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PETROLOGICAL CHARACTERISTICS OF LIGNITES FROM BEROVO DEPOSIT, FYROM

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The paper presents data about the petrographic and chemical composition of Neogene lignites from Berovo deposit, Republic of Macedonia. The coal material is built by the lithotypes humoclarain, humodurain, xylain, humovitrain, semifusain and fusain. Humoclarain is the one that dominates and displays also the role of a matrix for the rest lithotypes. Macerals from the three maceral groups were established, namely huminite, liptinite and inertinite, with those from the group of huminite being the major ones. Most well represented and of almost equal contents are the structural macerals textinite and ulminite. Mineral admixtures were also confirmed being represented by clay minerals, opal and pyrite. Based on the yield of volatile matter, on the contents of carbon, hydrogen, nitrogen, on the atomic ratios H/C and O/C, as well as on the mean reflection of the maceral ulminite, we determined a low degree of coalification of the studied coal or determining them as being of a low rank C-lignites. The data from the maceral analysis give basis to propose that Berovo lignites are formed in a flooded forest marsh.

Key words: Macedonian lignites, Berovo deposit, petrographic composition, ulminite reflection.

INTRODUCTION

Until now there are no published papers in the specialized literature upon the petrology of lignites from Berovo deposit, Republic of Macedonia. The conducted petrographic, chemical, mineralogical and geochemical analyses of the lignites and the rock varieties associated with them give us the opportunities to summarize the obtained results in a series of papers. In the present one we discuss the petrologic features and the chemical composition of the organic matter that builds up the lignites, while in the upcoming manuscripts we will discuss data about: (1) the mineral substance in the coal; (2) the phase transformations of the organic and mineral matter during self-ignition and fires in the coal seam; (3) the geochemical features of the paleobasin; (4) the inorganic chemical composition of the lignites, host rocks, clinker, and the para-lava.

The paper present results from the petrological investigations of samples from one section in a part of the coal seam that has been exploited in Brik mine of Berovo deposit. The purpose of the present study is to analyze the petrographic composition of the lignites, to describe the macroscopically differing coal varieties in the seam – the lithotipes, as well as the microscopic observations from the quantitative maceral analysis, and to determine the reflection of huminite. The studies are supplemented by data about the chemical composition of lignites obtained during the Proximate and Ultimate analyses.

GEOLOGICAL SETTING

This part of the paper was worked out entirely based on the information given about the geology, stratigraphy, lithology and tectonics of the region of Berovo coal deposit by Dumurdzanov et al., 2004.

"*Berovo*" open coal mine is part of the Delcevo-Pehcevo and Berovo grabens (Fig. 1 (8)). The grabens are located in SE Macedonia. The lithostratigraphy of these grabens is very similar. The NE-SW-trending Berovo graben extends 15 km south from the Bukovik volcano that forms the boundary with the Delcevo-Pehcevo graben. The sedimentary fill of the graben crops out across a ~6 km width, but the sedimentary section near the "Brik"-Berovo coal mine and other smaller relicts of Neogene sediments to the south and west suggest that the area covered by the basin was much larger. The sedimentary section in the Berovo graben is more than 500 m thick and is composed of two formations and Quaternary sediments (Fig. 2).

The upper Miocene Pancarevo Formation (PaF) is ~330 m thick and its stratigraphic section is similar to that of the same formation in the Delcevo-Pehcevo graben. Three superimposed lithological units are present in the Pancerevo Formation. The basal 125-m-thick unit of gravel, sandstone, and claystone? is very similar to the basal unit – the Delcevo-Pehcevo graben. The middle coal-bearing unit of Meotian age is ~100 m thick and is composed of siltstone, grey and green-grey claystone and 1–2 layers of coal developed at the SW margin of the basin at the "Brik" mine.

The upper unit consists of ~100 m of interbedded sandstone, siltstone and claystone. (Dumurdzanov et al., 2004). The uppermost part contains planktonic diatomite that belongs to late Miocene (Ognjanova-Rumenova, 2000). The presence of the kaolinitic claystone, diatomite and rare occurrences of bituminous siltstone could be related to sedimentation following the activity of the Bukovik volcano, but the age of this volcano has not been determined. The Pliocene Solnje Formation (SoF) is more than 150 m thick and in most inner parts of the basin it is eroded. It consists of gravel, sandstone and sandy claystone. Quaternary sediments (Q) are represented by proluvial-alluvial up to 80 m thick sediments. (Dumurdzanov et al., 2004).

