PETROGRAPHIC CHARACTERISTICS AND PETROLEUM POTENTIAL OF OIL SHALE FROM NA HONG BASIN, CHIANG MAI, THAILAND

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Abstract

The Na Hong basin is a small basin in the west of Chiang Mai Province, northern Thailand. Twenty-one oil shale samples were collected from Huai Phak La in the Na Hong basin. The aims of this study are petrologically to analyze the organic matter of the oil shale and to determine the oil source potential using geochemistry techniques, including proximate analysis, ultimate analysis, calorific value, and total organic carbon (TOC) analysis. The results of this study show that the organic matter of the oil shale is dominated by the liptinite group which ranges from 10.00–66.50%. The liptinite is composed mainly of lamalginite and liptodetrinite, with minor amounts of telalginite, sporinite, exsudatinite, and resinite. The vitrinite group which ranges from 4.00–40.00% is composed mainly of gelovitrinite, and the inertinite group has a range from 0.00–3.00% of fusinite. Mineral matter ranges from 21.00–80.50% and is composed mainly of clay minerals and quartz. Proximate analysis shows that the moisture content, ash content, volatile matter and fixed carbon content range from 3.26–5.90%, 53.70–78.98%, 15.83–34.52%, and 1.93–21.45%, respectively. Ultimate analysis shows that the carbon content, hydrogen content, sulphur content, and oxygen content range from 10.23–25.61%, 1.81–3.95%, 0.32–8.15%, 7.72–12.58%, respectively and the nitrogen content is mostly less than 1 %. The calorific values range from 821-3025 kcal/kg and the TOC content ranges from 4.16–22.31 wt%. Oil shale from this basin is the lamosite type. Apparently, the very dark gray and tough sample (sampling number PL1-23) shows a grade with high carbon, hydrogen, and calorific value. The study results indicate that the oil shales in the Na Hong basin show a trend to be good source rocks of Type II/III kerogen.

Keywords: Oil shale, organic petrology, maceral, petroleum source rock

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Introduction

Oil shale is a fine-grained, organic-rich sedimentary rock that can produce a substantial amount of oil and combustible gas upon pyrolysis. The organic components in oil shale and coal can be classified into 3 maceral groups: liptinite, vitrinite, and inertinite (Stach *et al*., 1982). The oil shales were deposited in a wide variety of depositional environments including freshwater to saline ponds and lakes, epicontinental marine basins, subtidal shelves, and in limnic and coastal swamps and very commonly with coal that was deposited there. These give a wide range of organic and mineral compositions and lead to different oil shale types (Hutton, 1987).

Most of the tertiary basins in northern Thailand have been intensively explored for coal and petroleum resources and this has led to production such as in the Fang Oil Field and Sirikit Oil Field. Ratanasthien (1997) studied algae types in oil source rocks in northern Thailand and reported that they are dominated by alginite B or lamalginite and that it is usual to find small amounts of alginite A or telaginite disseminated throughout the oil shale beds. This alginite is the most important maceral which can be a good indication of oil sources depending on its abundance, as well as other liptinite macerals which also contribute hydrocarbon.

The aims of this study are to analyse the oil shale which is associated with the coal-bearing formation in the Na Hong basin using organic petrology and geochemistry evidence, and to study the environment of the depositions.

Geology of the Na Hong Basin

The Na Hong basin is about 30 kilometers north of Mae Chaem District, Chiang Mai Province (between longitude 98°21′ and 98°42′ east and between latitude 18°42′ and 18°43′ north) (Figure 1). It is identified as a fault-bounded basin and the trend of the faults is mainly NE-SW and nearly N-S (Songtham *et al.,* 2000). The basin floor lies between 700-800 meters above mean sea level, surrounded by granite and Palaeozoic sedimentary rocks of the Late Silurian to Devonian and Carboniferous periods. The basin is bounded by Devonian-Carboniferous sedimentary rocks except in the northwest part where it is bounded by Upper Carboniferous conglomerate, sandstone, and shale. The east and south of the basin show Silurian to Devonian rocks. They are shale, sandstone, graywacke and chert with occasional limestone intercalations. The west and northwest side of the basin display an Upper Carboniferous clastic rock sequence. The tertiary sedimentary strata in the basin consist of conglomerate, gravel, sandstone,

