Geochemical Analysis and Origin of Organic Matter in Coal of Barapukuria Coal Mine, Bangladesh

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Abstract—The country's fuel energy efficiency is as prosperous as the country's prosperity. At present, country's development depends on their fuel energy efficiency. Bangladesh will going downwards day by day without fuel energy. In Bangladesh, some of the fuel energy comes from coal. Barapukuria coals in Bangladesh are one kind source of fuel energy in our country. To properly use of these coal, we should know the rank of coal. But to know the rank as quality of coal, coal is geochemical analyzed where total carbon (TC), total hydrogen (TH), total sulfur (TS) and total nitrogen (TN) concentrations are found. It's were measured by combustion method 1150°C in an CHNS elemental analyzer after removal of carbonates with HCI. Coal samples have recently been collected from Barapukuria Basin at -270 m and -460 m depths and analyzed by using CHNS elemental analysis method. In Barapukuria Basin at -270 m to -460 m depth, the average percentage of carbon is 64.59% (range between 56.50% & 73.50%), hydrogen is 4.67% (range between 3.852% & 5.619%), nitrogen is 3.24% (range between 2.89% & 3.62%) and sulfur is 0.45% (range between 0.377% & 0.532%). Where, the average C/N ratio is 19.93 (range between 19.5781 & 20.3019) and C/H ratio is 13.88 (range between 13.0802 & 14.6682). For average C/N ratio is 19.93 at -270 m & -460 m depths coal of Barapukuria Basin, it can be hoped that organic matter of coals are obtained from aquatic higher plants. And for the average percentage of C, H, N & S at -270 m & -460 m depths coal of Barapukuria Basin, it can be hoped that organic matter of coals are obtained from aquatic higher plants. And for the average percentage of C, H, N & S at -270 m & -460 m depths coal of Barapukuria Basin, it can be hoped that organic matter of coals are obtained from aquatic higher plants. And for the average percentage of C, H, N & S at -270 m & -460 m depths coal of Barapukuria Basin, it can be hoped that organic matter of coals are obtained from aquatic higher plants. And for the average percentage of

Index Terms—Barapukuria Coal Mine, Geochemical Analysis, Coal, Organic Matter, CHNS Elemental Analyzer, Basin and Fuel.

1 INTRODUCTION

In Bangladesh, coal is used as an electricity generation where some of coals come from Barapukuria coals and some of coals are imported coal. Coal is produced more than 550 million ton worldwide [1]. There are five coal fields discovered in the northwestern part of our country such as Barapukuria, Khalaspir, Dighipara, Phulbari and Jamalganj. The coal reserve is estimated more or less 3300 million tons and reserve is remained 2247 million tons in our country [2].

Barapukuria coals are origin in Permian age as Goudwana sequences which depths at 118 to 506 m below the earth surface. Geological Survey of Bangladesh (GSB) was discovered the first coal fields as Barapukuria coal field in 1985. According to discovery, Barapukuria coal field is the second coal field in our country but only one productive coal field.

The Barapukuria coal basin is located at a depth range between 195 and 535 m from the surface in the Parbatipur Thana of Dinajpur district. It covers an approximately 5.25 km² area [3]. There are six coal seams of different thickness and about 390 million tons coals are reserved in Barapukuria coal field. Now, coal is produced in seam VI where its thickness as about 36 m.

The study area is the Barapukuria coal mine which is situated at the northern part of Bangladesh that close to the village of Barapukuria of Hamidpur union under Parbatipur Thana of

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Dinajpur district (fig-1). The study area lies between latitudes 25°31'N to 25°35'N and longitude 88°57'E to 88°59'E [3].

The coal field has six coal-bearing seams which depth of 116 to 506 meters beneath the ground. The rank of coal in Barapukuria coal mine is bituminous coal and the average thickness of coal seam is 51 meters.

