Oil Shale, 2015, Vol. 32, No. 3, pp. 204–217
doi: 10.3176/oil.2015.3.02
ISSN 0208-189X

THE CHARACTERISTICS OF COAL AND OIL SHALE IN THE COASTAL SEA AREAS OF HUANGXIAN COALFIELD, EASTERN CHINA

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Abstract: *Coal and oil shale quality is important to evaluate for mining purposes worldwide. In order to evaluate the quality and analyze the mineralization of coal and oil shale in the Paleogene Lijiaya Formation in Huangxian Coalfield, Eastern China, 27 samples were studied by the methods of geochemistry, sedimentology and coal geology. The results indicated that coal belonged to brown coal with high moisture content, low-medium ash yield and low sulfur content. It is high-quality coal that is suitable for industrial electricity generation. However, the quality of oil shale varied by layers, while three quality levels could be distinguished: high-quality oil shale was located in layers A-1 and B with a thickness of 2.5 m, medium-quality oil shale was deposited in layer A-2 with a thickness of 1.71 m, and poor-quality oil shale was found in layers A-3 and C with a thickness of 1.55 m. So, oil shale layer A can be mined together with coal bed 1, while oil shale layer B can be extracted separately and layer C as a perspective resource would be subject to mining in future.*

Keywords: *Huangxian Coalfield, coal quality, oil shale, Eastern China.*

1. Introduction

Coal bed and oil shale have been mined worldwide for a long time. Coal and oil shale can be used as fuel to generate electricity, whereas oil shale can also be utilized for extracting oil [1, 2]. However, the use of coal and oil

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shale brings several kinds of environmental hazards and health risks [3]. In addition, the economic value of oil shale in the same bed varies vertically, which results in too high costs [3]. Some of these problems could be minimized, or even avoided if coal and oil shale quality information were available for decision makers.

There are many symbiotic coal and oil shale basins in the world, such as the Huangxian [4–9], Songjiang [10] and Heshentuoluogai [11] basins of China, the Intermontane Basin of USA [12], the Junggar Basin of China [13], the US Green River Basin [14], etc. Coal bed mined together with oil shale without prior quality evaluation induces too high costs in coal separation, oil shale refining, etc. [15–20]. So, the quality of coal and oil shale deposited in the same basin must be evaluated before mining. However, there are but few publications available on this topic at present. Although dealt with by several researchers before [21–25], it is very significant to further study the coal and oil shale succession in the same basin.

In this paper, the quality evaluation of coal and oil shale in the coastal sea areas of Huangxian Basin is presented. The quality analysis of coal mainly provides information on ash yield, contents of moisture, volatile matter, minerals, C, H, N and S, ash composition, calorific value, etc. The oil shale quality analysis mainly presents data on tar yield, calorific value, ash yield and elemental composition. Quality evaluation also gives additional information about the coal and oil shale forming environment and their quality evaluation system researched earlier [15–25].

2. Geological setting and previous study

Huangxian Coalfield is located in the east area of North China (Figs. 1a, 1b) [4–9]. There are eight land exploration areas, namely Beizhao, Haidai, Liangjia, Liuhai, Sangyuan, Wali, Xiangcheng and Yankou areas (Fig. 1c) [4–9]. The Zhubidian, Lijiaya and Xiaolou formations of the basin are mainly made up of Paleogene strata (Fig. 2). Coal and oil shale of the Lijiaya Formation have been mined for the last fifty years in land mining areas, while the underwater areas have been started to develop only recently. With the development of land mining exploration, underwater areas are becoming target regions with resource to be mined in future. While there are many researches on the coal and oil shale deposited in the land areas of Huangxian Coalfield, mostly dealing with their geochemistry [4, 5], sedimentary environment [6–9], etc., papers about the quality of coal and oil shale found in the field's underwater areas are few. The respective studies are yet highly important from the practical point of view.

