An Application of Coke Microstructure and Microtexture to Indonesian Coal

YA-WEN CHEN

Iron & Steel Research & Development Department China Steel Corporation

Taiwan is located at a compressive tectonic area and lack of natural resources; therefore most industrial feedstocks are imported. Metallurgical coal and iron ore are two main ingredients in steelmaking. It is necessary to be conversant with the characteristics from different resource areas. Indonesia is playing a major role in the coal market since Indonesian coal benefits from low ash and sulfur contents. Due to the ideal geographic position for Asian coal demand, it is time to engage in the study of Indonesian metallurgical coal. Basic coal and coke analysis are undertaken to generalize the chemical and physical properties. Nevertheless, the coke strength after a high temperature is unpredictable. Coal and coke petrographic methods are attempted to correlate the qualities of coke with coal. High vitrinite contents are found in most of Indonesian coals, and therefore the reactive maceral derived components are abundant in coke microtexture. The lack of inert maceral derived components and thin coke walls are discovered from the coke microstructure. Both microtexture and microstructure are adopted to confer the weak coke strength after reaction of Indonesian coal.

Keywords: Indonesian coal, Metallurgical coal, Maceral, Reactive maceral derived components, Inert maceral derived components

1. INTRODUCTION

Metallurgical coke is a fuel, a chemical reducing agent and a permeable support in the operation of blast furnaces and consequently stable coke quality and support are the fundamental need to maintain the process of iron production. Taiwan lacks the natural resources for most of the raw materials for iron making and counts on the exports of Australia, Canada, America and Brazil. Broadening the resource usage from different regions is a significant task of permanent operation for China Steel (CSC). Indonesian metallurgical coal commenced commercial production much later than Australian and American coals which were older in geological age. Unfamiliar coking characteristics led to unexpected coke quality and operation problem and therefore detail investigation of Indonesian coal is essential due to the great benefit of reduced shipping time.

The prediction of coke quality in coke production is critical for iron making and the linkage between coal properties and coke characteristics was investigated to fill the vacancy in our coal research project. The coke's physical properties and mechanical strength are used to control the coal blending sufficiency and porous structure which are the main determined properties. With increasing advances in imaging techniques, several studies have shown that microtexture and microstructure of coke identification were developed and connected to coke strength⁽¹⁻⁴⁾. The microtexture of the coke walls are the main support against compaction, degradation and abrasion during its descent down in to the blast furnace. The microstructureal features included pore size, pore density, and shape are related to the fracture creation that occurs through chemical reduction and degradation. This work adopted the current optical analysis methods to characterize microtexture and microstructure of Indonesian coals and the behavior of weak coke strength was discussed.

2. EXPERIMENTAL METHOD

2.1 Raw materials

Different origins of metallurgical coal are used to make out the regional geological diversity. This study analyzes twenty coal samples from Australia, Indonesia, America and Africa to adjust the characteristics of various sedimentary backgrounds. There have been three main geological periods with coal deposits which are Carboniferous, Permian and Jurassic-Tertiary (Fig.1). Indonesian coals are predominantly in Tertiary rather than Permian which Australian and South African coal deposits were formed in⁽⁵⁾. Deposited in these tectonically active regions, the low rank coals have been affected by post-formation igneous activity. The coking and caking properties of Indonesian coals are influenced by their geological environments and varies from mine to mine⁽⁷⁾. Due to the lower degree of compaction and induration of coal seams, most coals exhibit a high volatile matter and low coke strength which are shown in Table 1.



Fig.1. The geological ages of the world's major hard coal and lignite deposits⁽⁶⁾.

2.2 Coal analysis

2.2.1 Proximate and Ultimate analyses and ash composition

The fundamental standard practices of coal and coke are Proximate and Ultimate analyses. Proximate analysis covers the determination of moisture, volatile matter, ash and the calculation of fixed carbon⁽⁸⁾, which is able to show the rank of coals and the ratio of combustible to incombustible constituents. It is also an index to evaluate the beneficiation of coal for business transactions. Ultimate analysis is intended for the general utilization by applicable industries and the determination of carbon, hydrogen, sulfur, nitrogen and the calculation of oxygen⁽⁹⁾. The Ultimate analysis provides a convenient and uniform system for comparing coal and coke.