Although coal is very complex and contagious, it continues to be the most abundant resource and the principle fuel used for aluminum refineries, paper makers, chemicals and pharmaceutical industries. Many chemical products may be produced by coal products. Refined coal is used in term chemicals such as creosote oil, naphthalene, phenol and benzene.

Geologists predicted the good prospects of discovering huge coal deposits beneath the earth surface of North Bengal for more than hundred and seventy years. In 1961, the Geological Survey of Pakistan (GSP) conducted geological, geological and drilling operations in Bogra and Rajshahi districts, and in Jamalganj, about 1050 million tonnes of coal was found. And GSB was discovered the Barapukuria coal field in 1985 in Dinajpur district [5].

Considering the prevailing circumstances and scopes of this study, the main objectives of these studies are determined the TC, TS, TN, C/H and C/N value and the quality & origin of organic matter in coal of Barapukuria coal mine, Bangladesh.

2 GEOLOGY OF BARAPUKURIA COAL MINE

Barapukuria coal mine is able to make Bangladesh's current power crisis smaller and provide substantial investment in the country's electricity sector. Accordingly, all deposits of Barapukuria have been utilized for extensive research and research, which ends in the development of the country's first

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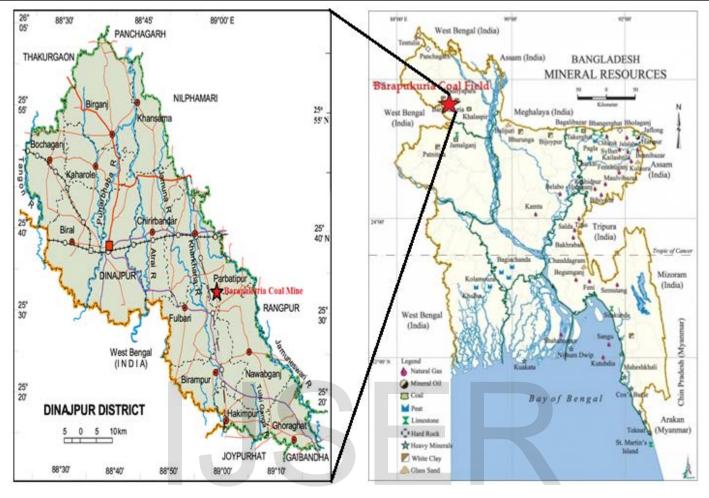


Fig-1: Location of the Barapukuria coal mine (Modified Banglapedia) [4].

TABLE-1 COAL RESERVES IN BANGLADESH [6]

Coal Field (District)	Year of Discovery	Discovered by	Depths of Coal Seam (in meter)	No. of Coal Seam	Reserve (million ton)
Jamalganj (Joypurhat)	1962	SVOC	640-1158	7	1053
Barapukuria (Dinajpur)	1985	GSB	118-506	6	303
Khalaspir (Rangpur)	1989	GSB	257-451	8	147
Dighipara (Dinajpur)	1995	GSB	250	7	200
Phulbari (Dinajpur)	1997	BHP	150-240	2	572

producer of underground coal mine. Geological evolution of Bangladesh is related to the emergence of large delta landmass by the main river system originating from the Himalayas. The basin of Bengal was produced by the collision of Indian plate and Eurasian plate.

These three plates from three major tectonic areas in Bangladesh;

i. A stable platform in the northwest which is divided into Rangpur saddle (Dinajpur shield) [7] and Bogra shelf.

ii. A central deeper (geosynclinals) basin.