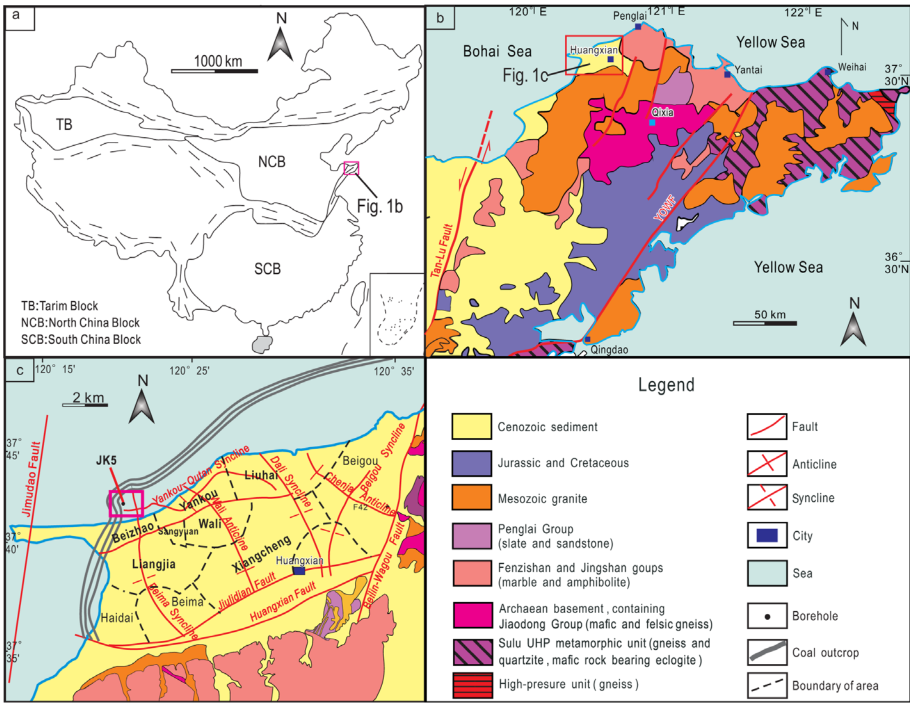

Fig. 1. The location of Huangxian Coalfield.

3. Sampling

For analysis 27 samples were collected from well JK5 located in the underwater area (the well location is shown in Fig. 1c), of which 19 samples were oil shale, two were kerogenic argillite and six samples were coal ones (Fig. 3). From top to bottom, the samples of the section were numbered from JK5-1a–f to JK5-23, and tested following the Chinese Standard Method GB482-2008. The distance between the oil shale sampling points was 20 centimeters. The samples were cut over an area of 10 cm in width and of predetermined depth (1 m for each sample) [26].

4. Methods

The bulk samples were air-dried, milled and split until a representative 0.5 kg sample milled to pass through a 0.250 mm sieve was obtained for mineralogical, proximate, ultimate and chemical analyses. Other splits were made at different size fractions (< 0.20 mm) for petrological studies. Proximate and ultimate analyses were performed following Handbook of Coal Analysis standard procedures [27]. The instruments used were Mettler AE-100 Digital Scale (USA), and DHG-9140A electricity heat drum wind drying oven, GW-100 muffle furnace, CS-S Coulomb sulfur meter, SDACM-5000 Calorimeter and BCH-1 semi-automatic hydrocarbon meter (all China). The results of petrographic and chemical analyses are given in Tables 1–6.

