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An organic geochemical study of the Miocene sedimentary sequence of Labuan Island, offshore western Sabah, East Malaysia

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Abstract: Labuan is an island, located offshore west of Sabah, East Malaysia. The rocks on Labuan Island are divided into three main units; Temburong, Setap Shale and Belait formations. The relationship between these units is still unclear. An organic geochemical study was performed on ten samples from these main rock units. The main aim of this study was to determine the thermal maturity of these sediments and to differentiate between the formations based on organic geochemical parameters. The study suggests the thermal maturity is early mature to mid mature for oil generation, the Belait Formation being least mature and Temburong the most mature. Based on organic geochemical parameters and supported by vitrinite reflectance data, the Layang Layangan Unit I should be grouped within the Belait Formation.

Abstrak: Labuan adalah sebuah pulau yang terletak di luar pesisir pantai di bahagian barat negeri Sabah, Malaysia Timur. Batuan di Pulau Labuan dibahagikan kepada tiga unit utama: Formasi Temburong, Syal Setap dan Formasi Belait. Hubungan di antara unit-unit ini masih kurang jelas. Suatu kajian geokimia organik dilakukan ke atas sepuluh sampel dari unit-unit utama batuan ini. Tujuan utama kajian ini ialah untuk menentukan tahap kematangan terma sedimen-sedimen ini dan untuk membezakan di antara formasi-formasi ini berdasarkan parameter-parameter geokimia organik. Kajian ini mencadangkan kematangan terma adalah diperingkat awal ke pertengahan bagi penjanaan minyak di mana Formasi Belait yang paling kurang matang dan Temburong yang paling matang. Berdasarkan parameter-parameter geokimia organik dan disokong oleh data dari pantulan vitrinit, Unit I Layang Layangan sepatutnya diletakkan di dalam Formasi Belait.

INTRODUCTION

The origin of organic matter in sedimentary rocks and crude oils has received much attention. In recent years, the information contained within biomarker distribution has been successfully used for the differentiation of depositional environments and assessment of thermal maturity (e.g. Peters and Moldowan, 1993). It is evident that the components of a particular sediment extract or oil are a reflection of both the paleoenvironmental conditions and the precursor compounds in the organisms which contributed organic matter at the time of sediment deposition, and therefore can provide valuable information about the organic input and the prevailing depositional environment. In this study, evaluation of thermal maturity and depositional conditions is made based on biomarker distribution and vitrinite data. The stratigraphic position of Layang Layangan Unit I in Labuan Island is reinvestigated in the light of the current data.

GEOLOGICAL SETTING

Labuan Island is located offshore west Sabah, east Malaysia (Fig. 1). It forms part of the Labuan-Muara redge, constituting one of a series of approximately north-south trending anticline structures within the Inboard Belt structural zone of the Sabah continental margin (Hazebroek and Tan, 1993).

The Island is made up entirely of sedimentary rocks. These rocks are Lower to Middle Miocene age and can be correlated to those of onshore west Sabah, Brunei, and northern Sarawak (Liechti *et al.*, 1960; Wilson, 1964; Potter *et al.*, 1984; Koopman and Schreurs, 1996). Wilson, 1964 divided Labuan rocks to three main units: Temburong Formation, Setap shale and Belait Formation. Mazlan, (1997) regrouped Layang Layangan Unit (lower part of Belait Formation according to Lee, 1977; see Table 1) with Temburong Formation.

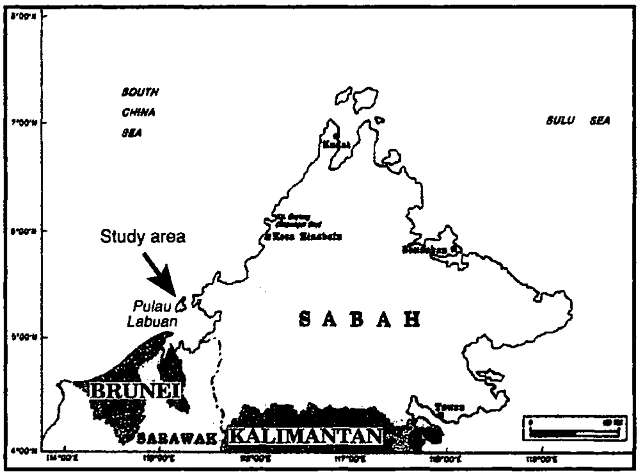

Figure 1. Location of Labuan Island.