iii. The folded belt in the east, known as the Chittagong-

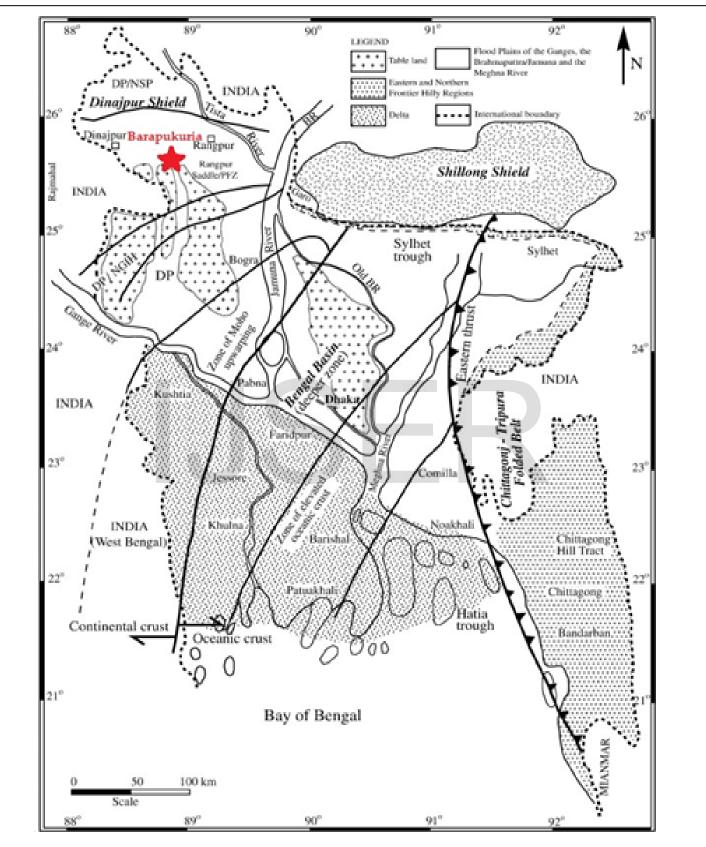
Tripura fold belt [7-9]. Each of these regions is separat-ed by a unique tectonic and stratigraphic history [9].

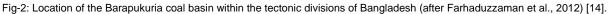
alaThe Barapukuria coal mine is lying in the Dinajpur shield
In(fig-2) of Bangladesh and is enclosed by Himalayan Fore deep to the north, the Shillong Shield/Platform to the east and the Indian Shield to the West [7-9]. The Garo-Rajmahal gap is located between the open Indian Shield and the Shillong
Shield (massif), which is related to a shallow enclosure known as the Platform flank zone [10, 7]. Most of the Gondwana coal basins including Barapukuria, Phulbari, Khalaspir, Dighipara are located within the Bangladesh part of the Garo-Rajmahal

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gap (known as the Rangpur Shield) [11-13].





3 STRATIGRAPHY OF BARAPUKURIA COAL MINE

The stratigraphy of the Barapukuria Basin is given in table-2 [3, 12]. The basin is completely concealed by an unconformable cover between 100-200 m of the late Miocene/Pliocene

Dupi Tila Formation. On the basis of age and lithology, the stratigraphy sequence of the Barapukuria Basin is correlated into four formations [3]:

- i. Madhupur clay
- ii. Dupi Tila (upper and lower)
- iii. Gondwana Group
- iv. Pre-Cambrian Archaean Basement

			DOB [3	3] and GI	
Age Group	Group	Formation	Thickness (m)		Lithology
Holocene-		0-11- 111-11-1	From	To 1	
Recent	-	Soils, Alluvium	0	1	Clay, very fine silt and sand
L.Pliocene- Pleistocene	-	Madhupur clay	3	15	Clay, mostly silty
L.Miocene- M.Pliocene	Dupi Tila	Upper Dupi Tila	94	126	Sand-unconsolidated to partly consolidated, medium coarse, occasionally gravelly with bands of silt
Permian	Gondwana	Lower Dupi Tila The upper coal sequence	0	80	Sandstone, silt and white clay
	Seam I	0	3	Sandstones-arkoses, fine to coarse with occasional conglomerates. Contain coal seam I to V. Occasional siltstones and mudstones.	
		Seam II	14	15	
		Seam III	0.30	6	
		Seam IV	3	10	
		Seam V	1	10	
		The upper sandstone			
		Sequence of Seam VI	15	140	Sandstones-arkoses, massive. Mostly medium coarse and conglomeratetic. Contains no coals and rare siltstones/mudstones.
		Seam VI	22	42	Coals-occasional thin partings of inferior coal, mudstone and sandstone.
		The lower sandstone			
		Sequence of Seam VI and VII	84	164	Sandstone/siltstone/mudstone/coal-rapid intercalation of mostly fine to medium sandstones with numerous coaly and argillaceous bands.
		The Tillites	0	55	Tillites-boulder beds, breccia-conglomerates with occasional intergraded siltstone, sandstone, and rare thin coal bands.
Archaean	-	Basement Complex			Diorite, meta-diorite, ophlitic gneiss and granite.

