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Geochemical Variations in Hydrocarbon Components Distribution in a Prograding Deltaic Sequence: A Case Study of the Baram Delta, Offshore Sarawak Basin, Malaysia

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Abstract: Studies on hydrocarbon distribution have evolved from basic reservoir characterization to complex studies today involving the interactions between oil components and clay minerals and sequential extraction studies on hydrocarbon extracts in reservoir rocks. Findings from such studies include the discovery of variations in oil fractions in reservoirs such as adsorbed oil and free oil. The theory that first oil charge preferentially interacts with clay minerals occurring in pores and as coatings in reservoirs was also proposed by some researchers. Despite, all these studies some aspects of variations in the composition of hydrocarbons in reservoir rocks still need to be investigated further. This study has been carried out particularly because the qualitative and quantitative composition of aromatic and aliphatic components of hydrocarbons in terms of the presence and quantities of hydrocarbon functional groups and how they relate to hydrocarbon migration have not been exhaustively discussed. This study uses Ultra-Violet visible light (UV-vis) and Fourier Transform Infra Red (FTIR) to characterize variations in hydrocarbon distribution in reservoir quality sandstones from three fields namely BD01, BD02 and BD03 in the Baram Delta, offshore Sarawak and to deduce how these variations relate to differential migration patterns in hydrocarbons. Hydrocarbon extraction was done in the ultra vilolet visible (UV-vis) experiment using 0.1M sodium pyrophosphate as solvent whereas in the Fourier Transform Infra Red (FTIR), the experiment was done on very fine powdered samples of the sandstones. Results from both the UV-vis and FTIR experiments indicate a dominance of aromatic functional groups in the samples. Most of the samples have E4/E6 ratio of more than 1 which indicates a high degree of aromacity. The BD01 field sandstones with a maximum porosity of 32% has the highest average E4/E6 ratio of 1.21, followed by the BD02 field sandstones with a maximum porosity of 29% and average E4/E6 ratio of 1.19 and the BD03 field with a maximum porosity of 20% and an average E4/E6 ratio of 1.09. The dominance of aromatics in the samples is interpreted as a possible indication of episodes of migration of aliphatics in the past leaving the aromatics behind with the variations in E4/E6 ratio reflecting the micro heterogeneities in the samples.

Keywords: Aliphatic, aromatic, fulvic acid, functional group, heterogeneity, humic acid, hydrocarbon components, migration, porosity

INTRODUCTION

The Baram Delta is one of the seven geological provinces found offshore the Sarawak Basin and is the most prolific of all the geological provinces in the basin (Tan *et al.*, 1999) (Fig. 1). As at 1.1.1998, a total of 132 exploration wells, 46 wildcats and 86 appraisal wells, have been drilled, representing about 30% of the total wells drilled in Sarawak (Tan *et al.*, 1999). The Baram Delta consists of nine fields. The area which was discovered in 1969 is estimated to have more than 400 million stock barrels of oil in place with multiple stacked sandstone reservoirs in a shallow offshore environment (Surdiman *et al.*, 2007). The Baram Delta was formed on an active continental margin with its shape and size suggesting that it may have developed initially as a pull apart basin whose length and width

were pre-determined by its bounding faults (Tan et al., 1999).

The offshore stratigraphy of the Baram Delta (BD) is characterized by the occurrence of coastal to coastal-fluviomarine sands which have been deposited in a northwestwards prograding delta since the Middle Miocene (from Cycle IV onwards) with the Cycle V (Middle to Upper Miocene) to Cycle VII (Upper Pliocene) being well developed (Fig. 2) (Ho, 1978; Tan *et al.*, 1999; Hutchinson, 2005).

