# Mineralogy and Chemical Composition of the Clay Fraction of Neogene Shales from the Surma Group in the Bengal Basin, Bangladesh

M. JULLEH JALALUR RAHMAN<sup>1\*</sup>, RUMANA YEASMIN<sup>1</sup> and TOM MCCANN<sup>2</sup>

<sup>1</sup>Department of Geological Sciences, Jahangirnagar University, Savar, Dhaka-1342, Bangladesh <sup>2</sup> Institute of Geology, University of Bonn, Germany \*Email: jrahman437@gmail.com

Abstract: Mineralogical and chemical investigations (<2 $\mu$ m clay separates) of shale samples from the Neogene-age Surma Group obtained from four wells (Habiganj-11, Shahbazpur-1, Titas-11, Titas-15) in the Bengal basin, Bangladesh, were carried out in order to reveal the clay mineral composition as reservoir exploration and exploitation requires a good understanding of the clay minerals. The samples were analyzed by X-ray diffraction (XRD), scanning electron microscope (SEM) and X-Ray fluorescence spectrometry (XRF). Mineralogically, the sub-surface Surma Group shales comprise predominantly quartz, plagioclase, illite, chlorite, kaolinite, with lesser amounts of K-feldspar, dolomite and smectite, and minor to trace amounts of calcite, siderite and pyrite. The chemical composition of the <2  $\mu$ m clay separates also suggests an illite and chlorite-rich composition. With increasing burial depth, the Surma Group shales are enriched in illite. The gradual decreasing of the smectite clays with depth and ultimate disappearance at greater depths ( $\geq$  3000 m) may have been responsible for the presence of the diagenetic illite. Based on the mineralogical composition it is most likely that the illite-chlorite associations together with quartz and feldspar were predominantly detrital in origin and thus reflect the presence of a rapidly-rising source terrain not subjected to intense weathering.

Keywords: Clay Minerals, Diagenesis, Neogene shales, Surma Group, Bengal basin, Bangladesh.

#### INTRODUCTION

The Bengal basin, one of the most potential hydrocarbonbearing sedimentary basins in the world, occupies Bangladesh and parts of West Bengal, Assam and Tripura in India (Fig.1). The basin fill, predominantly shallowmarine to continental clastics and minor carbonates, attains a thickness of c. 18 km. The succession is Tertiary in age, with 6 km of Neogene sediments present in the center of the basin (Hiller and Elahi, 1984).

The Neogene-age Surma Group in Bangladesh is the focus of exploration activity. This is largely due to the presence of deltaic to shallow marine sediments, which results in a succession comprising reservoir sandstones alternating with sealing shales. These units comprise the known petroleum deposits in Bangladesh (Table 1). Although the Surma Group is typically composed of subequal proportions of alternating shales and sandstones (Fig. 2), the unit is characterized by the predominance of shale in many sections. Over the last decades, a considerable geological, geophysical database has been established as a result of extensive hydrocarbon exploration. Much of the available geological data is related to organic geochemistry and hydrocarbon source rock potential. Very few works exist on clay minerals (e.g. Imam, 1989, 1994). A good understanding of the clay minerals in the reservoirs is necessary for effective and efficient reservoir exploration and exploitation.

The aim of the present study is to evaluate the mineralogy and chemical composition of the clay fraction of Neogene shales from the Surma Group of the core material collected from the Habiganj-11 (depth range 3088-3095 m), Shahbazpur-1 (depth range 997-3407 m), Titas-11 (depth range 2318-2790 m) and Titas-15 (depth range 2661-2720.6 m) wells in the Bengal Basin (Fig. 1). This study adds new results to earlier studies (Imam, 1989, 1994) from different wells.

#### **GEOLOGICAL SETTING**

The Bengal basin is a foreland basin which evolved as a result of the collision of three plates – the Indian, Tibetan (Eurasian) and Burma (West Burma Block) (Alam et al.,



Fig.1. Major tectonic elements of the Bengal Basin (Alam et al., 2003); the map also shows the locations of the petroleum exploration wells from which shale samples studied. HB-Habiganj, SB-Shahbazpur, TT-Titas, PT-Patharia.