Seam VI is the thickest and most laterally continuous seam which transversely the basin from 22 m to more than 42 m (average 36 m) and is the main coal seam in the sequence. Seam VI is usually overlain and underlain by thick sandstones which calculate for most of the coal resources at Barapukuria. The depositional environment of the Barapukuria coal seams have been clarify as forming in cold to cool temperate environments and were probably deposited in lacustrine conditions [3, 12].

4 METHODOLGY

Five coal samples were collected from Barapukuria coal

seam VI at -270 m and -460 m depths. Then, samples was dried and pulverized into particle before the test. EAI tin capsule are placed on the balance, press tare when the swinging motion has stopped. 4-8 mg of the sample is added to the capsule and crimps the capsule closed with the forceps and record the mass of the sample. Then the capsule are placed in a nickel sleeve and set it in the sample wheel. Finally, run to CHNS elemental analyzer and analyze the carbon, hydrogen, nitrogen and sulfur element percentage of the five coal samples. All coal samples are analyzed in Euro-vector elemental analyzer (fig-3) at Centre for Advanced Research in Sciences (CARS), University of Dhaka.



Fig-3: Euro-vector elemental analyzer.

The ultimate analysis of coal is vital important for classifica-tion of coal and combustion analysis as it generally deter-mines the % of elements. Working with unknown samples, it is very important to determine the working conditions and must use to optimize the performance of the instrument. To do that basically means to decide the standard and the sample size which are going to use.

Prepare the standards and samples, and drop the samples into the carrousel of the auto-sampler, according to the sample table of auto run, and put the carrousel of the auto-sampler on its position.

Looking for the right standard:

i. If know approximately the composition of unknown samples, must choose the right standard, considering that the ideal standard is the one who has percentages as simi-lar as possible to unknown samples in all the elements.

ii. If have no idea of the composition of samples, can weigh a little amount of sample (around 1mg for instance) and analyze it, just to see the signals in the chromatogram, that will show approximately if there is a very high amount of any element. This will help to choose standard, but in general if have no idea of samples which can use a standard with intermediate percentages of all the elements (Sulfanilic acid in CHNS or Acetanilide in Oxygen analysis, for instance).

Calibrating the instrument with the standard:

Once choose the standard which are going to use, it must perform a linear regression calibration with it, weighting 5 samples from 4 to 8 mg of the standard (more or less). Verify the quality of the calibration looking at the calibration curves to be sure that the results are good enough.

Determining the sample size:

i. Analyze around 4-8 mg of unknown sample and look at the chromatogram and try to compare it with the chromatograms of standards used for the calibration. If there no big differences between the areas of the standard and sample peaks, this could be a good choice for sample size.

ii. If there are very big differences between peak areas on standards and samples which can try to analyze more (or

less) amount of unknown sample, according to the differences could see.

iii. The analysis method is linear, so there will be no problem if a little bit out of the calibration range from the analytical point of view, but if working with unknown samples far away from this range, the quality of the results could be not as good as expected.