Several studies have been conducted over the years in trying to decipher the variations of hydrocarbon components and their interactions with reservoir rocks (Brooks, 1981; Waples, 1985; Wilhelms *et al.*, 1996; Schwark *et al.*, 1997; Pan and Yang, 2000; Pan *et al.*, 2005). Findings from such studies include the discovery

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Fig. 1: Location map of the Sarawak basin showing the geological provinces with well density (after Othman Ali and Salahuddin, 1999)



Fig. 2: Generalised stratigraphic columns for the onshore structural provinces of Sarawak modified from the Geological Survey of Malaysia (1995)

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Table 1: Sam	ple ID, depth and g	eneral description	of samples from BD01 field		
Sample ID	Depth (m), from	Depth (m), to	Sample description		
S1	1478.76	1479.47	Fine grained sandstone; very pale orange (10YR8/2); presence of vertical burrows; well indurated; massive structured; well sorted grains		
S2	1487.52	1488.31	Fine grained sandstone; very pale orange (10YR8/2); massive; well sorted; trace fossils		
S3	1491.51	1492.35	Fine grained sandstone; very pale orange (10YR8/2); massive structured; friable; well sorted		
S4	1494.84	1495.65	Fine grained sandstone; very pale orange (10YR8/2); massive structured; well sorted grains		
S5	1502.61	1503.27	Fine grained sandstone; grayish yellow (5Y8/4); parallel lamination (0.5-1 cm); laminations of coal intercalate (0.5-1 mm) with sandstone; well sorted		
S6	1506.73	1507.54	Fine grained sandstone; very pale orange (10YR8/2); horizontal burrows (3 cm length, 3 mm width); well indurated; well sorted; fossilferous		
S7	1507.54	1508.25	Fine grained sandstone; very pale orange (10YR8/2); well sorted; well indurated; massive		
Table 2: Sample ID, depth and general description of samples from BD02 field					
Sample ID	Depth (m), from	Depth (m), to	Sample description		
L1	1222.55	1223.47	Medium grained sandstone; dark yellowish orange (10YR6/6); massive structured; friable; hydrocarbon odour		
L2	1661.77	1662.68	Fine grained sandstone; grayish orange (10YR7/4); massive structured; well indurated		
L3	1682.19	1683.11	Fine grained sandstone; dark yellowish orange (10YR6/6); massive structured; friable; well sorted		
L4	1692.02	1692.86	Fine grained sandstone; dark yellowish orange (10YR6/6); massive structured; friable; well sorted		
L5	2101.50	2102.45	Fine grained sandstone; pale yellowish brown (10YR6/2); massive structured; well indurated		
L6	2109.52	2110.53	Fine grained sandstone; grayish orange (10YR7/4); massive structured; friable; well sorted		
L7	2122.89	2123.86	Fine grained sandstone; gravish orange (10YR7/4); massive structured; friable; well sorted		
Table 3: Sam	ples ID, depth and	general description	of samples for the BD03 field		
Sample ID	Depth (m), from	Depth (m), to	Sample description		
B1	851.31	851.89	Fine grained sandstone; dark yellowish orange (10YR6/6); parallel lamination (1-2 mm thick); friable; hydrocarbon/benzene odour		
B2	889.59	890.41	Fine grained sandstone; dark yellowish orange (10YR6/6); massive structured; well indurated		
B3	890.41	891.00	Fine grained sandstone; moderate yellowish brown (10YR5/4); friable		
B4	892.79	893.39	Fine grained sandstone; grayish yellow (5Y8/4); friable; massive structured		

of variations in oil fractions in reservoirs such as adsorbed oil on mineral surfaces, adsorbed oil on clay minerals, free oil and the mechanism of oil-clay minerals interaction in reservoir rocks. This mechanism suggests that first oil charge preferentially interacts with clay minerals occurring in pores and as coatings in reservoirs. These studies were necessitated by the need to know how the composition of hydrocarbons and reservoir rocks affect the distribution of hydrocarbons in reservoir pore networks and the interactions between the oil components and mineral surfaces in reservoir rocks. Despite, all these studies consistently producing very interesting results, some aspects of variations in the composition of hydrocarbons in reservoir rocks still need to be investigated further. Particularly, the qualitative and quantitative composition of the aromatic and aliphatic components (functional groups) of hydrocarbons in terms of the presence and quantities of hydrocarbon functional groups and how they relate to hydrocarbon migration have not been exhaustively discussed. The presence of these functional groups are usually expressed in the form of peaks at specific wavelengths in infrared spectrometry and their relative amounts expressed as peak intensities. The relationship between reservoir properties such as porosity and ratio of humic to fluvic components (E4/E6 ratio) in the reservoir sandstones has also not been sufficiently discussed in literature (Evdokimov and Losev, 2007; Ramli and Padmanabhan, 2011). Therefore the objective of this study is to characterize variations in hydro carbon distribution in reservoir quality sands tones from three fields in the Baram Delta, offshore Sarawak and to evaluate the possible causes of these variations. For the purpose of this study, the three fields will be referred to as BDO1, BDO2 and BDO3, respectively.

