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The Potential of Eocene Shale of Nanggulan Formation as a Hydrocarbon Source Rock

Potensi Serpih Eosen Formasi Nanggulan sebagai Batuan Sumber Hidrokarbon

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Abstract

In western Indonesia, Eocene shale is generally considered as a potential source rock. The Nanggulan Formation outcropping at Kulonprogo-Yogyakarta consists of a shale interval of Eocene in age. Analysis of its organic material content, kerogen type, and maturity level were conducted to know its potential. The laboratory analysis of eleven samples were done to measure its TOC content. Samples with TOC > 0.5% then were analyzed to measure its Rv and TAI. Maturity level was also calculated by TTI from burial history model. The result of analysis shows there are various TOC contents and seven samples of them are categorized into a good-excellent class (TOC > 1%). Kerogen content of those samples is type III (non fluorescene amorphous-humic kerogen). One sample has a good indication of hydrocarbon formation (PY = 9.0 mg HC/g rock). Unfortunately thermal maturity level of the samples is immature (highest Rv 0.39, T_{max} 422^{o}C, and TAI 2). Otherwise, TTI calculation result from subsurface burial history modelling indicates that some areas are mature having reached gas window since 0.4 mya, especially in the area which had been influenced by a volcanic intrusion at Oligocene (28.5 mya). Therefore, the Nanggulan Formation shale has a potential capacity as a source rock with some limitation in maturity level.

Keywords: potential, Eocene shale, hydrocarbon, source rock, maturity, Nanggulan

Sari

Di Indonesia bagian barat, serpih berumur Eosen umumnya dianggap sebagai batuan sumber yang potential. Formasi Nanggulan yang tersingkap di Kulonprogo-Yogyakarta terdiri atas serpih berumur Eosen. Analisis terhadap kandungan bahan organik, tipe kerogen, dan tingkat kematangan dilaksanakan untuk mengetahui potensinya. Analisis laboratorium terhadap sebelas percontoh dilakukan untuk mengukur kandungan TOC nya. Kemudian percontoh yang kandungan TOC > 0,5% diukur reflektan vitrinit (Rv) dan TAI (Indeks Alterasi Termal)-nya.Tingkat kematangan juga diukur dengan TTI dari model sejarah penimbunan. Hasil analisis menunjukkan adanya beragam kandungan TOC dan tujuh percontoh termasuk level baik-istimewa (> 1%). Kerogennya termasuk tipe III (kerogen humic-amorf non-fluorescence). Satu percontoh memperlihatkan indikasi bagus sebagai pembentuk hidrokarbon (S1=1,38 & S2=7,62 mgHC/g batuan), sedangkan tingkat kematangan percontoh termasuk belum matang (maksimum Ro 0,39, T_{maks} 422^{o}C, dan TAI 2). Sebaliknya, hasil perhitungan TTI dari model sejarah penimbunan menunjukkan bahwa beberapa tempat telah matang dan mencapai gas window sejak 0,4 jtl., terutama daerah yang terpengaruh oleh intrusi pada Oligosen (28,5 jtl.). Karenanya, serpih Formasi Nanggulan berpotensi sebagai batuan sumber dengan beberapa limitasi pada tingkat kematangannya.

Kata kunci: *potensi, serpih Eosen, hidrokarbon, batuan sumber, kematangan, Nanggulan*

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Introduction

The Eocene shale of Nanggulan Formation outcropping at Kalisonggo River, Kulon Progo, about 25 km to the west from Yogyakarta City (Figure 1), is present as black to grey shale and claystone with thickness of about 3 m (Figure 2). The shale is massive and relatively tilted to the south. Stratigraphically, the Nanggulan Formation was deposited unconformably on the basement and is overlain by Old Andesite Formation or OAF (Figure 3). The formation consists of shale, claystone, shaly sandstone, and some carbonaceous shale. Large foram is commonly found in the shale such as Nummulites and Discocyclina. Interpretation of gravity data (Figure 4 and Table 1) shows that the Eocene sediments in the subsurface are about 1,100 m thick distributed widely from Kulon Progo area to theYogyakarta Sub-basin.

The main aim of this research was to evaluate the source rock potential and estimate its level of thermal maturity from both surface samples and sub-surface modeling. Therefore, a total of eleven outcrop samples from Kalisonggo River and its surrounding areas were collected for hydrocarbon geochemical analysis.

Methodology

The methods used in the study were field and laboratory analyses. The field analysis was sample collecting for laboratory analysis preparation. Whilst laboratory analysis comprises total organic carbon (TOC) content, which then was followed by Rock-Eval Pyrolysis, reflected light microscopy, and transmitted light microscopy carried out for samples with TOC values > 0.50 %.

