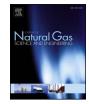
Contents lists available at ScienceDirect



Journal of Natural Gas Science and Engineering

journal homepage: www.elsevier.com/locate/jngse



Geochemical characteristics of the Paleogene-Neogene coals and black shales from Malaysia: Implications for their origin and hydrocarbon potential

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ARTICLE INFO

Keywords: Paleogene-Neogene Coals Black shales Malaysia Rock-Eval Origin Hydrocarbon

ABSTRACT

Paleogene-Neogene coals are widespread in Malaysia, cover a wide age spectrum and are considered the source rock of hydrocarbons in Malaysia. However, they have not been studied systematically up to now. Moreover, the black shales associated with these coals were not taken into consideration in previous studies. Therefore, this study presents systematic inorganic and Rock-Eval analyses of the Paleogene-Neogene coals and their associated black shales to examine their origin, depositional environment and hydrocarbon potential.

With the exception of coals from the Tanjong Formation, Sabah, the Paleogene-Neogene coals are characterized by very low ash yields and low concentrations of trace and rare earth elements including hazardous trace elements. The black shales are composed of quartz, illite, kaolinite and traces of pyrite in some samples. Al₂O₃/TiO₂ ratios in the coals (6-62) and black shales (16-34) suggest a mixture of felsic and intermediate igneous rocks as sources for their detrital fractions, which is supported by the Zr-Ti binary plot. V/(V+Ni) ratios average between 0.8 and 0.7, V/Ni between 13.5 and 3, and Ni/Co between 1.7 and 3.8 for coal and black shale samples; respectively, indicating suboxic to anoxic depositional conditions for both groups of samples. Rock-Eval analysis indicates that coal and black shale samples contain mixed Type II-III kerogens, which suggest similar organic input from terrestrial high plants. The samples also contain immature to mature organic matter and can produce gas and oil. These characteristics along with the high TOC contents (very good to excellent) indicate that the Paleogene-Neogene coals and black shales are potential source rocks for oil and gas fields in Malaysia. The coals and black shales from different ages and localities were shown to have similar source area composition, climate conditions during their deposition, terrestrial organic input as well as suboxic to anoxic depositional conditions. However, coals and black shales from the early-middle Miocene Tanjong Formation exhibit higher Al₂O₃/SiO₂ ratios than the rest of coals and black shales suggesting a prevalence of wetter climatic conditions during formation. In addition, coals and black shales from the early-middle Miocene Tanjong Formation and upper Pliocene Liang Formation show higher values for redox proxies such as V/(V+Ni), V/Ni, Ni/Co and Ce* compared with other formations, indicating the dominance of reducing conditions during the deposition of these formations.

1. Introduction

Coal is a multi-component unit of organic and inorganic materials. Organic matter is a major constituent that determines the combustible energy and sources of hydrocarbons, while inorganic matter consists of elements and minerals of environmental concerns (Romeo, 2014). The constituents of coal depend on the nature of country rocks, depositional environments, diagenesis as well as hydrological conditions (Sia and Abdullah, 2011; Arbuzov et al., 2011; Gürdal, 2011). In many cases, coals are associated with black shales (Baioumy et al., 2011). Therefore, the origin, depositional environments, hydrocarbon potential and environmental impacts of both coals and shales are frequently assessed using organic and inorganic geochemical investigations (Alias et al., 2012; Dai et al., 2016; Romeo, 2014).

The Paleogene-Neogene coals are geographically widespread in West and East Malaysia and are considered source rocks of hydrocarbon

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https://doi.org/10.1016/j.jngse.2018.01.007

Received 24 July 2017; Received in revised form 2 January 2018; Accepted 6 January 2018 Available online 08 January 2018 1875-5100/ © 2018 Elsevier B.V. All rights reserved.

fields in Malaysia. However, previous studies on the Paleogene-Neogene coals were not systematic. Abdullah (1997) investigated the organic geochemistry of coal-bearing sequence of Batu Arang, Peninsular Malaysia. Sia and Abdullah (2011) focused on the concentration and association of minor and trace elements in Mukah coal from Sarawak, with emphasis on the hazardous trace elements. Sia and Abdullah (2012a) highlighted the enrichment of As, Pb, and Sb in Balingian coal from Sarawak. Sia and Abdullah (2012b) studied the organic geochemical characteristics and thermal maturity of low-rank Balingian coal from Sarawak. Alias et al. (2012) investigated the organic geochemical characteristics and depositional environment of coals from Taniong Formation in Sabah. The general focus of the above-mentioned studies was on the organic geochemistry of Paleogene-Neogene coals in Malaysia (e.g. Batu Arang, Balingian, Sabah), without taking into consideration their inorganic geochemistry. Moreover, rare earth element geochemistry of the Paleogene-Neogene coals has not been studied, although coals from Mukah have been investigated for their trace element geochemistry.