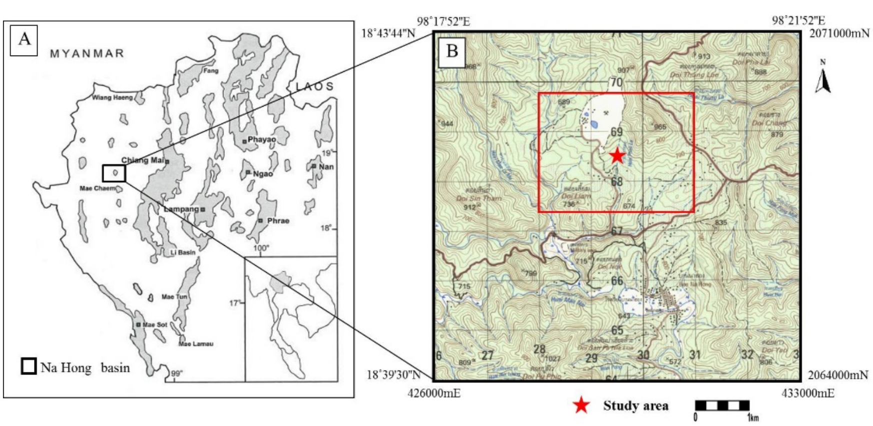

Figure 1. (A) Location map of tertiary basins in northern Thailand and Na Hong basin is in the box (modified from Ratanasthien, 2011).
(B) Study area in Mae Chaem District, Chiang Mai Province (modified from topographic map of the Royal Thai Survey Department, Map sheet 4646 II, Scale 1:50,000, 1999).

sand, shale, oil shale, and coal. Sedimentary strata is classified into 2 units: unit A is dominated by fine-grained sediments consisting of oil shale, shale, and coal, with thin layers of sand and sandstone intercalated: unit B is dominated by coarse-grained sediments including sandstone, conglomeratic sandstone, and conglomerate in the uppermost part with thin coal seams intercalated (Songtham, 2003).

Materials and Methods

Twenty-one oil shale samples from Huai Phak La in the Na Hong basin were systematically collected from a vertical succession of layers using the channel sampling method (Figure 2). They were dried, crushed, divided, and ground prior to chemical analysis. The solidified samples were cut both perpendicularly and parallel to the bedding for petrographic study purposes.

Petrographic Analysis

Organic petrography is the study of the organic matter in sedimentary rocks including coal, petroleum source rock, and oil shale by using a reflected polarizing microscope with UV-excitation to classify organic components or macerals. The samples were analysed with a Zeiss incident light microscope with polarizer at the Department of Geological Sciences, Faculty of Science, Chiang Mai University. A total of 400 points were counted, identified, and calculated for the percentage. The classification and characteristics of the oil shale in this study are based on Stach *et al.* (1982), Hutton (1987), and Hower *et al.* (1997).

Proximate Analysis, Ultimate Analysis, Calorific Value, and Total Organic Carbon Analysis

Each of the air-dried, fresh samples was crushed and split into 2 parts and 1 part was ground and passed through a 250 μm sized sieve. The other part was kept as a reference. Seven mixed analytical samples were obtained by mixing into equal portions according to the similar physical properties and successive layers (Table 2). They were subjected to the following analyses.

The proximate analysis is used to define the weight percentages of the moisture,

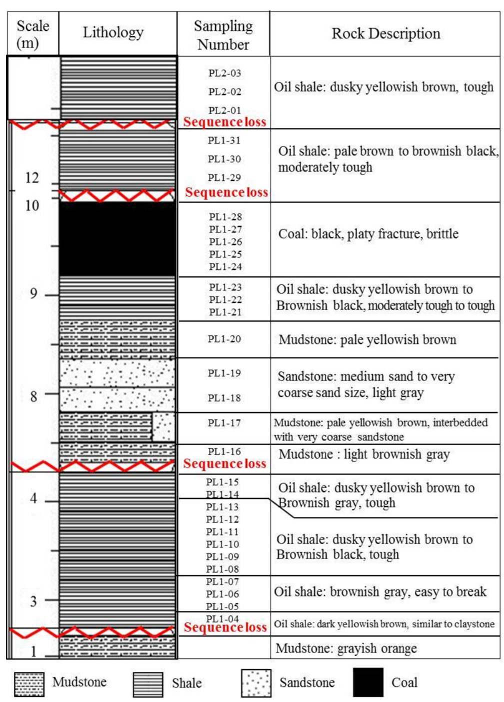

Figure 2. The sampling numbers and sample descriptions from Huai Phak La in the Na Hong basin