Checking the reproducibility:

If finally decide to use for instance 4-8 mg of sample, must be sure that using this sample size which obtain a good reproducibility in the analysis. To do that:

i. Prepare 3 repetitions of unknown samples with similar weights and close to the ideal sample size (4-8 mg).

ii. Analyze the 3 repetitions using the calibration with standard and look at the statistics results which can calculate after exporting to an excel file and calculate the standard deviation between the three repetitions.

iii. If the reproducibility is good enough which can use this sample size and this standard to analyze the samples.

iv. If the reproducibility is not good enough, there are having problems with the representatively and homogeneity of the samples. To solve this problem, it can try to use a bigger amount of sample (can try with 5 or 10 mg, or even bigger sample sizes if necessary). Of course, if increase the sample size, it will probably need to enlarge the calibration range, and must add more standards to calibration curve. Also consider that if increase the sample size; it will need to increase the volume of oxygen used for the combustion.

v. If cannot obtain a good reproducibility in the samples even increasing the sample size, but obtain good results when analyze the standards, it probably have a nonhomogeneous sample. Then it must improve the sample preparation procedure to solve this problem (may be with a better grinding or milling and a strong mixture of the product could be enough).

Analyzing samples:

When it finally obtain a good reproducibility in the samples, then can start to analyze them. To do that:

i. Prepare the samples with the sample size which chose. It must do at least two repetitions for each unknown sample. Analyze the two replicates and check if get similar results for all the elements. If the results are not similar must add another replicate at the end of the sample table to show which is the right result.

ii. From the analytical pint of view, it is recommended to do a calibration with standards everyday are going to analyze samples (5 standards would be enough).

iii. However, from one day to the following many times, it can use the calibration of the previous day. If want to do that, it must be sure that the response in the detector and all the instrument parameters are exactly the same. There is an easy way to determine if can use a previous calibration or not:

a. Import the calibration, want to use from Callidus SW and add a control sample before analyzing the unknown samples. This control sample will be the same standard used for the calibration, but declared as unknown sample.

b. After the control sample, can add the unknown samples. Save the auto run and start the analysis, and when

it's finished, check the results for all the elements of the control sample and compare them with the theoretical results. If the difference between the average percentage of each element in this control sample and the real percentages of each element in the standard is less than 0.3%, everything is ok and can use the previous calibration file to analyze the samples.

c. If the results are not good enough, must do a new calibration to analyze the samples.

iv. When are preparing the sample table to analyze the samples is always a good habit to add at the end of the sample table another control sample with the same standard declared them as unknown, and analyze them. In this way, it

can compare the results obtained before and after run the samples.

5 RESULTS & DISCUSSIONS

Elemental analysis is the determination of carbon, hydrogen, nitrogen and sulfur percentages of samples. In this research, coal samples from Barapukuria coal seam VI at -270 m and -460 m depths were elemental analyzed by Euro-vector elemental analyzer with standard as sulfanilic acid at Centre for Advanced Research in Sciences (CARS), University of Dhaka. So, carbon, hydrogen, nitrogen, sulphur composition (elemental composition) of coal samples and standard as

TABLE-3 ELEMENTAL COMPOSITION OF STANDARD AS SULFANILIC ACID AND COAL SAMPLES AT CENTRE FOR ADVANCED RESEARCH IN SCIENCES (CARS), UNIVERSITY OF DHAKA.