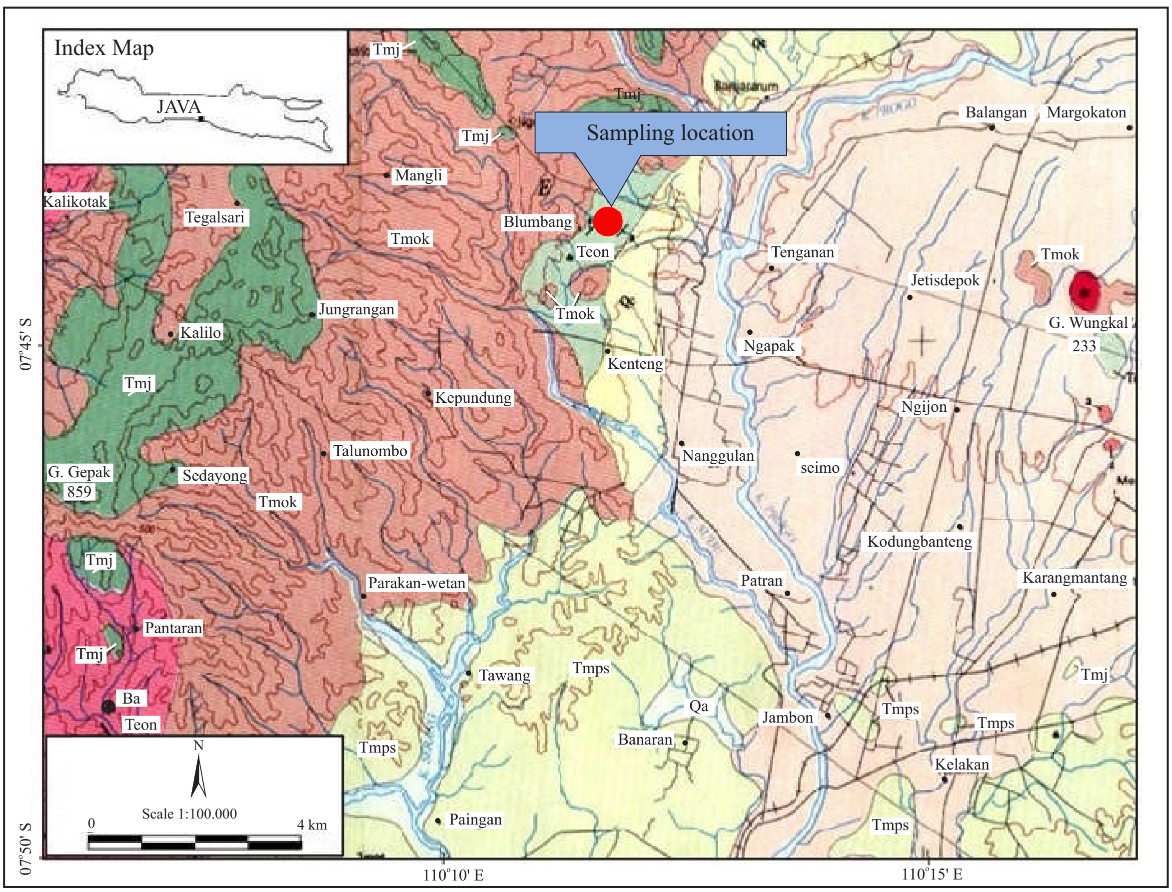

Figure 1. Map of sampling location (red dot), in Nanggulan area, West Kulon Progo.

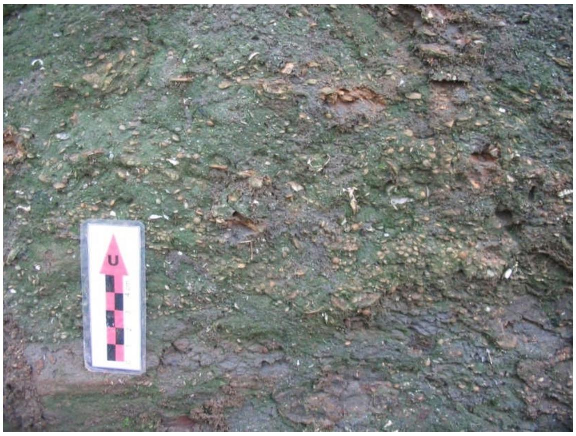

Figure 2. Photograph of Nanggulan Formation outcrop, showing greenish grey shale with large foram such as Nummulites and Discocyclina. The scale is about 20 cm height.

Sample Preparation

Preparation techniques are directed towards obtaining clean samples by washing them with cold water and crushed to approximately peas-sized fragments for kerogen preparation or finely milled for chemical analysis.