volatile matter, ash, and fixed carbon content. The samples were analyzed with a thermogravimetric analyzer (TGA701, LECO Corp., Saint Joseph, MI, USA) (ASTM, 2011). The ultimate analysis is used to determine the percentage that is present of the C, H, N, S, and O elements in a sample using a CHN determinator (TRuSpec CHN, LECO Corp.) which here was used for the carbon, hydrogen, and nitrogen contents, and an S determinator (SC632, LECO Corp.) for the sulphur content (ASTM, 2012). Oxygen is obtained by subtraction of the elementary percentage and the ash content from 100%. The calorific value or heating value is used to determine the energy released as thermal energy in a bomb colorimeter (AC600, LECO Corp.) (ASTM, 2011). The proximate analysis, ultimate analysis, and calorific value were applied to all these samples at the Laboratory Section, Geology Department, Mae Moh Mine, Lampang, Thailand. The total organic carbon (TOC) analysis was used to indicate the amount of carbon as an organic compound. The samples were analysed with a TOC analyzer for the solid sample (TOC-V_{CPH} and SSM-5000A, Shimadzu Corp., Kyoto, Japan) at the Center of Excellence on Hazardous Substance Management, Chulalongkorn University, Bangkok, Thailand.

Results

Petrographic Characteristic

The percentages of the maceral types (Table 1) indicate that the organic matter of the oil shales is dominated by the liptinite group which ranges from 10.00–66.50%. The liptinite group is composed mainly of alginite and liptodetrinite macerals. The vitrinite group ranges from 4.00–40.00%, which is composed mainly of gelovitrinite as a cementing material. The inertinite group is rare, and varies from 0.00–3.00% of fusinite. The mineral matter ranges from 21.00–80.50%, which is composed mainly of quartz, clay minerals, and pyrite.

Liptinite is the major maceral group. It consists mainly of alginite in the form of lamalginite, liptodetrinite, sporinite, exsudatinite, and resinite. Alginite originates from algae and is divided into 2 types: lamalginite and telalginite. Lamalginite ranges from 5.50–63.75% and appears as thin lamellar algal mats (Figure 3 (A), (C), (E), (F)). Telalginite ranges from 0.25–9.25% and can be found in 2 species: *Botryococcus* sp. and *Reinclia* sp. The *Botryococcus* sp. shows a polymorphous colony form which is a thick mass and similar to a flower (Figure 3(b)). *Reinclia* sp. shows a thick and long shape (Figure 3(e)). Liptodetrinite ranges from 1.00–12.75% consisting of small

Table 1. The percentage of macerals and mineral matter in Na Hong oil shale

Sampling number	Liptinite (%)	Vitrinite (%)	Inertinite (%)	Mineral matter (%)
PL1-04	25.75	7.75	0.00	66.50
PL1-05	27.50	15.75	0.25	56.50
PL1-06	56.50	17.75	0.25	25.50
PL1-07	55.25	16.25	0.00	28.50
PL1-08	58.50	17.50	0.00	24.00
PL1-09	59.00	11.50	0.00	29.50
PL1-10	55.25	19.00	0.00	25.75
PL1-11	55.25	14.75	2.50	27.50
PL1-12	58.00	15.75	0.00	26.25
PL1-13	55.75	17.00	3.00	24.25
PL1-14	42.75	20.75	0.00	36.50
PL1-15	43.50	7.25	0.00	49.25
PL1-21	50.50	11.75	1.00	36.75
PL1-22	62.00	8.50	1.00	28.50
PL1-23	66.50	11.00	1.50	21.00
PL1-29	10.00	40.00	0.00	50.00
PL1-30	15.50	4.00	0.00	80.50
PL1-31	37.50	11.75	0.00	50.75
PL2-01	49.00	19.75	0.50	30.75
PL2-02	49.50	17.50	2.50	30.50
PL2-03	50.00	24.00	0.00	26.00

fragments or finely detrital particles of liptinites. Sporinite is derived from spores and pollens which generally are preserved in their original forms; it ranges from 0.25–2.25% (Figure 3(d)). Exsudatinite, which ranges from 0.50–1.75%, is a secondary liptinite maceral that fills in cracks, veins, cell lumens, or any available space (Figure 3(c)). Resinite is rare, and is derived from resin and other secretions such as essential oils.