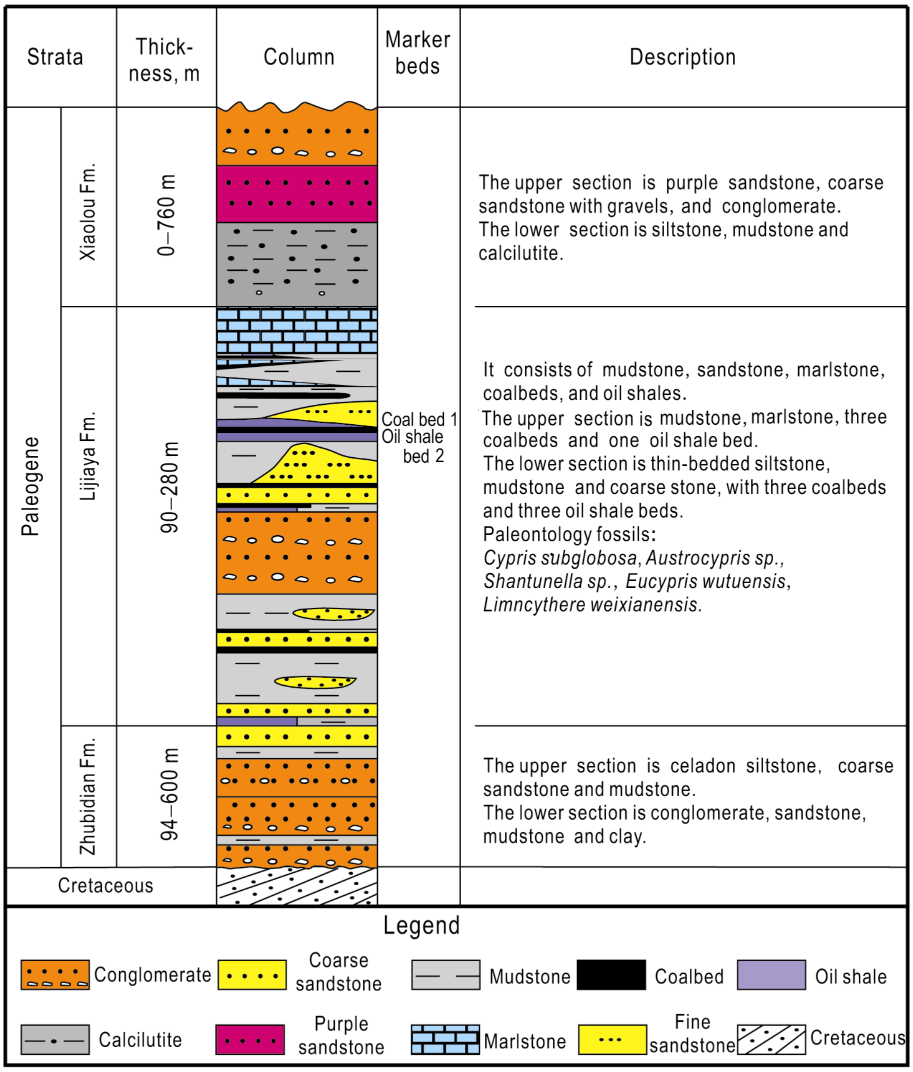

Fig. 2. The strata column of Huangxian Coalfield.

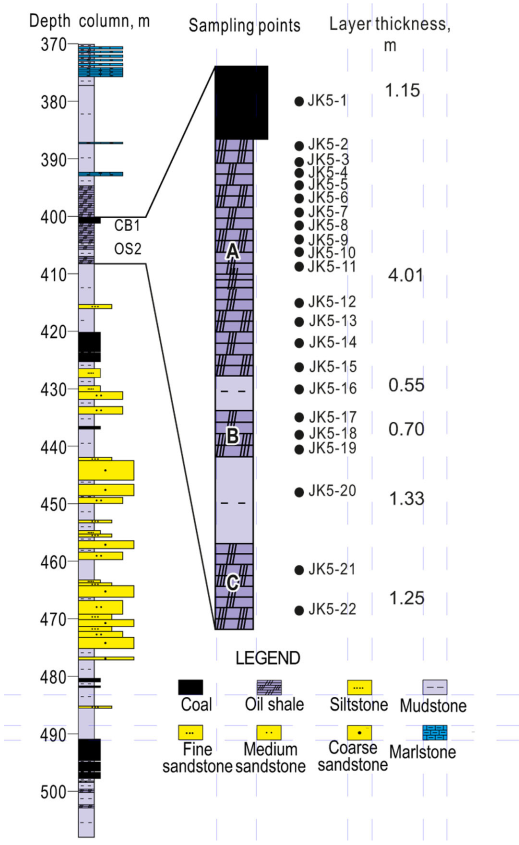

Fig. 3. Sampling points in well JK5.

5. Results

Table 1 presents the analytical results for coal samples, their ash yield and composition are shown in Table 2, the indexes for evaluation of coal quality are given in Table 3. The indexes for oil shale quality evaluation are shown in Table 4, the analytical results for oil shale and kerogenic argillite samples are presented in Table 5, their ash yield and composition are given in Table 6.

5.1. Coal sample

5.1.1. Moisture content

Moisture content is a basic parameter of the quality of coal to assess its economic value. The higher the moisture content in coal, the poorer its quality. According to Table 1, the moisture content in coal samples ranges from 7.52 to 20.89%.