Total Organic Carbon (TOC)

Total organic carbon content is determined by pulverizing depicted samples, treating carefully weighed portion with warm hydrochloric acid to remove carbonate minerals, and analyzing the residue for carbon content with a Leco Carbon Sulfur Determinator SC-144-DR. It is generally accepted that samples with less than 0.5 weight-% TOC cannot yield sufficient hydrocarbon to form commercial deposits and are therefore considered as non-sources;

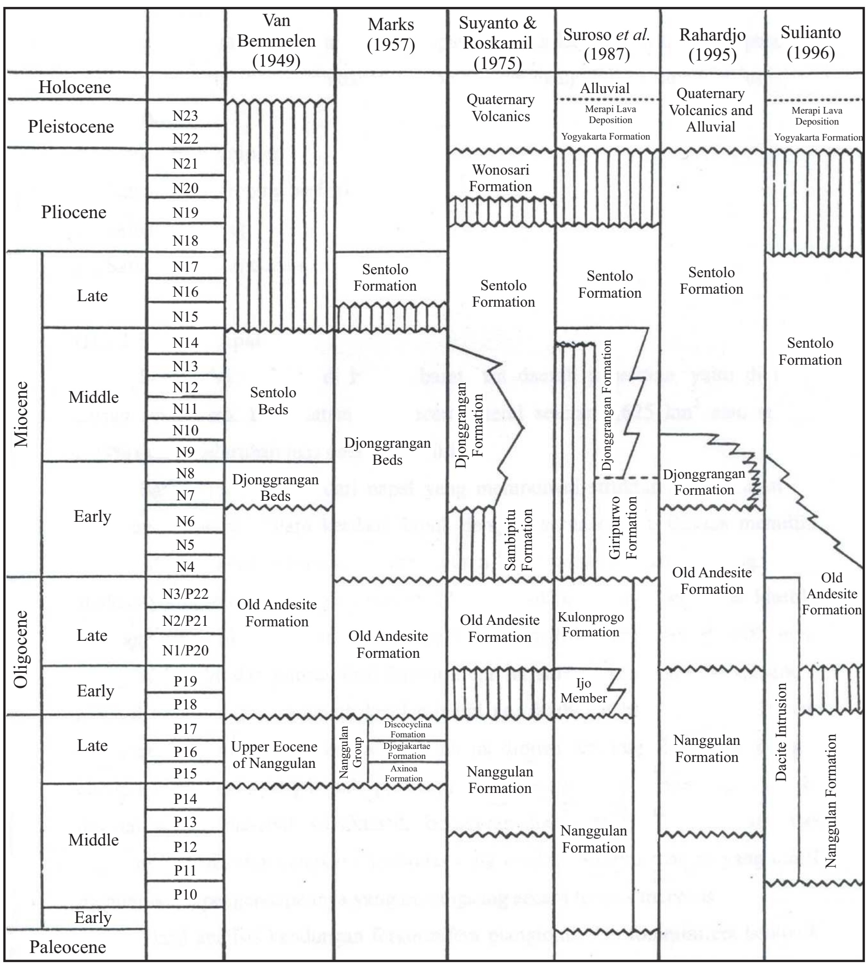

Figure 3. Regional stratigraphic columns from several authors, shows that Nanggulan Formation is Eocene in age and unconformably overlain by the Oligocene rock of Old Andesite Formation (OAF).

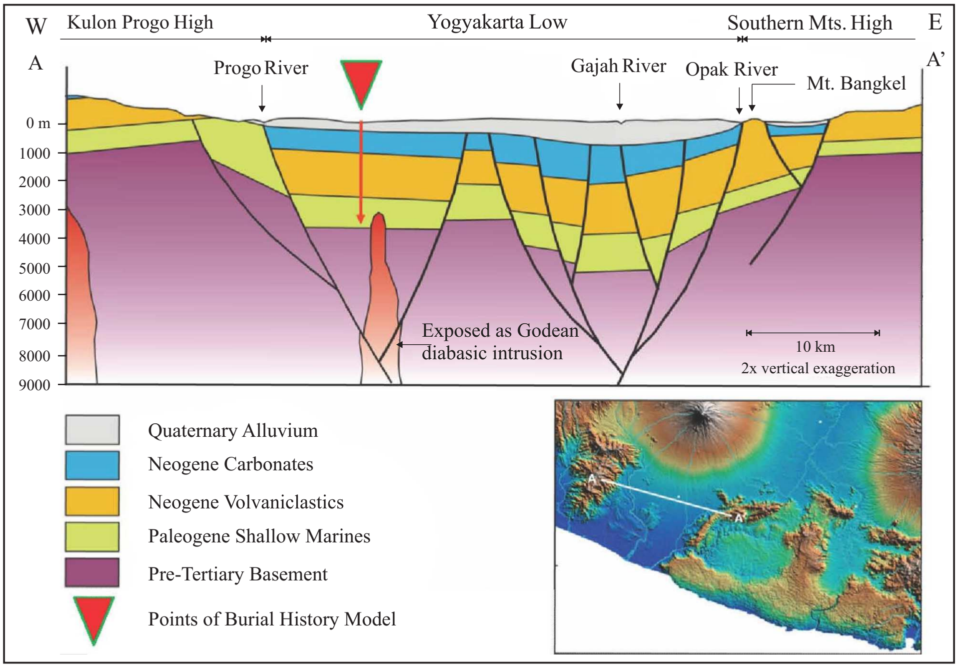

Figure 4. Subsurface interpretation from gravity data in Kulon Progo section and the point where the burial history model is build (modification from Pertamina, 2008).