In addition, there is the need to explain the phenomenon of coal association with black shales intervals. Furthermore, the geochemical features of these black shales have not been studied in previous research, particularly their inorganic geochemistry. Previous investigations suggested that coals and associated black shales from different ages and localities exhibit similar source area composition, climate conditions during their deposition, terrestrial organic input as well as suboxic to anoxic depositional conditions. On the other hand, some differences have been reported in the paleoclimate and redox conditions in some coals and black shales from various ages and formations. Therefore, this study shows, for the first time, comparative, systematic inorganic (major, trace and rare earth elements) and Rock-Eval analyses of the Paleogene-Neogene coals from Malaysia to determine existing geochemical variations, origin, hydrocarbon potential, depositional environments, maturation as well as environmental impact. The geochemical characteristics of the black shales were compared with those of associated coals.

2. Geological setting

Coals in Malaysia occur in the Paleogene-Neogene basins in three geographical provinces: Peninsular Malaysia, Sarawak and Sabah (Table 1 and Fig. 1A).

2.1. Peninsular Malaysia

Paleogene-Neogene sediments of Peninsular Malaysia occur on its West Coast in a series of small basins mainly at the Batu Arang area, which comprises a variety of sedimentary and meta-sedimentary rocks as well as Quaternary sediments (Fig. 1B). Recent analysis of palynomorph assemblages indicates the Batu Arang coals are Eocene to Oligocene in age (Ahmad Munif, 1993). The Paleogene-Neogene strata at Batu Arang are classified into two sequences: an upper sequence known as 'Boulder Beds' that unconformably overlies the lower sequence referred to as 'Coal Measures' (Roe, 1951; Stauffer, 1973). The Coal Measures consist of: 1) sandstone, with intercalations of shale and clay, 2) shale, 3) coal beds, shale and sandstone, and 4) sandstone with shale, clay and conglomerate intercalations (Roe, 1951). The Coal Measures are composed of two main coal seams (upper and lower). The upper seam averages 9 m, while the lower seam is approximately 6 m thick (Roe, 1953) (Fig. 2A). The coal occurs not only as thin laminae or streaks in black shales, but are also amassed as layers of more than 30 cm thick.

2.2. Sarawak

Coals from Sarawak were described and sampled in four formations, namely; Nyalau, Liang, Begrih and Balingian formations in chronological order (Table 1, Fig. 1).

The Nyalau Formation comprises fine to medium grained sandstones that alternate with shale. In Bintulu area, the Nyalau Formation contains frequent occurrences of coal beds and infrequent thin limestones (Fig. 1E). The contact with the underlying Buan is conformable and represents a sharp transition from argillaceous to arenaceous rocks. To the east, the Nyalau Formation is conformably overlain by the Sibuti Formation and to the northeast it interfingers with the Setap Shale Formation (Fig. 2B). Two coal beds were identified in the Nyalau Formation (Bintulu area). The coal beds range from 20 cm to 1 m in thickness and intercalate with grey massive mudstone. Black shales are observed at the base of the succession.

The Balingian Formation consists of a thick (> 3500 m) sequence of sandstone, pebbly sandstone, fossiliferous mudstone, coals and lignite (De Silva, 1986). The Formation is Late Miocene in age (Wolfenden, 1960; Nugraheni et al., 2014). It is unconformably overlain by the Begrih Formation along the Mukah road (Fig. 1C, D and Fig. 2B). The coalfield of Balingian Formation contains 12 coal seams, comprising 5 major (well-developed, with economic potential) and 7 minor seams. Black shales overlie the coal beds and also occur as thin beds.

The Begrih Formation (Fig. 1C, D and Fig. 2B) is composed of conglomerates, conglomeratic sandstones, mudstones, shales, tuffs and coals (Hutchison, 2005). According to Wolfenden (1960) and Nugraheni et al. (2014) Begrih Formation is Lower Pliocene in age. Two thin coal beds (varying thickness of 30–60 cm) intercalated with mudstone were sampled. No black shales were reported for this Formation.

The Liang Formation overlies the Begrih Formation (Fig. 1C, D and Fig. 2B) and has a thickness of approximately 950 m. The Formation is made up of thick and massive clays, sands, tuffs, coal seams and gravel lenses. The fauna identified in the coal zone suggest a brackish-water

Table 1

Locations, ages, formations, lithology, coordinates and number of samples of the Paleogene-Neogene coals and black shales from Malaysia.

Location	Formation	Age	Lithology	Coordinates	Number of analyzed black shale samples	Number of analyzed coal samples
Sarawak	Liang	Upper Pliocene	Clays, sands, tuffs, coal seams and gravel	N 02° 40′ 11.41″ E 112° 20′ 22.56″	2	5
	Begrih	Lower Pliocene	Conglomerates, conglomeratic sandstones, mudstones, shales, tuffs and coals	N 2°45′ 27.47″ E 112° 20′ 34.98″	-	2
	Balingian	Late Miocene	Sandstone, pebbly sandstone, fossiliferous mudstone, coals and lignite	N 02° 47′ 98.40″ E 112° 23′ 19.41″	2	5
Sabah	Tanjong	Early - Middle Miocene	Mudstones, siltstones, limestones, conglomerates and coals	N 04° 30′ 58.0″ E 117° 14′ 32.0″	5	1
Sarawak	Nyalau	Eocene-Oligocene	Sandstones, black shale, coals	N 3°11′ 32.98″ E 113° 5′ 17.62″	1	2
Peninsular Malaysia	Batu Arang		Sandstone, black shale, clay, coals	N 3°19′ 22.01″ E 101° 28′ 30.03″	3	3

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