Ash composition is determined through major and minor elements by using X-Ray Fluorescence (XRF) techniques⁽¹⁰⁾. It is useful to be able to predict slagging and fouling characteristics of combusted materials and the mineral matter in the coal has an important influence on the coke quality^(11,12).

Sample		VM (db)	ASH (db)	S (daf)	P ₂ O ₅ (in coal)	BAR	Max Fluidity	Ro	Vitrinite (%)	CSR (15 lb)	m ₃₀ (15 lb)
- - HV _ -	IN - 1	40.1	5.2	0.21	0.067	0.257	281	0.88	86.0	11.5	14.7
	AU - 1	35.8	6.4	0.64	0.281	0.100	1687	0.96	71.8	44.9	67.5
	AM - 1	33.1	6.2	0.98	0.024	0.149	6125	1.12	68.6	-	-
	IN - 2	35.4	7.4	0.74	0.014	0.181	1717	0.98	90.0	-	-
	AU - 2	35.8	8.1	0.51	0.024	0.078	71	0.79	70.8	-	-
	IN - 3	35.5	9.7	0.66	0.056	0.052	23412	1.16	93.4	29.4	65.3
	IN - 4	32.0	5.5	1.22	0.018	0.187	4485	1.03	95.7	40.9	60.5
MHV _	AF - 1	25.7	10.9	0.90	0.298	0.078	475	1.14	82.6	60.6	81.7
	AM - 2	25.8	9.4	0.63	0.186	0.086	233	1.09	64.4	63.3	71.7
	IN - 5	27.6	9.0	0.85	0.086	0.179	213	1.15	87.7	40.5	67.7
	AU - 3	28.8	9.2	0.53	0.222	0.124	1626	1.14	65.4	51.9	73.3
	AU - 4	27.8	9.0	0.37	0.368	0.118	35	0.99	58.2	41.0	74.6
 MLV	AU - 5	23.7	9.4	0.48	0.130	0.075	892	1.33	60.4	-	-
	AF - 2	25.0	10.3	0.83	0.278	0.077	757	1.36	80.3	61.9	85.0
	AU - 6	23.7	7.1	0.49	0.186	0.204	53	1.23	65.8	46.7	72.2
	AM - 3	22.7	7.7	0.50	0.039	0.066	20	-	73.8	49.8	70.9
	AF - 3	24.6	10.7	1.04	0.229	0.114	450	1.32	87.9	50.4	81.3
LV	AU - 7	19.5	10.6	0.62	0.074	0.082	31	1.52	72.0	70.5	75.2
	AM - 4	17.7	7.6	0.46	0.291	0.094	1.5	1.52	66.4	60.2	68.2
	IN - 6	19.3	6.5	0.39	0.007	0.168	35	1.76	98.3	15.5	61.0

Table 1 Coal and coke properties of samples from Australia, Indonesia, America and Africa

2.2.2 Coal petrography

Both maceral composition and vitrinite reflectance of coals are adopted to be an important index for the contents of organic substances and coal rank. Coal macerals are the complex of biological units which originate from plant tissues, secretions and exudates deposited into the peat. It has gone through several alteration processes which are peatification and coalification to form a coal seam $^{(13)}$. The identification of coal macerals and development of the coal maceral concept drove from the industrial use are established by the International Commission for Coal Petrology (ICCP). The microscopical determination method is in accordance with the standard test method^(14,15). The macerals are classified into three groups which are Vitrinite, Liptinite, and Inertinite. Coal samples are crushed to pass an 850 µm sieve mixed with a binder and formed into a briquette. The briquette is polished to a flat and scratch-free surface and observed with a microscopic system using oil immersion objective. According to the distinctive reflection, optical properties and morphology of different maceral groups, the proportions of individual groups are determined.

Due to vitrinite being the most abundant of the coal maceral groups, the degree of metamorphism in the natural geological process from lignite to anthracite are determined by vitrinite reflectance (Ro) to show the rank of the coal (Table 1).