Table 1. Subsurface Data estimated from Gravity Interpretation (Pertamina, 2008)

Unit	Age (mya)	Thickness (m)	Depth (m)	Density (g/cc)
Quaternary Alluvium	1.78	250	200	1.59
Neogene Carbonates	19	800	1000	2.49
Neogene Volcaniclastics	28.5	1500	2500	2.49
Paleogene Shallow Marine	54.8	1100	3600	2.63

samples between 0.5 and 1.0 weight-% TOC are rated as marginal in source quality; samples of 1.0 and 2.0 weight-% TOC are rated as average (good) in source quality, whilst samples with more than 2.0 weight-% TOC are considered to be above average (rich) in source quality.

Rock-Eval Pyrolysis

The rock-eval pyrolysis performed on whole rock samples is to distinguish the types of organic matter (OM) and to evaluate the source rock potential. The data of hydrocarbon are expressed as mg/g including basic parameters as follows:

1. S1 represents the quantity of oil (free hydrocarbons) present in the rock and is roughly analogous to the solvent extractable portion of the organic matter.
2. S2 represents the quantity of hydrocarbon released by the kerogen in the sample during pyrolysis between 300^o and 600^{o}C.
3. T_{max} in ^{o}C, is the temperature at which the maximum rate of generation (at the S2 peak) occurs.

In addition, when the ratio of S2/TOC showing a quantity of Hydrogen Index (HI=mg HC/g TOC) plotted against Tmax values, it provides a general indication of kerogen quality (type) and reveals whether oil or gas likely to be generated. Furthermore, the ratio of (S1)/(S1+S2) indicates the total production Index (PI) showing the thermal maturity.

Reflected Light Microscopy

A sample of ground rock is treated successively with hydrochloric and hydrofluoric acids to concentrate the kerogen, freeze-dried, mounted in an epoxy and polished. A determination using an image analysis QDI 2000 Microspectrophotometer with color digital imaging system.

Transmitted Light Microscopy

The transmitted light slide is prepared with standard palynological techniques and determined by a Nikon Fluophot Microscope. SCI values on all spores and pollen present are recorded on a 1 to 10 scale. A histogram prepared is used to interpret SCI maturities in a manner similar to that used for vitrinite reflectance.

Evaluation of maturity level fro the subsurface shale was conducted using TTI (Time Temperature Index) from burial history modelling which then was converted to its Rv values. Normal geothermal gradient in this basin is about 2.34^{o}/100 m (Terres and Mertani, 1987), but due to an intrusion in 28 mya, the geothermal gradient is predicted to increase to 3 - 4^{o}/100 m (Table 2).

Result and Discussion

Source Rock Potential

Data gained from Rock-eval pyrolysis are interpreted in the following manner (Tissot and Welte, 1984; Rad, 1984; Espitalie *et al.*, 1985: Bordenave, 1993):

1. S1 data are not used, except on a high S1 value associated with a low S2 value which is interpreted as the presence of migrated hydrocarbon.
2. S2 value is an indicator of source rock quality:
   - Poor source rock: S2 < 2.5 kg/ton
   - Marginal/fair source rock: S2 = 2.5-5 kg/ton
   - Good source rock: S2 > 5 kg/ton
3. Organic matter maturation degree is defined by T_{max}, *i.e.*:
   - T_{max} : < 435^{o} C ------- immature
   - T_{max} : 435^{o} - 470^{o}C --- oil
   - T_{max} : > 465^{o} C --------- gas
4. Production Index (PI) is used for sample with a richness of fair or better, associated within T_{max} value as follows:

Table 2. Temperature Gradient from some Basins in Indonesia (after Terres & Mertani, 1987)

Basin	Temperature Gradient (G) (^{o}C/100m)	Thermal Conductivity K (mcal/cm.cu^{o}C)	Heat Flow Value (Q) (10^{-6}cal/cm^{2}sec.=HFU)
N. Sumatra	4.69	5.01	2.35
C. Sumatra	6.76	4.84	3.27
S. Sumatra	5.22	4.90	2.56
Sibolga	2.14	5.65	1.21
Bengkulu	2.15	4.51	0.97
Sunda	4.63	4.49	2.08
N.W. Java	4.31	4.45	1.92
Biliton & Pati	3.84	5.26	2.02
South Java	2.43	4.86	1.18
N.E. Java	4.14	5.07	2.10
N.E. Java Sea	3.50	4.63	1.62
Makassar	3.18	5.34	1.70
Asem-Asem/Pasir	3.28	5.39	1.77
Barito	3.46	5.63	1.95
Kutei	3.13	5.24	1.64
Tarakan	3.40	4.94	1.68
E. Natuna	3.24	4.81	1.56
W. Natuna	3.81	5.36	2.04
Salawati	4.44	5.16	2.29
Bintuni	3.65	4.33	1.58