MATERIALS AND METHODS

Seven samples (S1-S7) were collected from reservoir quality rocks in field BD01, seven samples

(L1-L7) from field BD02 and four samples (B1-B4) from field BD03 located in the Baram Delta province. They belong to the cycles V and VI Middle to Upper Miocene prograding deltaic to shallow marine sandstones (Tan *et al.*, 1999). These samples have been described in Table 1 to 3. The color of the samples have been described according to the Munsell color chart.

About 0.1 g of fine powdered sample was analyzed using the Fourier Transform Infrared (FTIR) spectroscopy 8400S (Shimadzu FTIR Spectrophotometer) to determine the hydrocarbon functional groups and the results interpreted according to Coates (2000) and Stuart (2004). About 0.1 g of the sample was treated with 0.1 M sodium pyrophospate to extract the hydrocarbons and the extract was subsequently analyzed using a Shimadzu UV-3150 UV Vis Spectrophotometer and interpreted according to Schnitzer and Neyroud (1975), Stevenson (1982) and Ben-Awuah and Padmanabhan (2013).

RESULTS AND DISCUSSION

Distribution of functional groups: All the samples show maximum peak intensity around 990 cm⁻¹. This represents the aromatic C-H in plane bend functional group which is detected in all the samples except L1. The curves in field BD03 are closely spaced whereas the curves in field BD01 and BD02 are significantly spaced. In field BD01, the curve for S3 is distinctively spaced from all the other samples resulting in very high peak intensities of hydrocarbon functional groups. Most of the peaks are concentrated between the wavelengths 690-1700, 2300-3000 and 3200-3900 cm⁻¹ in all the samples (Fig. 3 to 5).

In field BD01, the presence of organic siloxane (Si-O-Si), aromatic C-H in plane bend, aryl-H (aromatic hydrogen bond), aromatic aryl ring, aromatic C-H out of plane bend was detected in all samples. Methyl C-H