Table 1. Analytical results for coal samples

Sample No/Index	Mad, %	Ad, %	Vdaf, %	$Q_{b,ad}$, MJ/kg	St,d, %	Cdaf, %	Hdaf, %	Ndaf, %
JK5-1a	20.89	17.06	50.04	28.98	0.68	71.68	4.79	2.11
JK5-1b	18.92	12.60	48.97	30.86	1.00	75.92	5.82	0.73
JK5-1c	21.72	21.38	51.67	29.30	0.57	76.11	7.54	1.50
JK5-1d	12.88	37.27	52.48	34.90	1.24	80.12	6.21	2.16
JK5-1e	7.52	29.33	41.66	32.86	1.15	73.55	5.21	1.32
JK5-1f	13.96	42.14	52.19	30.60	0.95	70.76	7.22	1.85

Mad – moisture (air-dried); Ad – ash yield (dry); Vdaf – volatile matter (daf); St,d – total sulfur (dry basis); Qb,ad – calorific value (air-dried); Cdaf – carbon content (daf); Hdaf – hydrogen content (daf); Ndaf – nitrogen content (daf).

5.1.2. Ash yield and composition

Ash is the residue of complete combustion of a substance under pre-determined conditions. In coal studies, ash yield and composition play an important role and act as indicators of its quality [3, 27]. The ash yield of the studied coal samples is from 12.60 to 42.14% (Table 1). In ashes, crystalline matters are formed through a series of physico-chemical processes such as fusion or a partial melting of discrete mineral matter, or coalescence of melted mineral in coal burning processes. The coal ashes consist mainly of SiO_2, Al_2O_3, CaO, Fe_2O_3 and SO_3, and small proportions of MgO, TiO_2, etc. (Table 3). According to chemical composition, the ashes belong to the $CaO–Fe_2O_3–Al_2O_3–SiO_2$ system (Table 2). The chemical composition usually changes much due to the different mineral components contained in the coals. The results of analysis showed that the content of the main minerals in coal was proportional to the chemical and mineral composition of ashes.

Table 2. Ash yield and composition of selected coal samples, Ad, %

Sample No/Constituent	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	TiO_2	K_2O	Na_2O	P_2O_5	MnO_2
JK5-1c	10.63	3.45	0.69	0.54	0.18	0.35	0.15	0.18	0.32	0.05	0.01
JK5-1d	10.50	3.55	0.71	0.56	0.23	0.40	0.20	0.23	0.27	0.10	0.02

Table 3. Coal evaluation indexes [18]

Index/Sample	Lignite	Low rank bituminous coal	High rank bituminous coal	Anthracite
Mad, %	28–5	5–11	1	1–2
Vdaf, %	63–46	24–46	10–24	2–10
C, %	60–75	75–87	87–91	91–96
H, %	6–7	5.6–6	4–5.5	1–4
$Q_{gr,d}$	16.7–29.3	29.3–36.17	≥ 36.17	≤ 36.17
Ad, %	< 15	15–25		> 25

The ash yield of coal is a key parameter to evaluate its quality. Coal ash is the residue that is derived from the inorganic and organic matter of coal during combustion. The yield and geochemical characteristics of ash are dependent on the conditions of coal formation and the temperature and mode of combustion [3, 16]. The chemical composition of coal ashes varies widely depending on the mineral and organic constituents present in the coal of a particular area [3, 16]. During coal combustion most non-volatile trace and minor elements form the matrix of ash as a homogeneous melt and crystals, which undergo a series of physico-chemical processes.

5.1.3. Volatile matter

According to Table 1, the volatile matter content in coal samples is between 41.66 and 52.48%. The volatile matter of coal has a close relationship with its metamorphic degree.

5.1.4. Sulfur

The forms of sulfur in coal are mainly pyretic sulfur, organic sulfur and sulfate sulfur. Pyretic and organic sulfur generally account for the bulk of sulfur in coal. Elemental sulfur is also present in coal in trace to minor amounts, and is usually not determined in routine coal analysis [3]. During combustion, almost all the organic sulfur, elemental sulfur, most of the pyretic sulfur and part of the sulfate sulfur in coal are emitted into the atmosphere as aerosols and gaseous products of flue gas emissions. But pyretic sulfur and sulfur from gypsum can during combustion form anhydrite ($CaSO_4$) with CaO from calcite, while Fe from pyrite forms with oxygen hematite. Bassanite may also be formed from gypsum. Sox emissions may have a noxious effect on air, water and living organisms, including humans. Several authors suggest that minor elements associated with the organic and sulfide fraction also volatilize when coal is burning [3]. The sulfur content of coal samples ranges from 0.57 to 1.24%, the total sulfur content being less than 1.5% (Table 1).