Table 1. Description of selected samples analysed in this study (letters A, B, C, D and E show locations on Figure 2).

	Lithology	Formation (Lee, 1977)	Environment (Lee, 1977)	Location
Lab 12	Siltstone	Belait Formation	Fluvial Deposits	Tg. Kubong (E)
Lab 10	Coal			
Lab 1	Coaly Sandstone			Tg Layang Layangan (D)
Lab 37	Coaly Sandstone	Layang Layangan unit I	Tidal Flat Between Channels; Delta Slope	Jalan OKK Daud (C)
Lab 36	Shale			
Lab 33	Siltstone			
Lab 7	Coal			Tg. Layang Layangan (B)
Lab 4	Shale			
Lab21	Coal	Temburong Formation	Proximal Turbidite	Shell Crude Oil Thrminal (A)
Lab 22	Calcareous coal			

A major unconformity at the base of the Belait Formation can be correlated with the mainland onshore areas and is dated Early Miocene by Liechti *et al.* (1960). The marine sequences of Setap Shale and Temburong Formation beneath the unconformity are highly deformed whilst the fluvial to shallow marine deposits of Belait Formation are mildly deformed.

The deposits in Labuan are equivalent to some of the hydrocarbon reservoirs in the offshore areas of NW Sabah (Mazlan *et al.*, 1999).

METHODS

Ten samples were selected for this study (Fig. 2 and Table 1). The samples for measurements of vitrinite reflectance study were crushed and mounted in a slow-setting resin and then polished using progressively finer alumina (5/20, 3/50, and Gamma grades). Measurements of vitrinite reflectance were carried out using a Leitz DMRXP MPV reflected light microscope under oil immersion. A windows-based software package (MPV GEOR) was used to acquire data.

For extraction, about thirty grams of powdered samples were Soxhlet extracted for seventy-two hours using an azeotropic mixture of dichloromethane and methanol (93:7). A glass column (30 cm x 0.75 cm) was used for separating the extracted fraction into three fractions (saturated hydrocarbons, aromatic hydrocarbons and polar compounds "NSO"). The column, with the tip blocked with cotton wool, is packed with approximately 20 cm of activated silica gel prepared as a slurry with petroleum spirit. The silica was topped with alumina (2 cm) before 1 ml of sample extract (about 0.40 g) was directly poured into the column. The column then successively developed with 100 ml of light petroleum, dichloromethane, and methanol. The respective solvents for each fraction are then reduced by Butchi evaporation. The saturated fractions were further analysed by gas chromatography-mass spectrometry.

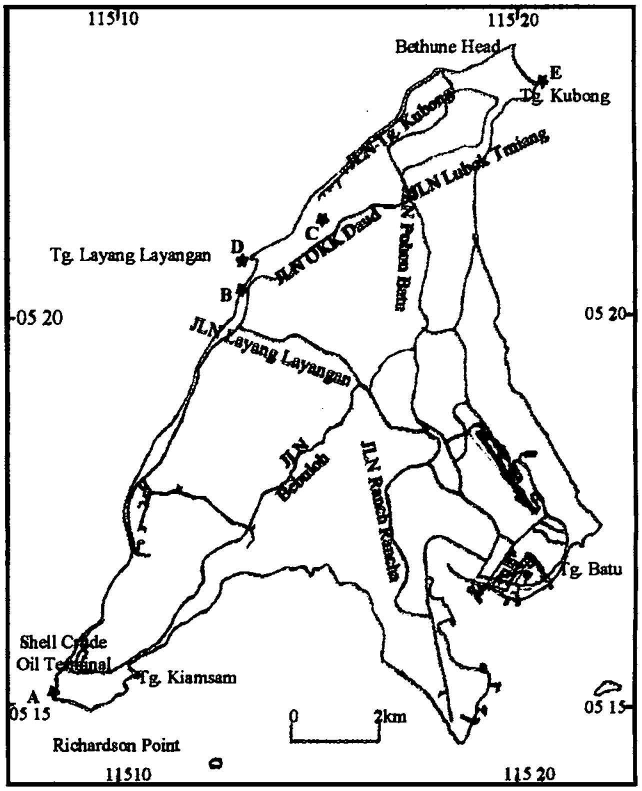

Figure 2. Locations of the studied samples.