2.2.3 15 lb coke oven test

The coking test was undertaken with a furnace, 112 mm wide, 305 mm high and 350 mm long, which heated electrically from both sidewalls. The charged coal was heated from 800° C to 900° C at the center and subsequently held for 15 min soaking time. The coal moisture was 6% and the weight was 7 kg. The coke after carbonization was quenched with water directly.

The determination of mechanical strength of this lump of coke was done with the ISO Micum drum and followed the ISO 556 standard⁽¹⁶⁾. The percentage of coke remaining on 30-mm-round aperture sieves after Micum test (m_{30}) was list in Table 1 to show the stability after carbonization. ISO 18894 and ASTM D5341 standard, patterned after the Nippon Steel test procedure, was adopted to measure indirectly the chemically reaction in the blast furnace⁽¹⁷⁾. Coke lumps are reacted with CO₂ and rubbed together during descending in the blast furnace, therefore two indices, Coke Reactivity Index (CRI) and Coke Strength after Reaction (CSR), are designed. CSR and m_{30} were used as the main coke strength data to review the coal quality from different region.

2.3 Coke analysis

2.3.1 Microtexture analysis

The true solids of coke are derived from coal carbonization, which include both a Reactive Maceral Derived Component (RMDC) and an Inert Maceral Derived Component (IMDC)⁽¹⁾. RMDC is classified according to the grain size of the mosaic textures and IMDC is originated in the Inertinite particles of the parent coal. ASTM D5061 standard was used as the principal test method to determine the types and amounts of coke carbon forms and the associated coal and process derived textural components in the coke⁽¹⁸⁾. The volume percent of the textural components in coke is useful to characterize the optical properties of coke and help to determine the coal blend proportions.

2.3.2 Microstructure analysis

The structure of coke pore wall and the properties of the pores are strongly suggested to govern the coke strength. Several studies have verified that the coke wall thickness, pore distribution, pore size and shape factor were characterized by image analysis. CSIRO Process Science and Engineering has developed the Mineral 4 / Recognition 4 software system for automated imaging analysis and textural identification⁽¹⁹⁾. The reflectivity of high resolution coke images were used to determine coke walls, pores and calculate morphological parameters. It is believed that microstructure provides an essential understanding of coke strength and changes under blast furnace conditions.

3. RESULTS AND DISCUSSION

3.1 Coal characterization

3.1.1 Chemical analysis

In order to sort out the coal characteristics from different regions, the analytic results were shown in a ternary plot and atom ratio plot (Van Krevelen Diagram) respectively (Fig.2). Most of Indonesian coal deposits are with high volatile matter due to being geologically young. Only a few coal mines are affected by igneous activity and produce high rank metallurgical coal. This property is also found in the hydrogen/ carbon and oxygen/carbon ratio, which is obviously why Indonesian coals exhibited a higher hydrogen/carbon ratio than the others.

3.1.2 Ash composition

Some mineral matters in coal are like catalysts as the coke descends to lower regions of the blast furnace. The alkalis and iron in coal affect fluidity and inhibit growth of crystallites when creating functional groups and consequently act as a catalyst of gasification by CO_2 ⁽¹⁶⁾. Since SiO₂ and Al₂O₃ are the most abundant composition in coal ash, these two components are taken as the basic elements compared with alkalis composition and Fe_2O_3 . Higher Fe_2O_3 was found in Indonesian coal (Fig.3); however there are also Australian and American coals holding the same ratio. While Na₂O, K₂O, MgO and CaO were considered entirely, Indonesian coals were distinguished from the others. This chemical property is one factor to cause the weak coke strength after reaction.

3.1.3 Maceral composition

It is known that vitrinite, liptinite, and part of semifusinite are the reactive coal components during carbonization. Based on the maceral composition analysis, Indonesian coals contain more than 85% vitrinite which is able to soften on heating and bind inerts (Fig.4). Making a good coke requires inert material in proper proportion because of avoiding crack growth. High volatile matter which suggests high gasification proportion and porous structure along with high vitrinite content lead to weak coke strength. Therefore, purified vitrinite is likely to be a nice condiment rather than a major material within coal blend assessments.