2003) resulting in Himalaya and Indo-Burman ranges and thereby loading lithosphere to form flanking sedimentary basin (Uddin and Lundberg, 1998). The basin is bordered by the Himalayas and the Shillong massif to the north, Indian shield to the west and the Indo-Burman ranges to the east (Fig. 1). Towards the south, the basin is open to the Bay of Bengal. The geological evolution of the basin began in the late Mesozoic with the breakup of Gondwana and is ongoing (Alam, 1989). The Bengal basin, containing mainly shallow marine to continental clastic sedimentary rocks (mostly

 Table 1. Tertiary succession of the Bengal Basin, Bangladesh, modified after Imam and Shaw (1987)

Age	Group/ Formation	Lithology	Environment of deposition
Pliocene	Tipam Group	Massive to cross- bedded sandstones. Minor shale and clay	Fluvial
Miocene	Surma Group Boka Bil Fm. Bhuban Fm.	Alternating sandstone, shale and siltstone	Deltaic- shallow marine
		Unconformity	
Oligocene	Barail Group	Sandstone, shale, coal	Deltaic- shallow marine
Eocene	Jaintia Group	Limestone, Sandstone, shale	Open marine



Fig.2. Lithofacies of the Surma Group encountered in Shahbazpur well-1, TT-11 (Titas-11) and TT-15 (Titas-15) wells

sandstone and shale of Tertiary age) and minor carbonates, forming a thick ( $\pm 22$  km) early Cretaceous to Holocene sedimentary succession (cf. Alam et al., 2003). The depositional setting has been predominantly deltaic since the Oligocene (Curray and Moore, 1974).

The Miocene-age Surma Group sediments are mainly deltaic to shallow marine in origin (Johnson and Alam, 1991) and comprise of two formations, the basal Bhuban Formation and the overlying Boka Bil Formation (Table 1). The lithologies are mainly mudstones and quartzofeldspathicquartzolithic sandstones. Shale-rich units display a spectrum of rock types, including, interbedded shale, mudstone, sandstreaked shale, and shale with sandy/silty lenses (lenticularbedding).

#### **METHODS**

X-ray diffraction analysis of the Surma Group shales was carried out in order to identify the clay minerals present by BRUKER-AXS D8 ADVANCE with CuK $\alpha$  radiation at 40 kV and 20 mA at the University of Bonn, Germany. Clay mineral fractions of less than 2mm were obtained by centrifuging clay slurries. The mineralogical compositions of some shale samples from the Titas well were determined by X-ray diffraction analysis using the RIGAKU: ULTIMA- IV (185mm) Goniometer (Cu / 40KV / 40 mA) with a scanning speed of  $2^{\circ}2\theta$  / min at the BAPEX Laboratory, Bangladesh. Clay mineral fractions of <2 µm were obtained by the sedimentation method. The samples were air-dried, ethylene glycolated and heated (550°C).

A Cam Scan MV 2300 scanning electron microscope (SEM) fitted with an energy dispersive X-ray spectrometer (EDX) at the University of Bonn, Germany was used to observe the nature of the clay minerals. The samples were gold coated and examined under an acceleration voltage of 20 kV and a beam current of 33 micro A.

Chemical analysis of 2-micron clay separates was performed by PAN analytical's Sequential Wave-length Xray fluorescence spectrometer (WDXRF - AXIOS PW4400/ 24 with 3 kW Rh-anode X-ray tube at the University of Bonn, Germany. For major element determinations, fused pellets were prepared using a mixture of 0.6 g of the powdered sample and 3.6 g of alkali metal borate at a ratio of 1:6. The loss on ignition was determined after heating the powdered sample (3 to 5 g) to a temperature of 1100°C for 2 hours. The major element program from fused pellets was controlled by GeoPT laboratory tests.