5.1.5. Calorific value

The calorific value of coal is the key factor to assess its economic usefulness [1–3]. The dry basis high calorific value is used to study and evaluate the quality of coal. According to Table 1, the calorific value of coal samples is 28.98–34.90 MJ/kg.

5.1.6. Organic element composition

5.1.6.1. Organic carbon

Organic carbon is an important constituent of organic matter in coal. The organic carbon content of coal increases gradually with increasing coalification degree. From Table 1 it can be seen that the content of organic carbon in coal samples is between 70.76 and 80.12%.

5.1.6.2. Organic hydrogen

Organic hydrogen is another important component of organic matter in coal. The organic hydrogen content of coal reduces with increasing coalification degree. According to Table 1, the content of organic hydrogen in coal samples is from 4.79 to 7.54%.

5.1.6.3. Organic nitrogen

Organic nitrogen is the only element existing in coal in organic state. The organic nitrogen content of coal decreases with increasing coalification degree at greater depths. According to Table 1, the nitrogen content in coal samples ranges from 0.73 to 2.16%.

5.2. Oil shale and kerogenic argillite

5.2.1. Tar yield

The tar yield of oil shale is determined by the standard Fischer-Schrader assay. According to Table 5, the tar yield of oil shale and kerogenic argillite samples ranges from 5.0 to 33.2%, with a minimum value for the kerogenic argillite sample JK5-16.

5.2.2. Calorific value

Calorific value referred to as heating value during the combustion of a unit weight of oil shale is an important parameter of oil shale quality [8]. As can be seen from Table 5, the calorific value of oil shale and kerogenic argillite samples varies from 2.86 to 19.67 MJ/kg.

5.2.3. Ash yield and composition

Oil shale ash is the residue deriving from its inorganic and organic matter during combustion.The yield and composition of ash are important parameters of oil shale quality. The lower the values of these parameters are, the higher the oil content of oil shale is and the better its quality is. The chemical composition of ashes varies widely due to the mineral and organic matter present in the oil shale of an area. The formation of ash particles during oil shale combustion greatly influences the heat and mass transfers in an oil shale-fired boiler, and the particles emitted from combustion into the atmosphere represent a hazard to human health and the environment [6–23].

In ashes, crystalline substances are formed through a series of physicochemical processes such as fusion or a partial melting of discrete mineral matter, and coalescence of melted mineral in oil shale burning processes [8]. Analysis of the investigated oil shale and kerogenic argillite samples showed their ash yield to range from 31.84 to 80.10%, with a maximum value for the kerogenic argillite sample JK5-16. The ash yield of oil shale is usually lower than that of kerogenic argillite (Table 5). The oil shale ashes consist mainly of SiO_2, Al_2O_3, CaO, MgO, Fe_2O_3 and SO_3, and minor proportions of TiO_2,

K_2O, Na_2O, etc. SiO_2, Al_2O_3, CaO and Fe_2O_3 make up 42.67–58.84% of the total ash.

5.2.4. Sulfur

The major forms of sulfur in oil shale are organic and inorganic sulfur [3]. During combustion, both organic and inorganic sulfur are almost completely emitted into the atmosphere as aerosols and gaseous products of flue gas emissions. Sox emissions may have a noxious effect on air, water and living organisms, including humans. Several authors suggest that minor elements contained in organic sulfur and the sulfide fraction also volatilize when oil shale is burning [1–8]. The sulfur content of oil shale samples ranges from 0.44 to 1.55%, that of samples JK-5 and JK-14 being more than 1.5%, which means medium-sulfur oil shale, while other samples are low-sulfur.