The vitrinite group, which is composed mainly of gelovitrinite, ranges from 4.00–40.00%. It is a homogeneous and structureless gel which fills in cracks or voids and forms a groundmass and/or cements material in oil shale. It shows shrinkage in the form of cracks due to desiccation (Figure 3(a), (d), (f)).

The inertinite group has fusinite in the range from 0.00–3.00%. Fusinite is highly reflective and the cell wall structures are well preserved. The cell lumen voids are usually filled with exsudatinite. (Figure 3(c)).

Mineral matter can be found in all samples ranging from 21.00–80.50%, and is composed mainly of quartz (Figure 3(f)), pyrite (Figure 3(e)), and clay mineral.

Proximate Analysis

From the proximate analysis, the variables of 4 parameters are shown in Table 2. The moisture content is from surface moisture (or free moisture) and inherent moisture (ASTM, 2011). The moisture content ranges from 3.26–5.90%. The highest moisture content is obtained from a dusky yellowish brown sample (A1-13). The ash content, ranging from 53.70–78.98%, is derived from the residue remaining after complete combustion of the samples. The highest ash content is from the brownish gray coloured sample (A1-2). The

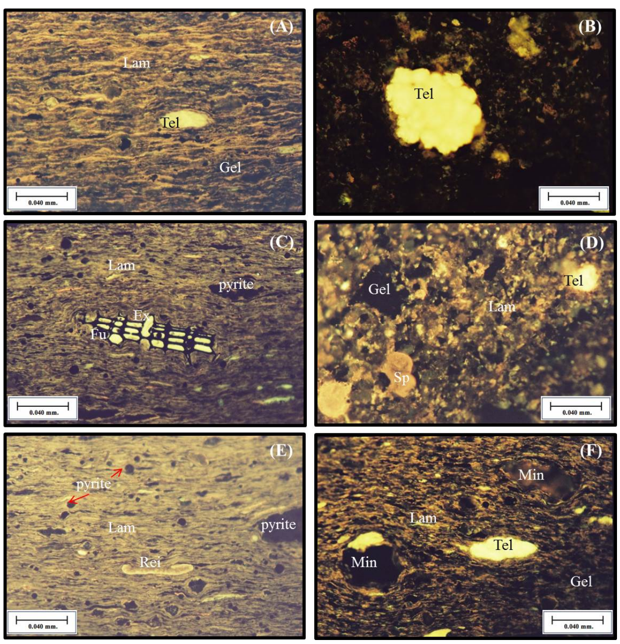

Figure 3. Results from petrographic study under UV-excitation (A) Sample A1-13 (cr.), lamalginite in association with telaginite and gelovitrinite. (B) Sample A1-10 (par.), telaginite (*Botyococcus* sp.). (C) Sample A1-23 (cr.), fusinite in association with exsudatinite inside, lamalginite in association with pyrite. (D) Sample A1-8 (par.), sporinite in association with lamalginite. (E) Sample A1-23 (cr.), lamalginite rich with *Reinchia* sp. and pyrite. (F) Sample A1-11 (cr.), mineral matter associated with lamalginite, telaginite and gelovitrinite. Tel: telaginite; Rei: reinschia sp.; Lam: lamalginite; Sp: sporinite; Ex: exsudatinite; Gel: gelovitrinite; Fu: fusinite; Min: mineral matter; par.: parallel section; cr.: cross section

volatile matter, ranges from 15.83–34.52% and includes methane, hydrocarbons, hydrogen, carbon monoxide, and incombustible gases like carbon dioxide and nitrogen. The fixed carbon is the combustible solid residue remaining after subtracting the percentage of moisture, ash, and volatile matter (on the same moisture basis from 100%). The fixed carbon content ranges from 1.93–21.45%. The highest volatile matter and fixed carbon content are from the brownish black and tough sample (A1-9).