Identification of biomarkers was performed by comparison of TIC retention time data and mass spectra with published data (Philp, 1985; Peters and Moldowan, 1993 and references therein). Relative peak intensities in gas chromatograms and mass fragmentograms were determined from their heights

RESULTS

Vitrinite reflectance data and molecular parameters, including isoprenoids, normal-alkane and triterpanes for studied samples, are presented in Table 2 and peaks identification shown in Table 3. Examples of biomarker distributions in these sediments are shown in Figure 3.

Temburong Formation

The Temburong Formation displays high abundance of low molecular weight n-alkanes compared to high molecular ones (Fig. 3). The Pr/Ph ratios are high and range from 2.20 to 2.75. The ratios of higher plant derived n-alkane to algae-derived n-alkanes (nC_{31}/nC_{17}) are from 0.42 to 0.86. Carbon preference index or CPI values are 1.09 in both samples studied from this formation. The Pr/nC_{17} and Ph/nC_{18} ratios are 0.31–0.68 and 0.11 to 0.31, respectively. In relation to tritepanes, Tm is higher than Ts with the Ts/(Ts+Tm) ratios of 0.41 – 0.32. The oleanane index is high in both samples (1.31–1.33). The $\beta\alpha/\alpha\beta$-C_{30} hopane ratios are low (0.11–0.13), whilst the hopane isomeration at C-22: 22S/(22S+22R) for C_{31} and C_{32} are high (0.62–0.63 and 0.57–0.59, respectively). Vitrinite reflectance values within the Temburong Formation range from 0.68% to 0.80%.

Layang Layangan Unit I

In contrast to Temburong Formation, the normal alkane distribution of the Layang Layangan Unit I is dominated by the n-alkanes in the range n-C_{13} to n-$C_{35\sim37}$ (Fig. 3). The maxima of the n-alkane distribution are at C_{31} and C_{14}. The Carbon Preference Index (CPI) values range from 1.43 to 1.81, show odd/even number predominance. Pr/Ph ratios range from 2.38 to 7.27 in this unit. The nC_{31}/nC_{17} ratios are greater than one in all samples. Pr/nC_{17} ratios range from 2.95 to 3.09. In relation to triterpanes, the ratios 22S/(22*R* + 22*S*) of Layang Layangan Unit I for C_{31} hopane