T_{max}	PI	Maturity
< 435°C	< 0.1	immature
> 435°C	0.1 - 0.2	early mature mature
> 0.2		contaminant

5. Source quality (hydrocarbon proneness) was also determined using hydrogen index (HI) as follows:

Source Quality	HI (mgHC/g TOC)
Oil-prone	>300
Gas-Oil prone	200 - 300
Gas prone	110 - 200
Non-source	<110

6. Generation zone of Rv is as follows: < 0.60
   - Rv: Immature 0.60 - 1.20 %
   - Rv: Mature (oil) >1.20
   - Rv: Over mature (gas)
7. Generation zone of SCI/TAI is as follows:

SCI	TAI	Maturity Level
< 4.5	<2+	Immature
4.5 - 7.5	2+ - 3+	Mature
> 7.5	>3+	Over mature

The source rock potential was estimated by TOC and Rock-Eval Pyrolysis analyses. The summary of geochemical or screening analysis is shown in Table 3. The samples comprise grey shales with some carbonaceous matter contents. The TOC content within these eleven samples are low to excellent, ranging from 0.37 wt.% to 11.28 wt.%. Furthermore, nine shale samples having TOC values between 0.58% and 11.28% are categorized as 'fair to excellent potential source rock' for generating hydrocarbon. Whilst the rest two samples is considered as 'non potential source rock (TOC < 0.5%) due to their TOC values less than 0.5%.

Detailed diagrams for interpreting the rock eval pyrolysis results are performed in Figures 5 and 6. Result of rock-eval pyrolysis analysis of the selected sediments indicates that only two samples of code: KP/03/S/WPR-38a and KP/03/S/WPR-38b have kerogen content (S2) > 2.5 mg/g rock. Those values being 3.85 and 7.62 mg/g rock respectively, are categorized as 'fair to very good' potential source rock for generating hydrocarbon, when they reach a mature stage in the thermal maturity level. The other sediments showing S2 < 2.5 mg/g rock tend to indicate their low potential for hydrocarbon source rock or non-source ones (Rad, 1984).

Kerogen Type

Determining kerogen type of nine selected samples has been examined by using transmitted light microscopy supported by hydrogen index values. Microscopic examination results shows that generally sediments in the formation are predominantly Non-Fluorescene amorphous types (humic kerogen, type III). Moreover, hydrogen index (HI) is generally low, shown by the dominant values of less than 68 mg HC/g TOC (Table 3). However, only one sample (KP/03/S/WPR 03) having value of 129. This condition suggest that those kerogen have no or low capability to produce hydrogen, except sample KP/03/S/WPR 03. Cross plots between HI *vs.* T_{max}, HI *vs.* OI, and TOC *vs.* S2 (Figures 5a,b,c,d) also indicate that the kerogen type mostly are type III, tending to be a gas-prone.

Thermal Maturity

Thermal maturity assesment was done on the basis of T_{max} (Table 3), vitrinite reflectance (Rv), and Thermal Alteration Index (TAI). No vitrinite reflectance measurements obtained from four samples due to the barren vitrinite particles. Only sediments with sufficient content of both vitrinite and/or spore/pollen for both Rv and TAI analyses are considered to be more reliable.

Some sediments show a very low S2 value and therefore no T_{max} data is considered to be reliable for inferring the thermal maturity. All of the samples have T_{max} value < 435°C. This condition tends to indicate an immature sediment level. Microscopic examinations show that all sediments have Rv values varying between 0.23 and 0.39 %. Those tend to show an "immature" thermal stage (Robert, 1988, in Bordenave, 1993). Moreover, it is slightly concommitant with the value of TAI ranging from 2- to 2+ or pale yellow to golden yellow, which falls under an immature - early mature level (Traverse, 1988). The corresponding Rock-Eval T_{max} values for these samples are consistent with the maturity level indicated by the vitrinite reflectance analysis (Figure 5d). However, PI having dominantly values of 0.15 - 0.27 (Table 3) suggests the early mature to mature thermal level. Therefore, the PI values are not reliable for a maturity level determination from a point of geochemical analysis view. High PI values