Ultimate Analysis

The results of the ultimate analysis are shown in Table 2. Carbon is an element which is derived from organic materials and carbonate components. From the determined basis, the highest carbon content is 25.61% and the lowest is 10.23%. Inorganic hydrogen in the samples is obtained from moisture (H_2O) and from minerals. However, they are usually a component of the organic compounds (mainly carbohydrate and hydrocarbon) in the source rocks. The highest hydrogen content is 3.95%, the lowest is 1.81%. The nitrogen content in all samples is less than 1.00%. The highest total sulphur content is 8.15%, the lowest is 0.32%. The oxygen content was calculated from the C, H, N, and S using the equation: O = 100–(C+H+N+S). The highest oxygen content is 12.58%, the lowest is 7.72%. The highest content of carbon and hydrogen is from the brownish black and the tough sample (A1-9). The lowest content is from the brownish gray sample (A1-2) which is also poor in nitrogen and sulphur content.

Calorific Value Analysis

The calorific value analysis results are shown in Table 2. The highest calorific value (dry basis) is 3,025 kcal/kg, which is obtained from the brownish black and tough sample (A1-9) and the lowest is 821 kcal/kg, which is obtained from the brownish gray sample (A1-2).

Total Organic Carbon Analysis (TOC)

The TOC is the amount of carbon in an organic compound. The results of the TOC analysis are shown in Table 2. The highest TOC content is 22.3%, which is obtained from the brownish black and tough sample (A1-9). The lowest content is 8.28%, which is obtained from the brownish gray sample (A1-2).

From geochemistry study, plot against the ash content shows the relationship between the carbon, hydrogen, volatile matter, fixed carbon, TOC, and calorific value (Figure 4). This chart shows the increase of the carbon, hydrogen, volatile matter, fixed carbon, and TOC against the decrease of the ash content.

Discussions

From the results, the petrographic study shows that the major maceral is alginite in the groundmass of gelovitrinite and liptodetrinite. The hydrogen rich macerals are mainly of the liptinite and some vitrinite groups, which are related to the high volatile matter and fixed carbon content. Oil shale in the Na Hong basin is concluded to be lacustrine oil shale of the lamosite type. From the appearance characteristics, the dark coloured and tough oil shale samples associated with very fine-grained sediments are related to a high nutrient, stagnant, and deep water environment. The pale coloured and loose oil shale samples are more related to the accumulation of turbulence that carried sediments from high land to be deposited in the lake. If a high sediment supply and low nutrients were transported, the paler coloured oil shale with a high ash content and low grade oil source can be produced. Apparently, the brownish black sample (A1-9) which has the highest liptinite group has the highest content of volatile matter, fixed carbon, carbon, and hydrogen, the highest calorific value, and highest TOC. It can be concluded that the dark coloured and tough samples follow a trend to be high quality oil shale and can be good grade source rocks.

The atomic ratio of hydrogen-carbon (H/C ratio) and atomic oxygen-carbon ratio (O/C ratio) can be used with the van Krevelen diagram (van Krevelen, 1983) to classify the origin of kerogen, as is shown in Figure 5. It shows that the Na Hong samples are a Type II/III kerogen. The relationship between the macerals and kerogen type shows that Type I kerogen is formed from alginite and other macerals in the liptinite group such as liptodetrinite, sporinite, and resinite. They can

Table 2. The result of proximate analysis, ultimate analysis, gross calorific value, and total organic carbon analysis

The mixed sampling number	Sampling number	Appearance characteristics	Proximate analysis (wt.%, on as-received basis)				G (kcal/kg)	Ultimate analysis (wt%, on as determined (air dry) basis)					TOC (wt%)
			M	A	V	F		C	H	N	S	O	
A1-2	PL1-05 PL1-06 PL1-07	Brownish gray, easy to break	3.26	78.98	15.83	1.93	821	10.23	1.81	0.13	0.32	8.53	8.28
A1-3	PL1-08 PL1-09	Dusky yellowish brown, moderately tough	3.26	76.86	16.60	3.28	1,043	11.90	1.91	0.17	0.45	8.70	9.85
A1-4	PL1-10	Brownish black, tough	3.52	69.13	22.50	4.85	1,628	16.22	2.59	0.26	1.98	9.82	13.75
A1-5	PL1-11 PL1-12 PL1-13	Dusky yellowish brown to Brownish black, tough	3.70	66.33	23.79	6.18	1,796	17.51	2.74	0.30	2.92	10.18	15.12
A1-8	PL1-21 PL1-22	Dusky yellowish brown, moderately tough	3.31	76.10	18.03	2.56	1,091	11.17	2.02	0.08	2.91	7.72	8.87
A1-9	PL1-23	Brownish black, tough	3.41	53.70	34.52	8.37	3,025	25.61	3.95	0.38	8.15	8.21	22.31
A1-13	PL1-29 PL1-30 PL1-31	Pale brown to brownish black, moderately tough	5.90	67.22	20.19	6.69	1,436	15.80	2.26	0.38	1.76	12.58	13.04