Fig. 3: FTIR spectra of field BD01



Fig. 4: FTIR spectra of field BD02



Fig. 5: FTIR spectra of field BD03

Table 4: Fu	nctional groups in field	d BD01						
Sample	P 011	Methyl C-H	Methyl ether		Carbonyl ($C = O$)	Alkenyl $C = C$ stretch	C = C stretching, aromatic	Methylene C-H
ID	Free OH	asym/sym	(O-CH ₃)	B-H stretch	ketone	(olefinic alkene)	ring (aryl) stretch	bend
SI	3300-3600	2945	2839	-	-	-	-	-
S2	3600-3800	2920	2851	-	1746	1654	1597	1466
S3	3300-3800	2926	2874	-	-	-	-	1487
S4	3300-3900	2943	-	2345	-	-	-	-
S5	3300-3900	-	2846	-	-	-	-	1486
S6	3400-3900	2936	-	2366	-	-	-	-
S7	3500-3900	2926	-	-	-	-	-	-
Sample	Vinyl C-H in	Methyne		Organic siloxane	Aromatic; C-H bending	Aryl-H; aromatic		Aromatic C-H out
ID	place bend	CH-	P-O stretch	(Si-O-Si) /silicone	(in plane bending)	hydrogen bond	Aromatic ring; aryl	of plane bend
S1	-	-	1163	1091, 1030	1009	914	779, 797	693
S2	1418	1278	-	1113, 1031	1008	913	778, 796	691
S3	1416	1265	1184	1093, 1030	1008, 937	913	796, 779	694
S4	-	-	-	1031	1009, 939	914	797, 779	693
S5	-	-	1163	1030	1009, 940	912	797, 778	693
S6	-	-	1164	1030	1008	913	796, 778	692
S7	1429	-	1163	1031	1009	913	797, 777	693
Table 5: Fu	nctional groups in field	d BD02						
	8 F	Methyl C-H	Methyl ether		Carbonyl ($C = O$)	Alkenvl $C = C$ stretch	C = C stretching aromatic	Methylene C-H
Sample ID	Free OH	asym/sym	(O-CH ₂)	B-H stretch	ketone.	(olefinic alkene)	ring (aryl) stretch	bend
I 1	3300-3900	2050	2882	2322	Retone	(oternine untenie)	ing (alf) succes	John
1.2	3300-3700	2004	2002	2322	1722	-	-	-
L2 1.2	2200 2800	2904	2001	-	1722	-	-	-
L3	3200-3800	2942	2833	2550, 2552	-	10/4	-	-
L4	3200-3800	2912	-	-	-	-	1603	-
L5	3300-3900	2917	2850	-	-	-	1618	-
L6	3500-3800	2950	2877	2366	-	-	1625	-
L7	3300-3900	-	-	2346	-	-	-	-
	Vinyl C-H in			Organic siloxane	Aromatic; C-H bending	Aryl-H; aromatic		Aromatic C-H out
Sample ID	place bend	Methyne CH-	P-O stretch	(Si-O-Si) /silicone	(in plane bending)	hydrogen bond	Aromatic ring; aryl	of plane bend
L1	1415	-	-	1029	-	911	795, 779	696
L2	1418	-	1163	1030	1007, 936	912	797, 778	693
L3	1432	-	-	1086, 1031	1008, 942	912	797, 778	694
L4	1418	-	-	1030	1007, 936	912	795, 778	692
L5	1424	1282	-	1028	1006	912	797, 778	694, 670
L6	1437, 1414	1267	1162	1031	1007, 935	912	797, 777	693
L7	-	-	-	1029	1008	914	797, 777	693
Table 6: Fu	nctional groups in field	d BD03						
	6 1	Methyl C-H	Methyl ether			Alkenvl $C = C$ stretch	C = C stretching aromatic	Methylene C-H
Sample ID	Free OH	asym/sym	(O-CH ₂)	B-H stretch	Nitrate (N-O) stretch	(olefinic alkene)	ring (aryl) stretch	bend
D1	2400 2000	2002	(0 CH3)	2207	Tituate (IT O) succes	(oternine arkene)	ring (aryi) stretten	bend
D1 D2	2500 2000	2903	-	2397	- 870	-	-	-
D2	2600 2800	-	2030	2500	0/7 921	-	-	-
B3	3000-3800	-	-	-	851	-	-	-
<u>B4</u>	3300-3900	2955	28/5	-	-	1009	-	150/
	Vinyl C-H in place			Organic siloxane	Aromatic; C-H bending	Aryl-H; aromatic		Aromatic C-H out
Sample ID	bend	Methyne CH-	P-O stretch	(Si-O-Si) /silicone	(in plane bending)	hydrogen bond	Aromatic ring; aryl	of plane bend
B1	1414	-	1164	1031	1007	913	797, 776	693
B2	1418	-	-	1030	1007	913	797, 778	694
B3	-	-	1163	1029	1008, 987	911	796, 777	693
B4	-	-	-	1026, 1095	1008	-	795, 777	692

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functional group was detected in all samples except S5. Methyl ether (O-CH₃) was detected in all the samples except S4, S6 and S7. The carbonyl (C = O), alkenyl C = C stretch (olefinic alkene) and C = C stretch aromatic ring functional groups are detected in only S4. The presence of the methyne CH-, vinyl C-H in place bend and methylene C-H bend functional groups were detected in S2 and S3, S2, S3 and S7 and S2, S3 and S5, respectively.