Table 3. Result of Geochemical Analysis

NO	Sample No.	Lithology	TOC (%)	S1	S2	S3	PY	S2/ S3	PI	PC	T_{max} (0C)	HI	OI	RO
				mg/g										
1	KP/03/S/WPR 28	Clyst, gngy	0.37	-	-	-	-	-	-	-	-	-	-	-
2	KP/03/S/WPR 41	Clyst, ltgy/gy	0.48	-	-	-	-	-	-	-	-	-	-	-
3	KP/03/S/WPR 17	Clyst, dkgy	0.58	0.05	0.21	0.19	0.26	1.11	0.19	0.01	NDP	36	33	-
4	KP/03/S/WPR.32	Clyst, gy/dkgy,slty	0.68	0.19	0.20	0.30	0.39	0.67	0.49	0.03	418	29	44	0.34
5	KP/03/S/WPR 03	Clyst, gy/dkgy	1.56	0.46	2.01	0.61	2.47	3.30	0.19	0.21	419	129	39	-
6	KP/03/S/WPR 37	Clyst, dkgy	1.57	0.13	0.36	0.47	0.49	0.77	0.27	0.04	398	23	30	0.33
7	KP/03/S/WPR 27	Clyst, dkgy	1.64	0.09	0.32	0.96	0.41	0.33	0.22	0.03	405	20	59	0.29
8	KP/03/S/WPR.30	Sh, dkgy, sl calc	1.81	0.16	0.73	0.77	0.89	0.95	0.18	0.07	422	40	43	0.28
9	KP/03/S/SG 66	Clyst, dkgy, sl carb	4.07	0.31	0.92	4.24	1.23	0.22	0.25	0.10	413	23	104	0.26
10	KP/03/S/WPR.38a	Sh, dkgy/blk, sndy, carb	9.27	0.74	3.85	3.61	4.59	1.07	0.16	0.38	401	42	39	0.39
11	KP/03/S/WPR 38b	Sh, dkgy/blk, carb	11.28	1.38	7.62	5.02	9.00	1.52	0.15	0.75	404	68	45	0.23

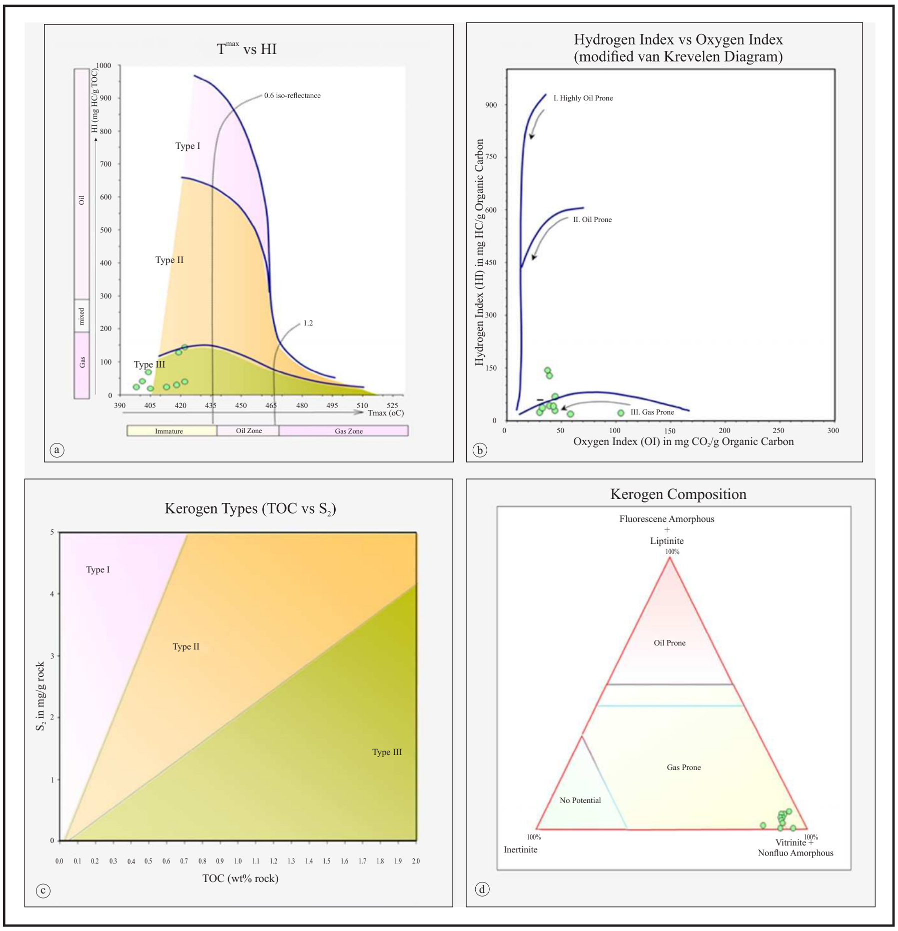

Figure 5. Kerogen type and composition determination from cross plot T_{max}-HI (a), HI-OI (b), TOC-S2 (c), and vitrinite reflectance analysis (d), showing that the kerogen mostly are type III and derived from vitrinite and non fluorescence amorphous material.