Berovo coal deposit is situated to about 8 km from Berovo town in the southern most part of Delcevo-Pehcevo-Berovo basin (Fig. 1 (8)). The landscape has been modeled during Neogene and from a morphological viewpoint it represents a high plane of about 880–940 m above the sea level. One coal seam was established in the

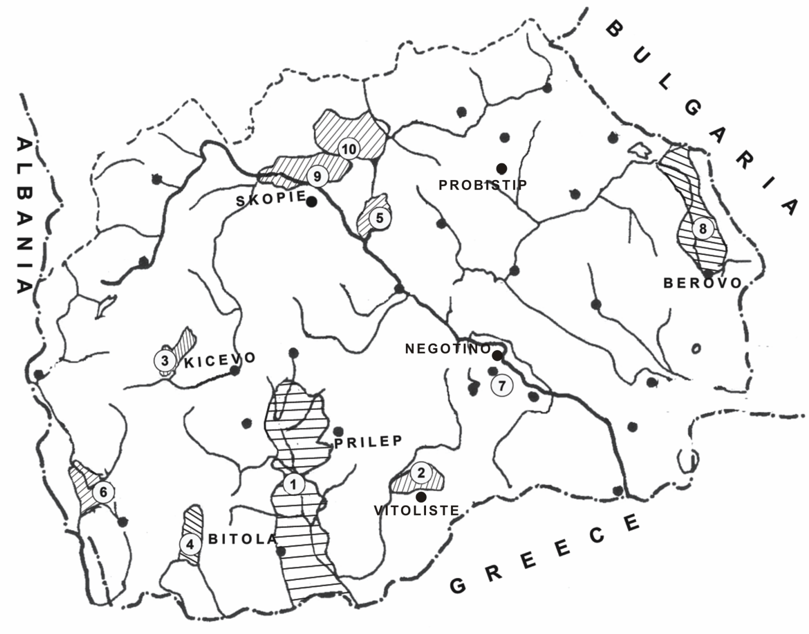

Fig. 1. Location of Macedonian coal deposits (according to Andreevski, 1995)
1 – Pelagonian basin; 2 – Mariovo basin; 3 – Kicevo basin; 4 – Prespa basin; 5 – Katlanovo basin; 6 – Strumica basin; 7 – Tikves basin; 8 – Delcevo-Pehcevo-Berovo basins; 9 – Skopje basin; 10 – Kumanovo basin

aleurolite-productive facies displaying thickness of 2.20 to 8.75 m (with mean value of 5.40 m). In the peripheral parts of the deposit this seam stratifies into 2 to 3 separate layers. The depth of dipping of the upper part of the seam and the thickness of the cover vary, respectively from 4.20 to 28 m, with mean value of 13 m. The geological reserves of the deposit have been calculated to 2×10^6 t (Andreevski, 1995). Mining works are conducted in Berovo deposit, respectively in Brik mine, since 1986. At one and the same time there are performed both, uncovering and mining of lignites. In the time of sampling of the seam during June 2007 it was realized that the mining is performed on 3 or 4 levels with thicknest of the swaths of 1.0 to 2.0 meters.

The geological section in the central part of the mine field has the following sequence: under reddish sandy Quaternary clays of the cover there follow grey-whitish up to greenish massive and dense aleurolites displaying semi conchoidal fracture and well expressed cleavage along the stratification. Their thickness varies from 3.0 to 4.0 m and in the lower part there is observed one interbed of gray-black coaly clays of 0.20 m thickness. Then come the uppermost parts of the coal seam having thickness of about 0.60 m. The coals are of lignite type with greay-black color with dense outlook when they are fresh and then becoming fissured and schistose after drying up. Remnants of leaves, leaf imprints and plant detritus are observed along the surfaces of layering.