3.2 Coke characterization

3.2.1 Microtexture

The properties of individual coke microtextures were measured using LEICA DM4500 polarized light microscope with a 500x oil immersion objective with



Fig.2. Proximate and Ultimate analytic results of metallurgical coals from different regions. (a) Proximate analysis results shown in ternary plot (b) Ultimate analysis results shown in the atom ratio plot.



Fig.3. Ash composition results of metallurgical coals from different regions. (a) Comparison with Fe_2O_3 , (b) Comparison with Alkali (Alkali (%) = $K_2O(\%) + Na_2O(\%) + MgO(\%) + CaO(\%) + Fe_2O_3(\%)$).

the polar crossed at 105°. RMDC is corresponding to the binder phase in ASTM D5061-05 standard. Volume percentage of the various types of binder phase carbon forms were counted with at least 1000 binder phase points and carbon forms were determined and contrasted with the parent coal V-Type (Vitrinite Type; R₀×10) which is shown as Corresponding V-Type in Fig.5. The distributions of Corresponding V-Type from coke were wider than their parent coal V-Type which was owing to the coking process. This behavior is the main advantage of metallurgical coal to blend with each other. The positive correlation between the V-Type of coal and the Corresponding V-Type of coke infers that the microtexture of coke is able to restrict from coal blend, and therefore the weak coke strength of Indonesian coal is not bound up with the microtexture of coke.



Fig.4. Maceral compositions of metallurgical coals from different regions.



Besides the binder phase carbon forms were measured, the filler phase which is corresponding to IMDC was counted. Seldom IMDC was founded in Indonesian coals which is shown in Fig.6. The connected boundary between IMDC and RMDC was observed form cokes from the other regions. Those observations were responded to the maceral composition results.

3.2.2 Microstructure

Coke microstructure features included pores and coke walls. The coke strength was related to coke wall thickness, coke microtexture, pore size, pore density, pore shape and IMDC. The size of IMDC interfered with generating cracks, and the low roundness pores is capable to connect pores after reaction. The observations of four coke images from Indonesian coals were shown in Fig.7. The coke walls were thin and the pores were small and widely spread which was the perfect gas pathway in the blast furnace. The weak strength of cokes from Indonesian coals were clarified from microstructure properties.

4. CONCLUSIONS

Indonesian metallurgical coals were produced later than Australian & American coals. Acquainting with the exercise of Indonesian coal is essential since the shipping time is shorter than the other regions. The fundamental characteristics are summarized below:

1. Higher alkali oxide proportion in ash was found in most Indonesian coals, and that acts like a catalyst as the coke descends to lower regions of the blast furnace.



Fig.5. Distribution of V-Type and Corresponding V-Type from different regions. (a) V-Type of coals (b) Corresponding V-Type of cokes.



Fig.6. Polarized reflected light microscope images (a) AU-1 (b) AU-3 (c) IN-2 (d) IN-5 (e) AM-1 (f) AM-2 (g) AF-1 and (h) AF-2.

- 2. All of the Indonesian coals have more than 85% of vitrinite content which was softened and flowed during carbonization. Lots of small pores and thin coke walls were created and originated from high vitrinite content. The microstructure observation also supports the outcome of high RMDC content.
- 3. The Corresponding V-Type of coke was inferred

from the microtexture which was based on types of binder phase carbon forms. The correlation between V-Type of coal and the Corresponding V-Type of coke is positive and restrict from the coal blend, hence the microtexture of coke is able to be predicted from that of the V-Type of coal blend.



Fig.7. Large scale microscope images of cokes from Indonesian coals (a) IN-4 (b) IN-3 (c) IN-5 and (d) IN-6.

4. The microstructure of coke in the formation of coke walls and pores are believed to have significant impact on coke strength. Small pores which provide gas pathways and react with CO₂ were deduced from the microscope analysis, and therefore the microstructure of Indonesian coal is unquestionably of a weak coke strength.

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