6. Discussions

6.1. Coal quality

Yang and Pan have proposed a table to classify coal and determine its rank (Table 3) [18]. According to Table 1 and Table 2, the moisture content of coal samples is 5–28%, their ash yield varies from 12 to 43%, the sulfur content is between 0.5 and 1.5%, the calorific value is from 29.3 to 36.17 MJ/kg, and the volatile matter content ranges from 63 to 46%. The content of organic carbon of most coal samples is 60–75%, the organic hydrogen content varies from 6 to 7%. The above values allow coal to be classified as low-sulfur lignite. The coal can be used to generate electricity and as fuel. Considering the favourable characteristics of humic acid present, the high coking quality and high volatile matter content, coal is suitable for industrial applications.

6.2. Oil shale quality

According to many scientists, oil shale with the tar yield on dry ash basis more than 5% has an ash content of 40–83% and ash yield of 40–65%, and its tar yield on dry air basis is higher than 8% [23–26]. Oil shale is a high-organic combustible sedimentary rock with an oil content more than 3.5% and calorific value of 4.18 MJ/kg, which can release shale oil by low-temperature carbonization [28, 29].

Tar yield, ash yield, calorific value and total sulfur content are the four main parameters to evaluate oil shale quality (Table 4). There are three layers of oil shale in oil shale bed 2, analytical results for oil shale samples are presented in Table 5. There are eight samples with low ash yield, of which seven are from layer A (layer A can be further subdivided into layer A-1, layer A-2 and layer A-3). The ash yield of samples increases from layer A to layer C. The tar yield of most samples is high, however, some are of medium tar yield. From layer A to layer C, the tar yield of samples decreases. The sulfur content of the 14 samples from the lower part of layers A, B and C is low, decreasing from layer A to layer C. The calorific value of samples decreases from layer A to layer C. It is significant to explore oil shale of lower ash yield, higher tar yield, lower total sulfur content and higher calorific value [25]. On the basis of these parameters, three oil shale quality levels can be distinguished in the studied layers. High-quality oil shale is deposited in layers A-1 and B with a thickness of 2.5 m, medium-quality oil shale lies in layer A-2 with a thickness of 1.71 m, and poor-quality oil shale is found in layers A-3 and C with a thickness of 1.55 m. Oil shale layer A can be mined together with coal, oil shale layer B can be mined separately (Fig. 4), and layer C can be treated as a perspective resource for future.

Table 4. The evaluation principles of oil shale (according to [24], modified)

Index/Quality level	Low	Medium	High
Tar, %	3.5–5	5–10	> 10
Ad, %	< 65	65–83	83–88
St,d, %	1.0–1.5	1.5–2.5	> 2.5

Table 5. Analytical results for oil shale and kerogenic argillite samples

Layer	Sample No	Sample type	Tard, %	Qb,ad, MJ/kg	Ad, %	St,d, %
A	JK5-2	Oil shale	18.8	14.38	45.01	1.25
	JK5-3	Oil shale	16.2	14.27	42.42	1.08
	JK5-4	Oil shale	21.1	12.64	50.57	1.07
	JK5-5	Oil shale	23.3	13.84	43.88	0.73
	JK5-6	Oil shale	33.2	19.67	31.84	1.04
	JK5-7	Oil shale	14.1	9.07	56.19	1.54
	JK5-8	Oil shale	8.9	7.83	58.46	0.86
	JK5-9	Oil shale	12.1	7.41	66.50	0.98
	JK5-10	Oil shale	13.0	7.90	66.58	0.75
	JK5-11	Oil shale	13.0	7.15	66.74	0.85
	JK5-12	Oil shale	11.0	6.67	70.66	0.99
	JK5-13	Oil shale	11.7	6.83	67.46	1.08
	JK5-14	Oil shale	7.9	5.25	74.43	1.55
	JK5-15	Oil shale	9.8	5.74	71.47	0.78
	JK5-16	Kerogenic argillite	5.0	2.86	80.10	0.57
B	JK5-17	Oil shale	16.6	8.99	65.41	0.77
	JK5-18	Oil shale	17.7	10.21	62.23	0.96
	JK5-19	Oil shale	14.7	8.07	65.92	0.94
	JK5-20	Kerogenic argillite	5.5	2.98	76.85	0.44
C	JK5-21	Oil shale	9.6	4.53	76.52	0.77
	JK5-22	Oil shale	8.1	3.88	76.05	0.79

Tard – tar yield (dry basis); Ad – ash yield (dry); St,d – total sulfur (dry basis); Qb,ad – calorific value (air-dried).