In field BD02, the presence of organic siloxane (Si-O-Si), aryl-H (aromatic hydrogen bond), aromatic aryl ring, aromatic C-H out of plane bend was detected in all samples. Methyl C-H functional group was detected in all samples except L7. Methyl ether (O-CH₃) was detected in all samples except L4 and L7. The carbonyl (C = O) and alkenyl C = C stretch (olefinic alkene) functional groups were detected in L2 and L3 respectively. The presence of C = C stretch aromatic ring functional group was detected in L4, L5 and L6. The presence of the methyne CH- and vinyl C-H in place bend functional groups were detected in L5 and

L6 and L1-L6, respectively. Methylene C-H bend functional group was not detected in any of the samples.

In field BD03, the presence of organic siloxane (Si-O-Si), aromatic C-H in plane bend, aromatic aryl ring, aromatic C-Hout of plane bend were detected in all samples. Aryl-H (aromatic hydrogen bond) was detected in B1-B3. Methyl C-H functional group was detected in B1 and B4. Methyl ether (O-CH₃) and alkenyl C = C stretch (olefinic alkene) functional groups were detected in B2 and B4 respectively. C = C stretch aromatic ring and methyne functional groups were absent in all samples. The presence of the methylene C-H and vinyl C-H in place bend functional groups were detected in B4 and B1 and B2, respectively.

Table 4 to 6 summarizes the different functional groups in all the samples. The numbers in the tables represent the wavelength at which the particular functional groups are determined in the sample. These variations can be seen within the same facies and

, aromatic Methylene C-H bend - 84.7 120.7
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84.7 120.7
84.7 120.7
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Aromatic C-H
ryl out of plane bend
96.1
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124.9
97.2
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between similar facies in the same field and between different fields. The FTIR confirms the dominance of aromatic functional groups in the samples as suggested by the E4/E6 ratio from the UV-vis experiment. In cases where the same peaks were identified, differences peak intensities reflecting differences in in transmittance were evident (Fig. 3 to 5). Table 7 to 9 gives the transmittances in percentage of each functional group. The differences in transmittance according to Coates (2000) and Stuart (2004) is an indication of differences in the quantity of that particular functional group. Higher peak intensities represents higher amounts. From Fig. 3 to 5, the most distinct peak in all the samples except L1 occurs between 1007-1009 cm⁻¹. This is the peak for aromatic C-H in plane bend functional group. This peak has the highest peak intensity in all the samples. The presence of free-OH functional group is detected in all the samples between 3200-3900/cm. This functional group indicates the presence of either water or minerals containing oxides. Transmittances of the same hydrocarbon functional group varies within similar

lithologies and within the same field. This indicates variations in hydrocarbon distribution within similar facies. These variations also emphasize the importance of micro heterogeneties in rocks which influences the distribution of hydrocarbons.

Evaluation of aromacity: The focus of the UV-vis experiment is the E4/E6 ratio. This is the ratio of optical densities or absorbencies of humic and fulvic acid solutionsat 465 and 665 nm and is used mainly for the characterization of organic matter (Schnitzer and Khan, 1978). The E4/E6 ratio is also widely used as a measure of aromacity or humification (Stevenson, 1982). From Table 10, the general trend of the E4/E6 ratio in most of the samples is a ratio greater than 1 except in samples S5 and B3. This general trend indicates the dominance of aromatics in reservoir quality sandstones in the three fields and by extension the Baram Delta.

Field BD01 with a maximum porosity of 32% has the highest average E4/E6 ratio of 1.21, followed by field BD01 with the maximum porosity of 29% and

Table 10: Average porosity and average E4/E6 ratios	
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Field ID	Average porosity	Average E4/E6 ratio
Field BD01	32	1.21
Field BD02	29	1.19
Field BD03	20	1.09