The mineralogical composition of the shales was identified after Brindley and Brown (1980), JCPDS (1986), and, Moore and Reynolds (1997). Semi-quantitative estimation of the bulk minerals was made after Schultz (1964). The quantification of the clay minerals was made by considering the peak height above the background for each diagnostic reflection multiplied by a correction factor, for example 1 for illite, 0.54 for chlorite 0.5 for kaolinite and 0.35 for smectite. In the case of chlorite and kaolinite, the intensity at 7Å was split by taking the intensity relation at 3.52 Å for chlorite and at 3.57 Å for kaolinite.

## RESULTS

### **Bulk Minerals**

The Surma Group whole rock shale samples comprise of mainly clay minerals, quartz, plagioclase and potassiumfeldspars, as well as minor amounts of carbonates and traces of pyrite.

In all the samples quartz is the most abundant mineral among the non-clay minerals. The average quartz content is  $\sim$ 32 wt% with values varying from  $\sim$ 25 to  $\sim$ 42 wt %. Plagioclase is more common ( $\sim$ 6-12 wt%) than potassium-feldspar (trace to  $\sim$ 4 wt%). Dolomite is the predominant mineral phase among the carbonate minerals (average content of 3 wt%) and is identified by the 2.89 Å reflections. Calcite (av.  $\sim$ 0.4 wt%) and siderite (av.  $\sim$ 1 wt%) are present in minor amounts. Clay minerals constitute  $\sim$ 45-61 wt% of the bulk minerals.

#### **Clay Minerals**

The principal clay minerals found in the Surma Group shales are illite, chlorite, kaolinite and minor smectite. The semi-quantitative estimation of the clay minerals from the  $<2 \mu m$  size fraction with depth of burial is shown in Table 2. The characteristic X-ray diffractogram pattern of the samples is shown in Fig. 3.

Illite is the most abundant mineral in the shales and varies from ~44wt% (depth 2662.3 m, Titas-15) to ~87.9wt% (depth 3095.5 m, Habiganj-11) (Fig. 4). The percentage of illite remains constant at greater depths (2318.3 m onward). As revealed by SEM (Fig. 5A), the illite is characterized by cornflake and crenulated morphology indicating that there might be presence of some smectite.

Chlorite is the second most common mineral (~6% to ~31wt%) and is present in all the studied samples. Chlorite increases incrementally with the depth interval from 997 m to 3407 m in the Shahbazpur-1 well (Fig. 4C). The

 Table 2. Semi-quantitative abundance of different clay minerals (%) of the Neogeneshalesin four wells in the Bengal basin. HB = Habiganj-11, SB = Shahbazpur-1 and TT = Titas (11 & 15) (for location, see Fig. 1)

Well	Sample	Depth (m)	Semi-quantitative abundance of different clay minerals (%)					
			Illite	Kaolinite	Chlorite	Smectite		
Shabazpur-1	SB-1 SB-3 SB-5 SB-6	997-1006 2016 3020-3021 3406-3407	66.2 78.2 81.8 76.2	6.2 7.5 8.1	7.2 9.5 10.1 23.8	20.4 4.8		
Titas - 11	TT-11-1 TT-11-2 TT-11-3 TT-11-4 TT-11-5 TT-11-6 TT-11-7 TT-11-8 TT-11-10 TT-11-10 TT-11-10 TT-11-10 TT-11-11 TT-11-12 TT-11-13 TT-11-15 TT-11-16 TT-11-17 TT-11-18 TT-11-18 TT-11-18	2318.3-2318.9 2712.72 2715.46 2716.2-2716.8 2717.29 2719.73 2721.56 2721.86 2722.47 2728.26 2736.9-2737.8 2737.56 2738.32 2740.15 2745.33 2745.64 2782.82 2785.4-2786.3 2789-2790	78.8 53.0 49.4 84.7 45.8 47.9 48.1 48.9 49.0 50.0 76.2 52.0 52.8 49.4 50.8 49.4 50.8 49.2 66.8 82.3 80.6	8.8 12.5 20.8 - 23.0 22.3 23.5 20.5 19.8 20.5 7.0 18.7 20.8 21.3 20.4 21.1 10.3 8.0 6.2	9.5 31.1 26.7 15.3 28.9 27.7 26.5 27.7 31.2 29.5 10.0 27.0 24.0 29.3 27.1 28.5 22.9 9.7 6.1	2.9 3.4 3.1 2.3 2.1 1.9 3.0 - - 6.8 2.3 2.4 - 1.7 1.2 - 7.1		
Titas - 15	TT-15-1 TT-15-2 TT-15-3 TT-15-4 TT-15-5 TT-15-6 TT-15-7	2661 2662.25 2663.5 2716.8 2717.3 2719.4 2720.6	62.4 43.8 45.4 54.2 51.9 53.4 64.2	18.3 23.1 23.8 19.4 20.5 20.6 10.4	19.3 30.3 28.2 24.1 25.5 24.8 23.6	2.8 2.6 2.3 2.1 1.2 1.8		
Habiganj-11	HB-2 HB-4 HB-5	3088 3095 3095.5	87.5 87.3 87.9	4.8 5.5 5.1	7.7 7.2 7.0	- - -		