M: moisture; A: ash; V: volatile matter; F: fixed carbon; G: gross calorific value; C: carbon; H: hydrogen; N: nitrogen; S: sulphur; O: oxygen.

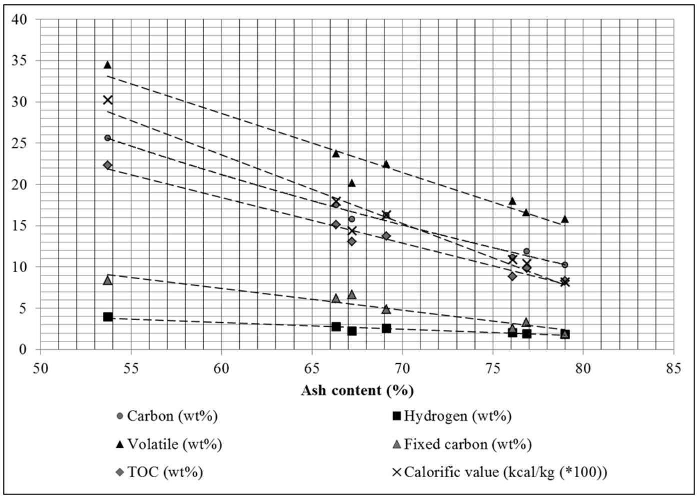

Figure 4. The relationship between carbon, hydrogen, volatile matter, fixed carbon, TOC, and calorific value plotted against the ash content

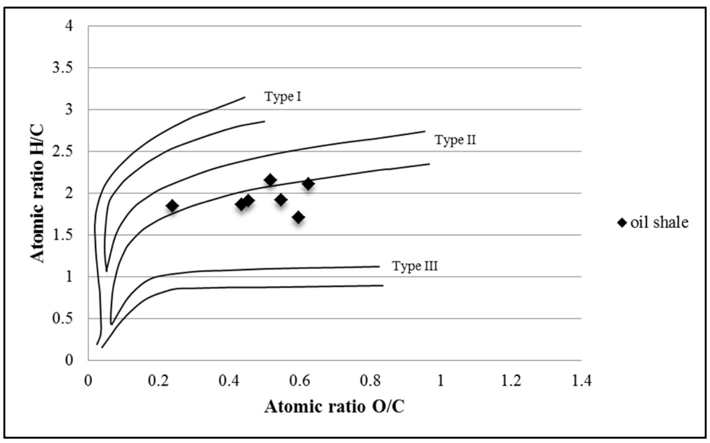

Figure 5. The ratio of H/C and O/C by using Van Krevelen's diagram indicates that the Na Hong samples are a Type II/III oil source (modified from van Krevelen, 1983)

be an excellent source material for oil. Type II kerogen is dominated by the vitrinite group and some of the liptinite group such as sporinite. They are a major source of oil and gas. Type III kerogen is dominated by the vitrinite macerals group that is lacking in lipids or waxy matter. The material of this type is derived from woody terrestrial material; therefore, the oil generation is low but the gas generation is high (Hunt, 1995). The gelovitrinite in the groundmass of all samples indicated that this Na Hong oil shale has a sapropelic origin that is associated with the adjacent coalfield and which contributed to the Type III kerogen.