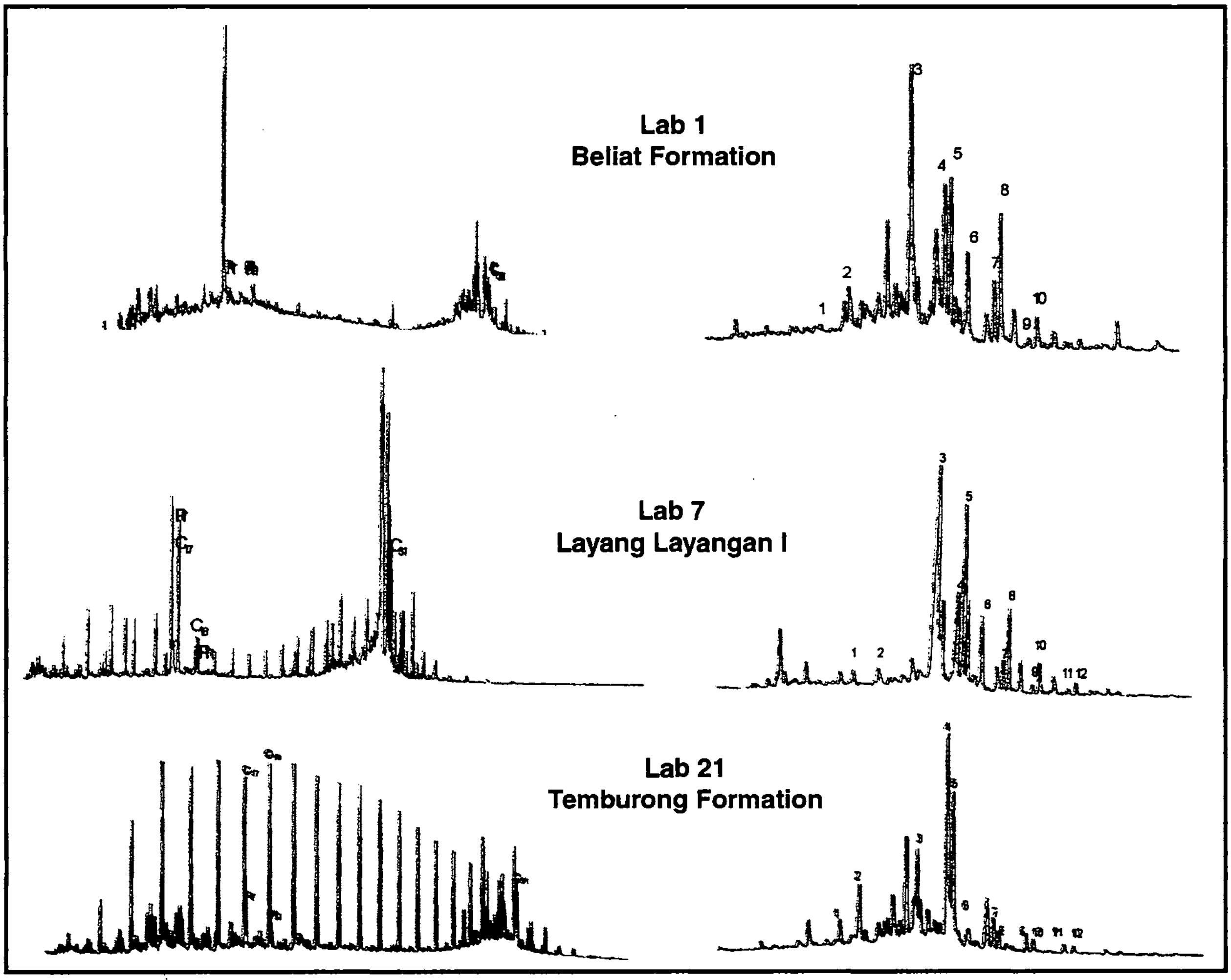

Figure 3. Biomarker distributions in Labuan sediments: n-alkanes and isoprenoids (left); distribution of triterpanes in the m/z 191 (right), peaks identification is shown in Table 3.

Table 2. Vitrinite reflectance data, normal-alkanes, isoprenoids and triterpanes parameters.

Sample number	*Lithology*	*Formation*	*nC_{31}/nC_{17}*	*CPI*	*Pr/Ph*	*Pr/nC_{17}*	*Ph/nC_{18}*	*$\frac{Ts}{(Ts+Tm)}$*	*$Ole./C_{30}H$*	*$C_{29}N/C_{30}H$*	*$Mor./C_{30}H$*	*$C_{31}\ \frac{S}{(S+R)}$*	*$C_{32}\ \frac{S}{(S+R)}$*	*$\%R_0$*
Lab12	Siltstone	Belait formation	1.19	1.43	5.67	2.43	0.43	0.47	0.95	1.98	0.43	0.21	0.38	
Lab10	Coal			1.42	7.50			0.05	0.12	0.47	0.58	0.35	0.25	0.48
Lab1	Coal Sandstone		2.13	2.33	1.63	0.81	1.60	0.17	0.94	1.74	0.60	0.34	0.27	0.49
Lab37	Coal Sandstone	Layang Layangan units		1.54	7.27			0.35	0.96	3.59	0.96	0.23	0.18	0.49
Lab36	Shale		3.32	1.62	3.29	2.95	0.89	0.28	0.80	2.16	0.60	0.35	0.42	
Lab33	Siltstone			1.43	2.38			0.32	0.85	1.90	0.40	0.39	0.40	0.52
Lab4	Shale		3.29	1.81	4.00	3.09	0.57		0.81	1.75	0.42	0.42	0.39	
Lab7	Coal			1.60	4.29			0.47	0.53	1.21	0.43	0.28	0.26	0.50
Lab21	Coal	Temburong Formation	0.42	1.09	2.75	0.31	0.11	0.32	1.31	0.58	0.13	0.62	0.59	0.68
Lab22	Calcareous Coal		0.86	1.09	2.20	0.68	0.31	0.41	1.33	0.59	0.11	0.63	0.57	0.80

CPI = $\{[(C_{25}\text{-}C_{33}$-odd No.)/($C_{24}\text{-}C_{32}$-even No.)] +[($C_{25}\text{-}C_{33}$-odd No.)/($C_{26}\text{-}C_{34}$-even No.)]\}/2 Bray and Evans (1961); Pr = Pristane; Ph = Phytane; $\%R_0$ = Vitrinite reflectance

ranges from 0.23 to 0.42 and from 0.18 to 0.42 for the C_{32} homologue. The ratios of moretane/C_{30} hopanes are high and ranging from 0.42 to 0.96 for this unit. The high abundance of Tm compared to Ts is reflected in Ts/(Ts+Tm) ratios, which range from 0.28 to 0.47. The oleanane index ranges from 0.53 to 0.96. Vitrinite reflectance ranges from 0.49% to 0.52%.