Fig.3. X-ray diffraction patterns of clay fraction (<2 μm) of shale sample, depth 2719m in Titas-11 well. Sm= smectite, Ch= chlorite, Ill= illite, Kao= kaolinite, Q= quartz

SEM photomicrographs (Fig. 5B) show the chlorites as small irregular to pseudohexagonal platelets, oriented perpendicular to the grain surfaces.

Kaolinite shows no systematic change in the amount with depth (Fig. 4B) and occurs as thin stacks of pseudohexagonal crystals with a book-like morphology (Fig. 5 C). It's content varies from  $\sim$ 5% to  $\sim$ 24 wt%.

Smectite could be identified based on the X-ray diffraction profiles of the ethylene glycol treated samples; a distinct 16.97-17.51 Å (001) glycolated peak was identified as expanded smectite. Smectite ranges from ~2.9wt% (2318.3-2318.9 m depth, (Titas-11) to ~20.4wt% (997-1006 m depth, Shahbazpur-1). It decreases at depths >1006 m, and is found only in minor amounts at depths of 1277.5 - 2790 m and absent at deeper depths ( $\geq$  3000 m) (Fig. 4, Table 2).

# **Chemical Composition**

The major element composition of the 2  $\mu$ m clay-sized fractions of the Surma Group shales with depth is shown in Table 3. The amount of the measured K<sub>2</sub>O content < 7.5 % K<sub>2</sub>O in illite (Azzaro et al., 1988) implying that the mixed layers of smectite might be responsible. Significant amounts of MgO possibly indicate the presence of chlorite.

# DISCUSSION

The mineralogical and chemical compositions of the  $<2\mu$ m clay-sized fractions from the Neogene shales comprises mainly Illite, followed by chlorite, kaolinite and minor smectite. Diagenetic alteration of smectite to illite and chlorite in the Surma Group shales is clearly evidenced by the decrease in smectite and the corresponding increase





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Fig.5. Scanning electron micrographs of shales showing: A. illite-smectite (ill-sm)- depth 2789-2790 m, well Titas-11 (TT-11);
B. chlorite (ch)- depth 3020-3021 m, well Shahbazpur-1 (SB); C. kaolinite (K), chlorite (Ch)- depth 3095 m, well Habiganj-11 (HB);
D. Late quartz overgrowths (QO) partly occludated pore-throats- depth 3020-3021 m, well Shahbazpur-11 (SB).