Conclusions

From the results of the organic petrography and organic geochemistry studies, the following conclusions can be drawn:

1) The petrographic study shows that the Na Hong oil shales are dominated by 10–66.5% of the liptinite group, which is composed

mainly of alginite and liptodetrinite macerals. The vitrinite is composed mainly of gelovirinite which ranges from 4.00–40.00%. Inertinite is rare, with fusinite in the range from 0.00–3.00%. Mineral matter ranges from 21.00–80.50%, and is composed mainly of quartz, pyrite, and other minerals. The petrographic characteristics imply that the depositional environment of the Na Hong basin is a lacustrine environment and that the deposited lacustrine oil shale is of the lamosite type.

2) The van Krevelen diagram shows that the Na Hong samples are a Type II/III kerogen. It is suggested that the oil shales in this basin produce a mixture of gas and oil. The oil shale in the Na Hong basin is related to its sapropelic origin.

3) The appearance characteristics, petrographic characteristics, and geochemistry of oil shale can indicate the grade of the oil shale. The dark coloured and tough oil shale samples are dominated by alginite and liptodetrinite macerals. They are low in ash content, have a low percentage of mineral matter, and are high in fixed carbon, volatile matter, carbon, hydrogen, calorific value, and TOC content. Moreover, they can be related to high grade oil shale. The richness of alginite and liptodrtrinite macerals indicates high nutrient and high water level but a low energy environment. On the other hand, the small number of pale coloured and loose oil shale samples have a low percentage of maceral. They are high in ash content and the percentage of mineral matter and low in fixed carbon, volatile matter, carbon, hydrogen, calorific value, and TOC content. That can be related to low grade oil shale. The poor maceral level indicates low nutrient, shallow water, and a high energy environment that transported larger amounts of inorganic sediments. From the large number of dark coloured and tough oil shale samples, it can be concluded that the oil shale samples in the Na Hong basin have a trend to be high quality oil shale and could lead to good potential oil and gas source rocks.

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References

ASTM. (2011). Standard Test Method for Gross Calorific Value of Coal and Coke. ASTM: D5865-11a. ASTM International, West Conshohocken, PA, USA, 19 p.

ASTM. (2011). Standard Test Methods for Proximate Analysis of Coal and Coke by Macro Thermogravimetric Analysis. ASTM: D7582. ASTM International, West Conshohocken, PA, USA, 9p.

ASTM. (2012). Standard Test Method for Sulfur in the Analysis Sample of Coal and Coke Using High-temperature Tube Furnace Combustion. ASTM: D4329-12. ASTM International, West Conshohocken, PA, USA, 6p.

Hower, J.C., Stanton, R.W., Gammidge, L.C., and Hutton, A.C. (1997). Coal petrology. In: Atlas of Coal Geology. Papp, R.A., Hower, J.C., and Peters, C.D., (eds). American Association of Petroleum Geologists, Tulsa, OK, USA, CD–Rom, Vol. II.

Hunt, J.M. (1995). Petroleum Geochemistry and Geology. W.H. Freeman and Company, NY, USA, 743p.

Hutton, A.C. (1987). Petrographic classification of oil shales. Int. J. Coal Geol., 8:203-231.

Ratanasthien, B. (1997). Algae types of oil source rocks in northern Thailand. Proceedings of the International Conference on Stratigraphy and Tectonic Evolution of Southeast Asia and the South Pacific; August 19-24, 1997; Bangkok, Thailand, p. 606-612.

Ratanasthien, B. (2011). Coal deposits. In: The Geology of Thailand. Ridd, M.F., Barber, A.J., and Crow, M.J. (eds). The Geological Society, London, UK, p. 393-414.

Songtham, W., Ratanasthien, B., Watanasak, M., and Mildenhall, C.D. (2000). Temperate palynological elements from Na Hong basin, northern Thailand. Mahidol Univ. J., 7(2):121-126.

Songtham, W. (2003). Stratigraphic correlation of tertiary basins in Northern Thailand using algae pollen and spore, [Ph.D. thesis]. Graduate School, Chiang Mai University, Chiang Mai, Thailand, 280p.

Stach, E., Mackowsky, M.Th., Teichmüller, M., Taylor, G.H., Chadra, D., and Teichmüller, R. (1982). Stach's Textbook of Coal Petrology. Gebrüder Borntraege, Stuttgart, Germany, 535p.

van Krevelen, D.W. (1993). Typology Chemistry Constitution. 3rd ed. Elsevier Science, Arnhem, Netherlands, 514p.