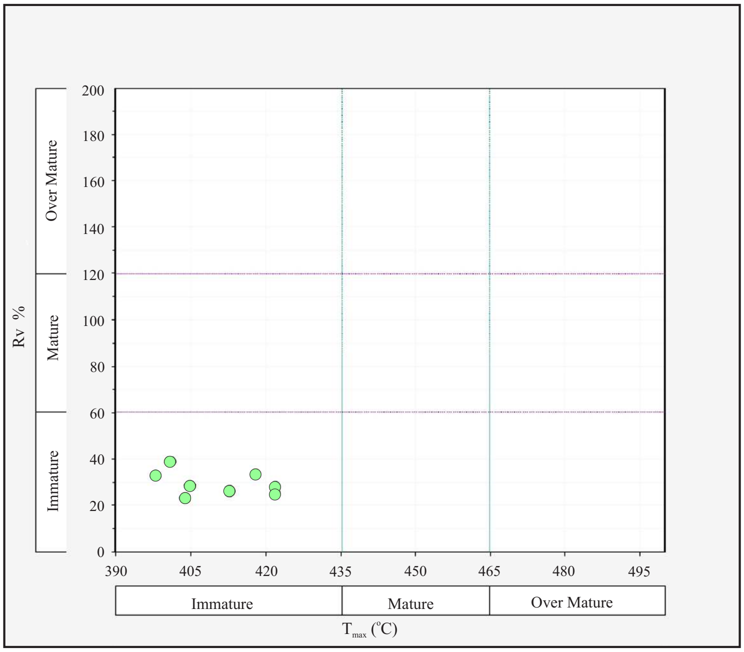

Figure 6. Subsurface interpretation from gravity data in Kulon Progo section and the point where the burial history model is build (modification from Pertamina, 2008).

within samples poor in organic matter do not reflect the maturation level. The PI, generally, is more suited as a maturity trend parameter (Rad, 1984).

Burial History Model

Although maturity level of the surficial samples indicates an immature stage, there is another chance to evaluate maturity level from subsurface data, since the overburden and the presence of volcanic intrusion would give a higher temperature. Therefore, a burial history model and calculated maturity level by Time Temperature Index (TTI-Lopatin method) were used which then were converted to Rv value.

Location point for creating the burial history model was chosen from subsurface-gravity interpretation (Figure 4). The site is near the Godean diabasic intrusion predicted to increase the geothermal gradient up to 3 - 4°C/100 m, higher than normal gradient in South Java basinal area which is about 2.43°C/100 m (Table 2). The intrusion took place about 28.5 mya (Late Oligocene). The surface temperature which was used for the model is 24° C.

Other subsurface data such as density and sediments thickness (Table 1) were estimated from gravity interpretation (Pertamina, 2008). Those data were used for creating burial history model (Figure 7). There were four sediment layers interpreted from the gravity data *i.e.* Paleogene shallow marine sediments overlying Eocene shale of the Nanggulan Formation, Neogene volcaniclastics equal to OAF & Jonggrangan Formation, Neogene carbonates of the Sentolo Formation, and Quaternary alluvium sediments in the uppermost part of the layers. The deepest sediments reach up to 3600 m in depth.

Time Temperature Index and Rv

Using Lopatin method, Time Temperature Index (TTI) from burial history model is calculated (Table 4) and converted to Rv value (Table 5). Since the kerogen is type III, Rv values for oil window are ignored and it focused on gas window. The temperature for wet gas generation would be reach if the Rv is equal to 1.35. The converted number to TTI value is 180 and in the modeling that number had reached at 0.4 mya. So, it looks like that the maturity level of subsurface sediments from Paleogene (Eocene shale of the Nanggulan Formation) in the studied area was mature enough to generate gas (Figure 8).

Table 4. The Result of TTI Calculation

Temp Interval [C]	Temp Factor	Time Factor	Interval TTI	Total TTI	Time (mya)
30 - 40	0.01	10.00	0.08	0.08	39.00
40 - 50	0.02	9.50	0.15	0.23	29.50
50 - 60	0.03	2.80	0.09	0.31	26.70
60 - 70	0.06	1.70	0.11	0.42	25.00
70 - 80	0.13	1.80	0.23	0.65	23.20
80 - 90	0.25	1.20	0.30	0.95	22.00
90 - 100	0.50	1.00	0.50	1.45	21.00
100 - 110	1.00	1.20	1.20	2.65	19.80
110 - 120	2.00	0.80	1.60	4.25	19.00
120 - 130	4.00	5.60	22.40	26.65	13.40
130 - 140	8.00	5.40	43.20	69.85	8.00
140 - 150	16.00	6.00	96.00	165.85	2.00
150 - 160	32.00	2.00	64.00	229.85	0.00

Table 5. TTI to Rv Conversion and Its Time Estimation, Rv 1.35 (wet gas generation) equal to TTI 180 reached about 0.4 mya

Maturity Level	Rv	TTI	Time (mya)
Early mature of oil	0.6	10	17.6
Peak mature of oil	0.65	15	16.3
Late mature of oil	0.9	50	10.9
Early mature of wet gas	1.35	180	0.4