in illite and chlorite with depth in the Shahbazpur-1 well (Fig. 4A, C, D). Smectite has not been observed at depths >3000 m (Fig. 4). In the Shahbazpur-1 well, there is a clear trend of gradually decrease of smectite with depth and its ultimate disappearance at depth around 3020m. This is broadly consistent with the observation of Imam (1994) in the case of the Patharia anticline (Surma basin, NE Bengal Basin) (see Fig. 1). There is no evidence of discrete or pure smectite in the Neogene shales (Surma Group) of Patharia anticline but instead illite/smectite mixed layer and diagenetic illitization has been reported from the Patharia-5 (Imam, 1994). The alteration of smectite to illite (illitization), accompanied by a decrease in kaolinite content and an increase in chlorite content, is recorded frequently in diagenetic studies (Perry and Hower 1970; Howeret al. 1976; Jahren and Aagaard, 1989). According to Weaver and Beck (1971), the presence of potassium ( $K^+$ ) is necessary for the conversion of smectite to illite, which is introduced into the shale by complete decomposition of potassium feldspar and some mica.

Table 3. Major element analyses (wt %) of the  $<2\mu$ m clay sized fractions of the Surma Group shales in three wells in the Bengal Basin. HB = Habiganj-11, SB = Shahbazpur-1 and TT = Titas-11; 1, 2, 3, .....= core 1, core 2, core 3

Sample	Depth(m)	$SiO_2$	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	TiO <sub>2</sub>	$P_2O_5$	LO1
TT-2	2318.3-2318.9	34.16	18.41	11.43	0.07	8.04	2.09	0.71	2.79	0.80	10.77	10.12
TT-4	2736.9-2737.8	38.14	20.43	8.02	0.16	6.56	1.00	0.70	3.20	1.00	6.56	13.4
TT-6	2789-2789.9	38.14	20.36	9.24	0.08	5.75	1.16	0.61	3.34	0.82	7.63	12.1
SB-5	3020	31.84	17.60	10.32	0.06	8.77	1.61	0.77	2.88	0.88	10.75	13.8
SB-5(1)	3020.5	29.68	16.02	9.45	0.05	7.61	1.51	0.62	2.57	0.75	9.2	21.6
SB-5 (2)	3021	28.81	16.00	10.36	0.06	9.37	1.81	0.73	2.52	0.75	12.35	16.35
HB-2	3088	37.02	20.64	9.72	0.10	7.47	1.21	0.70	3.21	1.03	9.72	8.8
HB-4	3095	36.06	19.96	9.00	0.08	6.13	1.25	0.67	3.42	0.96	7.0	14.7
HB-5	3095.5	35.50	19.82	8.81	0.09	6.55	1.33	0.74	3.51	1.00	7.88	13.88
	Average	34.37	18.80	9.59	0.08	7.36	1.44	0.69	3.05	0.89	9.09	13.86

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Fig.6. Illustrating the major elemental composition with depth burial of the Neogene shales. Major element data from (Rahman and Suzuki, 2007). Samples marked with \* indicates chemical composition of <2 µm clay fraction of shale</p>

The K<sub>2</sub>O content of deeply buried shales is slightly higher than that of more shallow shales, and this is probably related to the development of illite layer (Fig. 6). Si<sup>+4</sup> produced as byproduct of illite/smectite or kaolinite/feldspar reaction, may precipitate as diagenetic quartz (SiO<sub>2</sub>) in deeply buried shales (Fig. 5D). This observation may explain the high SiO<sub>2</sub> contents in some of these shales (Fig. 6). SEM observations reveal that thin plates of chlorite seem to occur on kaolinite surfaces indicating that kaolinte is being altered to chlorite at depths of > 3000 m (Fig. 5C). The variation in abundances of clay minerals (Fig. 4) with depth suggests mainly various inputs from source area controlled by source rock lithology and was thus mainly detrital in origin. Therefore, the observed changes may be both diagenetic as well as being related to source rock composition.

Acknowledgements: The authors would like to thank Alexander von Humboldt Foundation (AvH), Germany for granting Return Fellowship and Ministry of Science and Technology Bangladesh for providing grant to Rahman (First Author) to carry out this research project. We are grateful to BAPEX (Bangladesh Petroleum Exploration and Production Company) for giving permission to analyze core samples.

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(Received: 4 June 2015; Revised form accepted: 28 December 2015)