Belait Formation

The normal alkane distributions for the Belait Formation are shown in (Fig. 3). The TIC (total ion current) distributions show low abundance in n-alkane and show evidence of slight biodegradation based on the presence of humps of unresolved complex mixture. The CPI values are more than 1, ranging from 1.42 to 2.33. The Pr/Ph ratio ranges from 1.63 to 7.50. The Pr/nC_{17} ratio ranges from 0.81 to 2.43. The 22S/(22R + 22S) at C-22 for C_{31} and C_{32} hopane ratios for Belait Formation range from 0.21 to 0.35 for C_{31} and from 0.25 to 0.38 for C_{32}. The Ts/(Ts+Tm) ratios are from 0.17 to 0.47 and oleanane index range from 0.12 to 0.95. Vitrinite reflectance ranges from 0.48% to 0.49%.

DISCUSSION

Organic matter input and depositional environments

Temburong Formation

The high abundance of low molecular weight n-alkanes, compared to high molecular weight, suggest low contribution of terrigenous organic matter, which can be supported by low nC_{31}/nC_{17} ratios. The nC_{31} indicates higher plant inputs and nC_{17} show contribution of algae (Hunt, 1996; Tissot and Welte, 1984). The low Pr/nC_{17} and Ph/nC_{18} ratios (031–0.68 and 0.11–0.31, respectively) have been reported to suggest deposition in a deep marine environment, which generally possesses relatively lower amounts of acyclic isoprenoids compared to n-alkanes (Wan Hasiah, 1999).

Table 3. Identification of triterpanes in the m/z 191 mass fragmentograms.

Peak	Compound	Abbreviation
1	18α(H),21β(H)-Trisnorneohopane (C27)	Ts
2	17α(H),21β(H)-Trisnorhopane (C27)	Tm
3	17α(H),21β(H)-Norhopane (C29)	N
4	18α(H)-Oleanane (C30)	Ole.
5	17α(H),21β(H)-Hopane (C30)	H
6	Moretane (C30)	Mor.
7	22S-17α(H),21β(H)-Homohopane (C31)	
8	22R-17α(H),21β(H)-Homohopane (C31)	
9	22S-17α(H),21β(H)-Bishomohopane (C32)	
10	22R-17α(H),21β(H)-Bishomohopane (C32)	

Layan Layangan Unit I

The n-alkanes contained in the samples of this unit are dominantly high carbon number with an odd over even carbon preference. Their n-alkane maximums appear generally at C_{31}, indicative of terrestrial material input (Tissot and Welte, 1984; Hunt, 1996). The nC_{31}/nC_{17} ratios are very high in measured samples (>3). Pr/Ph ratios are variable from 2.38 to 7.27 and Pr/nC_{17} in measured samples are 2.95–3.09, suggesting an oxic depositional environment (Didyk *et al.*, 1978).

Belait Formation

The low abundance of n-alkanes made the measurements of these samples difficult. The high CPI value, high Pr/Ph ratios and nC_{31}/nC_{17} ratios, suggesting higher plant input deposited under oxic environment.

Thermal Maturity

Vitrinite Reflectance (VR) is the most commonly used organic maturation parameter (e.g. Hunt, 1996). The thermal maturity of samples studied shows a wide range, i.e. ranging from 0.48 to 0.80%. Based on vitrinite reflectance, the Temburong Formation is of relatively high maturity (0.68 to 0.80%). The biomarker data also supports this level of maturation. The Temburong Formation exhibits CPI values of about 1.09. CPI values close to 1.0 indicate complete maturation (Simoneit, 1978). In the hopane isomeration at C-22: 22S/(22S+22R), the 22S parameter is probably the most widely applied for maturity parameters, and records the relative abundance of the more thermally stable 22S isomer (e.g. Kolaczkowska *et al.*, 1990; Peters and Moldowan, 1993) compared to the biologically-derived 22R stereochemistry. These ratios for the Temburong Formation range from 0.62 to 0.63 C_{31} and for C_{32} from 0.57 to 0.59, and indicate at least moderate maturity.

Ts/(Ts+Tm) ranges from 0.32 to 0.41. This parameter is commonly used for maturity assessment, although it varies with the source of organic matter (Seifert and Moldowan, 1978). In the studied samples the source influence of organic matter appeared to be strong and effecting these ratios, therefore it could not be used as a maturation indicator here.