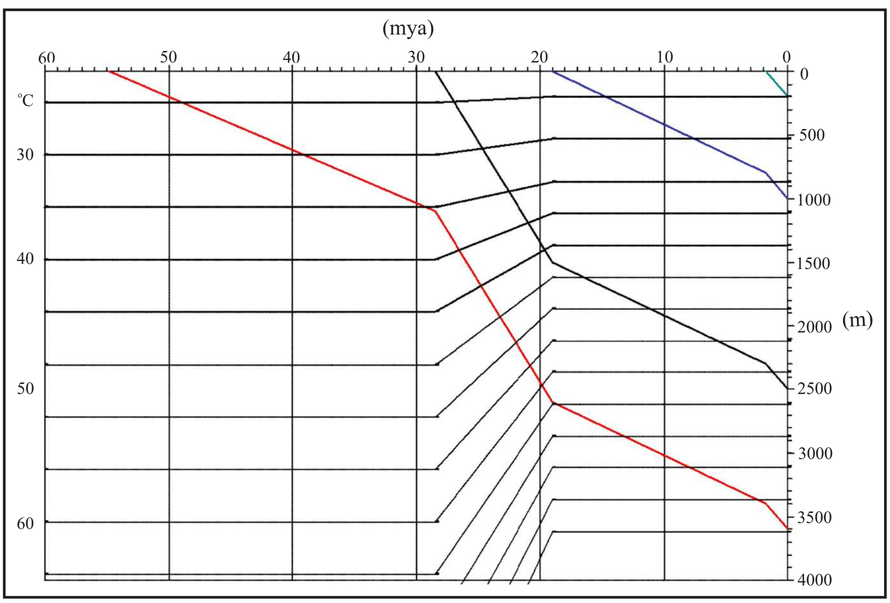

Figure 7. Burial history and plot of temperature gradient. For normal temperature is estimated about 2.43°C/100 m and the intrusion at 28.5 mya influenced to increasing the gradient about 3° - 4°C/100 m.

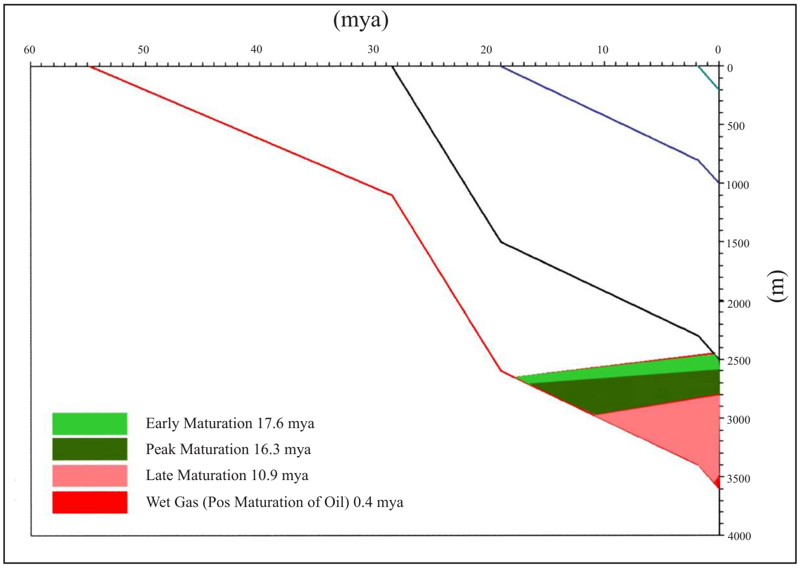

Figure 8. Maturity model created in study area, and because the kerogen is type III then hydrocarbon produced is wet gas. Generation time for reaching gas window level is estimated about 0.4 mya.

Differences of Thermal Maturity Stage

Geochemical analysis of the surficial samples indicates that all the samples are immature. However, from the subsurface modeling point of view, it is predicted that the Eocene shale of Nanggulan Formation had reached a mature stage to produce gas since 0.4 mya. The most important factor in this difference result of the analysis is the presence of intrusion that had influenced the geothermal gradient in subsurface area. The increasing number of the gradient is believed to be able to make the temperature in subsurface reached the gas window. Therefore, it is suggested that the next research related with this topic should be focused on a study about the relationship between intrusion and geothermal gradient to know precisely the correction number which must be added to a normal geothermal gradient in a similar case.

Conclusions

Some surficial samples show a fair to excellent TOC content for nine samples, however two of them have kerogen content (S2) > 2.5 mg HC/g rock; so generally the Eocene shale of Nanggulan Formation is categorized as a low to very good potential source rock.

From a microscopic examination and cross-plot Sbetween HI *vs*. T_{max}, HI *vs*. OI, and TOC *vs*. S2, the kerogen type mostly are of type III (humic/ non-fluorescene amorphous) leading to gas prone conditions.

Although the surface samples indicate an immature stage, except the PI values showing an early to mature level, a subsurface modeling shows that the Eocene shale of Nanggulan Formation had reached a mature stage to produce gas since 0.4 mya.

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