Table 11: E4/E6	ratios		
Sample ID	E4 (absorption at 465 nm)	E6 (absorption at 665 nm)	E4/E6
S1	1.08	0.83	1.30
S2	2.44	2.23	1.09
S3	2.59	2.08	1.24
S4	1.79	1.36	1.32
S5	1.09	1.32	0.83
S6	0.92	0.71	1.30
S7	0.92	0.71	1.30
B1	1.29	1.16	1.11
B2	1.86	1.61	1.16
B3	0.99	1.06	0.93
B4	2.42	2.07	1.17
L1	1.39	1.25	1.11
L2	2.31	2.01	1.15
L3	1.58	1.41	1.12
L4	1.23	1.09	1.33
L5	1.58	1.25	1.27
L6	1.29	1.05	1.23
L7	1.56	1.34	1.16

average E4/E6 ratio of 1.19 and field BD03 with a maximum porosity of 20% has an average E4/E6 ratio of 1.09 (Table 10). This means field BD01 with the highest porosity allows a lot more migration of the aliphatics and retention of the aromatics resulting in a relatively higher average E4/E6 ratio. Evidence of past episodes of migration of aliphatics and retention of aromatics is reflected in the high values of E4/E6 ratio. The lowest E4/E6 ratio of 0.83 measured in S5 can be attribute to the traces of coal laminations present in this sample. The presence of extra Carbon from the coal has a net effect of altering the carbon-carbon bonds in the hydrocarbons in this sample.

Despite the general trend of E4/E6 ratio of more than 1 in most of the samples, the minor variations in the values is a reflection of the micro (small scale) heterogeneities in the sandstones which will subsequently result in different rates of migration (Mikes and Bruining, 2005; Ramli and Padmanabhan, 2011) (Table 11).

Variations in hydrocarbon components: Variations in hydrocarbon components (aromatics and aliphatics) may be from several factors but most importantly usually include biodegradation, different source rocks for the hydrocarbons and differences in migration patterns of hydrocarbons. In the Baram Delta, the first two possibilities can be cautiously eliminated based on work by other researchers such as Tan et al. (1999), who proposed through gas chromatography, oil-oil correlation and oil-source correlation the very similar nature of the source rocks that generated the hydrocarbons and a negligible degree of biodegradation of organic matter in the Baram Delta. According to Tan et al. (1999), gas chromatography-mass spectrometry confirms the terrigenous nature of the source rocks by displaying ubiquitous occurrence of bicadines and oleanane in the tritepane m/z 191 traces, low tricyclic

terpanes, moderately abundant diasteranes, predominance of C_{29} regular steranes and high hopane to sterane ratios. This means that the variations in hydrocarbon functional groups distribution is not as a result of differences in source rocks or differences in the degree of biodegradation. It can therefore be attributed to differences in migration patterns and rates.

The dominance of aromatics in the samples therefore can be attributed to episodes of migration of aliphatics in the past. This finding is in agreement with the findings of Ramli and Padmanabhan (2011). Aliphatics are generally lighter than aromatics and so migrate more easily. This property of aliphatics coupled with the relatively good porosity in the sandstones and the large pore throats means that the aliphatics are more susceptible to migration compared to the aromatics. Results from both the UV-vis and FTIR supports the preferential migration of aliphatics and retention of aromatics in the Baram Delta. The high E4/E6 ratio confirms the high aromacity of the samples and retention of aromatics. Distribution of hydrocarbon functional groups can therefore be used as indicators of migration in reservoir sandstones in petroleum exploration.

CONCLUSION

Variations in the presence of hydrocarbon functional groups and in the peak intensities of these functional groups within similar facies in the same field and within similar facies in different fields were observed. Both the UV-vis and FTIR results indicate a dominance of aromatic functional groups. Differences in transmittance in the same functional groups within similar facies in the same field were observed. These variations can be attributed to past episodes of migration where most of the aliphatics migrated out of the sandstones due to the relative ease with which aliphatics migrate compared to aromatics and the relatively higher porosity of these sandstones. The retention of the aromatics has resulted in a generally higher E4/E6 ratio greater than 1. The variations in the E4/E6 values is a reflection of the micro heterogeneities in the sandstones which will subsequently result in different rates of migration. Field BD01 which has the highest maximum porosity of 32% and the biggest average pore throat also has the highest average E4/E6 ratio of 1.2 confirming the relationship between porosity, pore throat sizes and migration of hydrocarbons.

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