The moretane/C_{30} hopane ratios are low in all samples and range from 0.11 to 0.13. Moretanes are much less stable than hopanes, and thus decrease in concentration more rapidly with increasing maturity (e.g. Peters and Moldowan, 1993). All or most of the moretane loss occurs at very low maturities. Moretanes/hopane ratios, like 22S/(22S+22R) ratios for extended hopanes, are mainly useful as a qualitative indicator of immaturity (Grantham, 1986). This ratios is variable from about 0.8 in immature bitumens to values of less than 0.15 in mature source rocks and oils to minimum of 0.05 (Mackenzie *et al.*, 1980; Seirfert and Moldowan, 1980).

The oleanane indexes (oleanane/C_{30} hopane) for these samples are 1.31 and 1.33. Oleanane is thought to be a Cretaceous or younger higher-plant (probably angiosperm) marker (Ekweozor, 1988). The oleanane index tends to increase by destruction of hopanes. High maturity oils show a higher oleanane index than original organic matter in immature or low maturity level source rocks (Alberdiand and Lopez, 2000).

In contrast to Temburong Formation, vitrinite reflectances range from 0.48 to 0.52% for Layang Layangan unit I and Belait Formation, and indicate low maturity. CPI values range from 1.42 to 2.33, and also reflect also low maturity. In the hopanes, the low degree of thermal maturity is reflected in a low degree of conversion of the biologically produced 22R form for C_{31}–C_{33} 17α (H) hopanes (homohopanes) to their 22S epimer. The ratios 22S/(22*R* + 22*S)* of Layang Layangan Unit I for C_{31} range from 0.28 to 0.42 and from 0.18 to 0.42 for the C_{32} homologue. These ratios for the upper part of Belait Formation range from 0.21 to 0.35 for C_{31} and from 0.25 to 0.38 for C_{32}.

Moretane is thermally less stable than C_{30} hopane (e.g. Seirfert and Moldowan, 1980). The ratio of moretane/C_{30} hopane ranges from 0.42 to 0.96 for Layang Layangan Unit I and from 0.43 to 0.60 for Belait Formation. The range of moretane/C_{30} hopane ratios indicate the low thermal maturity of the organic matter (Mackenzie *et al.*, 1980; Seirfert and Moldowan, 1980).

The high abundance for Tm compared to Ts is reflected in Ts/(Ts+Tm) ratios, which range from 0.05 to 0.47 for both Layang Layangan Unit I and Belait Formation and may indicate low maturity. However, these ratios must be interpreted with considerable care as an indicator of thermal maturity, as it depends to a considerable degree on the conditions of sedimentation and the character of the source of organic matter (Peters and Moldowan, 1993).

CONCLUSIONS

The Pristane/Phytane and CPI ratios in all samples studied are greater than 1, which suggest a terrigenous origin and deposition under oxidizing environments. The nC_{31}/nC_{17} ratios for Temburong Formation are lower than 1, this may suggest some alga contribution during deposition of this sediments.

Vitrinite reflectance data indicate that the Layang Layangan Unit I and Belait Formation are immature (0.49–0.52 %Ro and 0.48 Ro%, respectively). Temburong Formation is relatively more mature (0.68 to 0.80%)

The variation in maturity is also observed in some organic geochemical parameters. The extremely low thermal maturity of Layang Layangan Unit I and Belait Formation is reflected in the CPI values which vary in the range from 1.42 to 2.33 and in the high ratios of moretane/C_{30} hopane. The low maturity for this unit can be seen in the hopane isomeration at C-22: 22S/(22S+22R) for C_{31} and C_{32} which range from 0.21 to 0.42 for C_{31} and for C_{32} from 0.18 to 0.42.

On the other hand, the Temburong Formation shows high maturity. The high maturity is reflected in the CPI values i.e. from 1.09 and the moretane/C_{30} hopane ratios from 0.11 to 0.33.

The hopane isomeration at C-22: 22S/(22S+22R) for C_{31} and C_{32} also show high thermal maturity, they range from 0.62 to 0.63 and from 0.57 to 0.59 for C_{31} and C_{32} respectively. The oleanane/C_{30} hopane ratios (oleanane index) are high.

Thus, base on organic geochemical parameters and supported by the vitrinite reflectances data, the Layan Layangan Unit I and Belait Formation are thermally immature and Temburong Formation are mature. This thermal maturity suggests the Layang Layangan units be grouped within the Belait Formation.

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