000. Coke strength after reaction. Signification regarding with various operation and various cokes. Proc. 59th Ironmaking Conf., ISS-AIME, pp. 47–54.

Walker, D.N., 1999. Sun Coke heat recovery coke technology at Indiana Harbor. Cokemaking technology. Technical Exchange Session. Int. Iron and Steel Institute (IISI), Committee on Technology, Brussels, pp. 1–8.

Wessiepe, K., Karsten, E., 2000. Recent developments in the world coke industry. Coke demand/coke production. Cokemak. Int. 12 (1), 38–43.

Westbrook, R.W., Schuett, K.J., 2000. Two years of heat recovery coke production at Sun coke company's Indiana Harbor facility. Proc. 4th European Coke and Ironmaking Cong., ATS-RM, Paris, vol. 2, pp. 654–658.