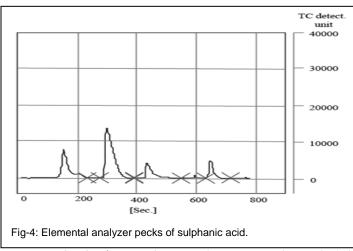
Weight [mg]	4.6170	6.1500	7.2760	7.1211	6.7643	6.5520
Name	Sulfanilic acid	Si	S2	Sa	S4	Ss
Method	2mgChem80s	2mgChem80s	2mgChem80s	2mgChem80s	2mgChem80s	2mgChem80s
N area	21723	12940	12199	12770	12377	12561
C area	43376	101948	92723	98840	95798	97222
H area	11479	21646	17406	19689	19083	19367
S area	8316	466	402	439	425	432
N[%]	8.09	3.62	2.89	3.28	3.17	3.23
C[%]	41.61	73.50	56.50	65.37	63.18	64.37
H[%]	4.070	5.619	3.852	4.704	4.559	4.627
S[%]	18.500	0.532	0.377	0.452	0.438	0.445
C/N ratio	5.1434	20.3019	19.5781	19.9299	19.9306	19.9288
N factor	0.6066	0.6066	0.6066	0.6066	0.6066	0.6066
C factor	1.1001	1.1001	1.1001	1.1001	1.1001	1.1001
H factor	1.3007	1.3007	1.3007	1.3007	1.3007	1.3007
S factor	1.0868	1.0868	1.0868	1.0868	1.0868	1.0868
N Blank	0	0	0	0	0	0
C Blank	0	0	0	0	0	0
H Blank	0	0	0	0	0	0
S Blank	0	0	0	0	0	0
C/H ratio	10.2236	13.0802	14.6682	13.8967	13.8583	13.9118
Humidity [%]	0.00	0.00	0.00	0.00	0.00	0.00

sulfanilic acid are given in below as table-3.

In sulfanilic acid analysis, carbon is 41.61%, hydrogen is 4.070%, nitrogen is 8.09%, sulfur is 18.500% and C/N & C/H

ratio is 5.1434 and 10.2236 respectively. Elemental analyzer peck of sulfanilic acid are given in below as fig-4.

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At -460 depth of Barapukuria coal mine in coal sample (S_1) analysis, carbon is 73.50%, hydrogen is 5.619%, nitrogen is 3.62% and sulfur is 0.532% respectively. Elemental analyzer peck of coal sample (S_1) are given in below as fig-5.

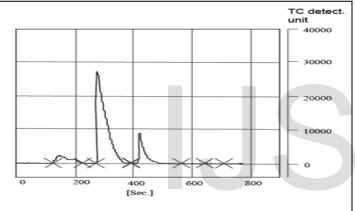
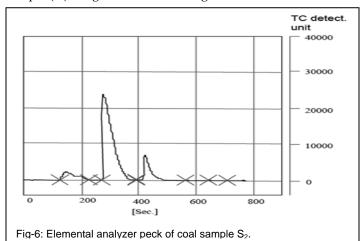


Fig-5: Elemental analyzer peck of coal sample S₁.

At -270 depth of Barapukuria coal mine in coal sample (S_2) analysis, carbon is 56.50%, hydrogen is 3.852%, nitrogen is 2.89% and sulfur is 0.377% respectively, when C/H and C/N ratio is 14.6682 and 19.5743. Elemental analyzer peck of coal sample (S_2) are given in below as fig-6.



At -460 depth of Barapukuria coal mine in coal sample (S_3) analysis, carbon is 65.37%, hydrogen is 4.704%, nitrogen is 3.28% and sulfur is 0.452% respectively. Elemental analyzer

peck of coal sample (S₃) are given in below as fig-7. At -270 depth of Barapukuria coal mine in coal sample (S₄)

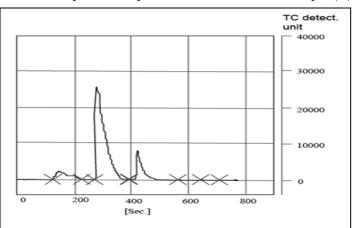
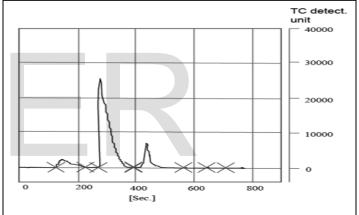


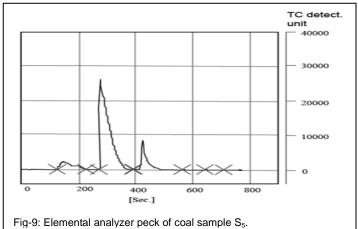
Fig-7: Elemental analyzer peck of coal sample S₃.

analysis, carbon is 63.18%, hydrogen is 4.559%, nitrogen is 3.17% and sulfur is 0.438% respectively, when C/H and C/N ratio is 13.8583 and 19.9306. Elemental analyzer peck of coal sample (S₄) are given in below as fig-8.