Table 6. Ash yield and composition of selected oil shale samples, Ad, %

Sample No/Constituent	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	TiO_2	K_2O	Na_2O	P_2O_5	MnO_2
JK5-3	20.98	3.85	4.73	13.11	1.07	2.85	0.10	0.34	0.49	0.40	0.202
JK5-10	38.37	2.84	4.53	8.93	1.12	2.18	0.22	0.49	0.75	0.32	0.224
JK5-18	44.56	1.06	4.78	8.44	1.38	2.00	0.34	0.90	0.97	0.28	0.144
JK5-23	36.05	2.40	4.69	9.89	1.21	2.29	0.24	0.61	0.77	0.33	0.185

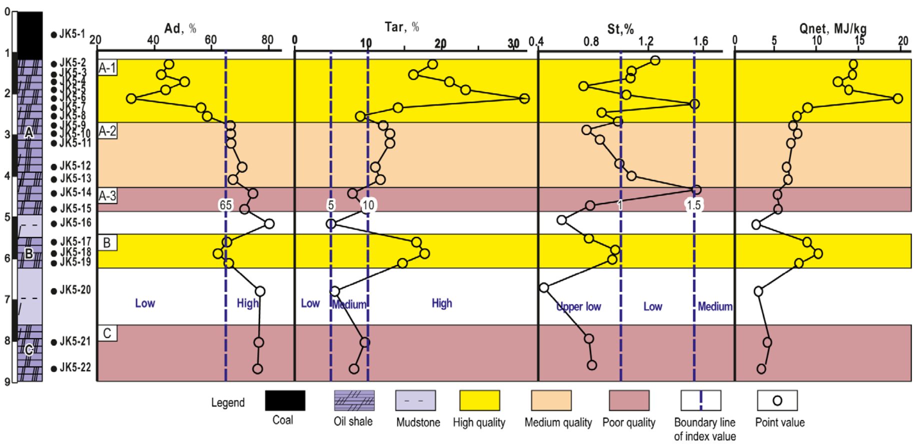

Fig. 4. Diagram for oil shale quality evaluation.

7. Conclusions

Coal and oil shale of the coastal sea areas in Huangxian Coalfield were studied using methods of geochemistry, sedimentology and coal geology. The conclusions drawn are as follows:

(1) The quality of coal was evaluated. The moisture content of coal ranges from 7.52 to 20.89%. The ash yield is 12.60–42.14%, the volatile matter content in coal is 41.66–52.48%, the sulfur content ranges from 0.57 to 1.24%. The calorific value is 28.98–34.90 MJ/kg. The content of organic carbon in coal is from 70.76 to 80.12%, the content of organic hydrogen is between 4.79 and 7.54%, the content of organic nitrogen ranges from 0.73 to 2.16%. According to the above data, the coal is regarded as lignite with low sulfur content, and is suitable to be utilized for industrial electricity generation and as fuel.

(2) The quality of oil shale and kerogenic argillite was evaluated by four indexes. The tar yield of oil shale and kerogenic argillite is 5.5–33.2%, the calorific value is between 2.98 and 19.67 MJ/kg, the ash yield ranges from 31.84 to 80.10%. The sulfur content of samples JK-5 and JK-14 is more than 1.5%. On the basis of the above indexes, three oil shale quality levels can be distinguished. High-quality oil shale is in layers A-1 and B with a thickness

of 2.5 m, medium-quality oil shale lies in layer A-2 with a thickness of 1.71 m, poor-quality oil shale is deposited in layers A-3 and C with a thickness of 1.55 m. Oil shale layer A can be mined with coal bed 1, oil shale layer B can be mined individually, and layer C can be treated as a perspective resource for future.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (41272172, 41202070), the Research Fund for the Doctoral Program of Higher Education of China (20123718110004), the Natural Science Foundation of Shandong Province (ZR2015JL016), and the Shandong Provincial Key Laboratory of Depositional Mineralization & Sedimentary Mineral (DMSM201401). We thank Professor Linhua Sun and Dr. A. K. Verma for providing many constructive comments.

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Presented by E. Reinsalu and J. Qian
Received December 14, 2014