At -460 depth of Barapukuria coal mine in coal sample (S_5) analysis, carbon is 64.37%, hydrogen is 4.627%, nitrogen is 3.23% and sulfur is 0.445% respectively. Elemental analyzer peck of coal sample (S_5) are given in below as fig-9.



From the above discussion, it is known that the percentage of carbon, hydrogen, nitrogen and sulfur in the coal samples

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of Barapukuria coal mine in seam VI range from 56.50 to 73.50 wt%, 3.852 to 5.619 wt%, 2.89 to 3.62 wt% and 0.377 to 0.532 wt% respectively. And the average percentage of carbon, hydrogen, nitrogen and sulfur in the coal samples of Barapukuria coal mine in seam VI are respectively 64.59 wt%, 4.67 wt%, 3.24 wt% & 0.45 wt%.

It is also known that the ratio of C/H and C/N in the coal samples of Barapukuria coal mine in seam VI range from 13.08 to 14.67 and 19.58 to 20.30 respectively. And the average ratios of C/H and C/N in the coal samples of Barapukuria coal mine in seam VI are 13.88 and 19.93 respectively.

If the % of C is greater as 64.59 wt%, so the quality and calorific value of Barapukuria coal is better. But, H is mainly present in combination with O as water it lowers the calorific value of coal, the percentage of H is lesser as 4.67 wt%, the quality is better of Barapukuria coal. N is an inert and not combustible gas and doesn't contribute any useful property. It is generally found in small amounts (around 1%). If percentage of N is low as 3.24 wt%, so it is the good quality of coal. S is present in metallurgical coal is harmful for use in metallurgy as it transfers to the metal and adversely affects the properties of metal. S is react with oxygen as form SO_2 and SO_3 that also causes atmospheric pollution. So, the presence of S is undesirable. In Barapukuria coal, the percentage of S is very low as 0.45 wt%.

If the carbon percentage is high with nitrogen and sulfur percentage is low, hence organic matter comes from terrestrial plants [15]. And if C/N ratio is greater than 15, then organic matter is also originating from terrestrial plants [16-17]. In Barapukuria coal samples, the % of C is 64.59 as high with the % of N and S are 3.24 and 0.45 as low within 100 wt% and the C/N ratio is 19.93, so it is said that the organic matter of Barapukuria coal are derived from terrestrial plants.

Again, the low biodegradation is consisting of the high C/N ratio as (>12) and the high biodegradation is consisting of the low C/N ratio as (<12) [18]. In Barapukuria coal samples, the C/N ratio range from 19.58 to 20.30 (average 19.93), so it is said that terrestrial plants are degraded into lower degree of biodegradation.

6 CONCLUSIONS

In Bangladesh, coal is the source of electricity generation. Elemental analysis of coal is determined the element composition as C, H, N and S wt% which help the indicative of quality of coal and origin of organic matter. Five coal samples are collected from seam VI in Barapukuria coal mine at -270 m and -460 m depth. In Barapukuria coal mine for seam VI, the average % of carbon, hydrogen, nitrogen and sulfur of the coal samples are respectively 64.59 wt%, 4.67 wt%, 3.24 wt% & 0.45 wt%. And the average ratios of C/H and C/N in the coal samples of Barapukuria coal mine are 13.88 and 19.93 respectively. From the elemental composition as C, H, N and S wt% of Barapukuria coal mine, it is said that the quality and calorific value of Barapukuria coal is better and organic matter were originated from terrestrial plants.

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