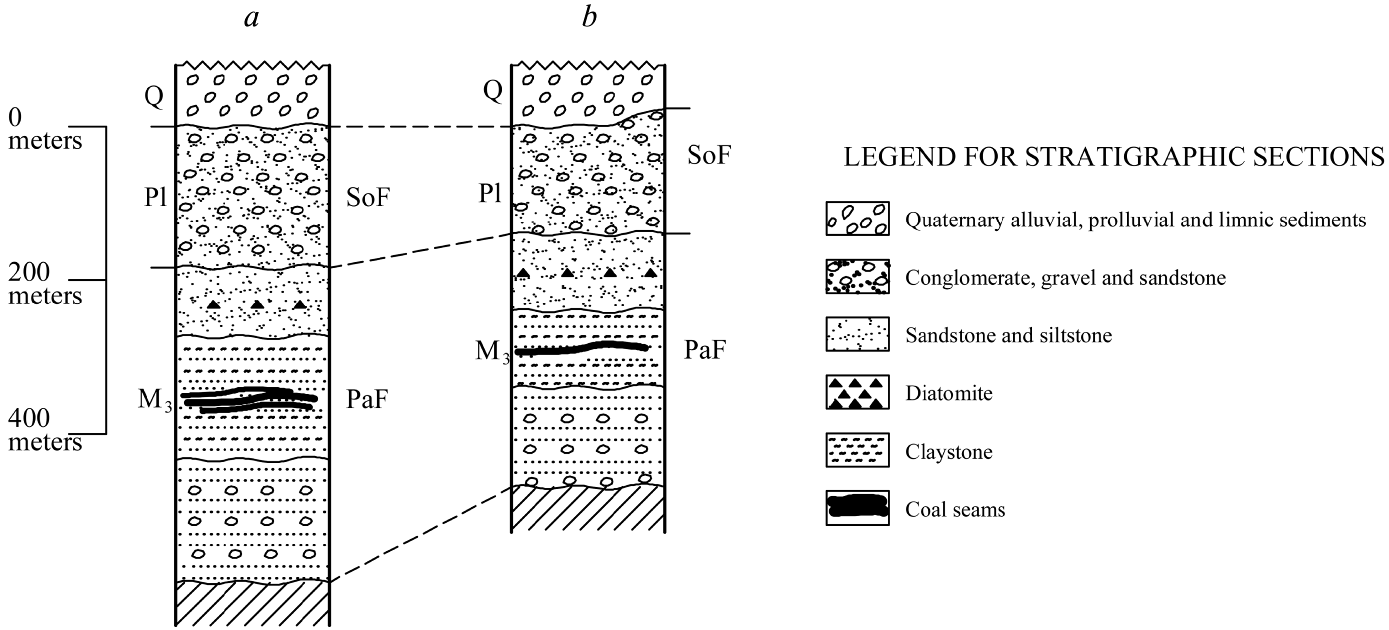

Fig. 2. Stratigraphic sections of the Delcevo-Pehcevo (a) and Berovo (b) grabens
Q – Quaternary, Pliocene (Pl): SoF – Solnje Formation; Miocene(M): PaF – Pancerevo Formation (according Dumurdzanov et al., 2004)

The second mining level comprises 1.0 to 1.2 m of the productive seam and in the upper part there is separated about 0.2 m thick beam of gray up to whitish aleurolites. The lignites are dark brown to gray-black, dense, on places with earthy outlook and inclusions of wood fragments, leaf detritus and fusainized remnants.

The third level with thickness of 1.20 m is separated from the second one with an interbed of gray-white aleorolites with thickness of 0.15–0.25 m. Lenses of fusainized wood fragments are followed between the aleurolites and lignites from the third level. Lignites are dark brown with massive outlook and large inclusions of xylain and lenses of fusain.

Samples from the fourth level of mining were not collected due to technical reasons.

SAMPLES AND METHODS

The sampling was performed downwards along the section of the seam following the levels of the digging in the mine. The objects of study are 15 samples (55B to 70 B). Five samples (55B-59-60B) were taken from the first level along the seam in east-west direction (Fig. 3A). Another 5 samples were taken from the uncovered coal from the second and third level (61B-66B), as well as sample 63B from the aleurolites croping out between second and third level.

The samples with numbers from 67B to 70B were chosen for the characterization of the separate lithotypes – xylain, liptain, humovitrain, semifusain, fusain and some transitions between them. Sample 71B from the upper part of the layer is represented by gray-white to white aleurolites that on places are replaced by clays or fine grained and weakly cemented quartz sands.

The petrographic studies include macroscopic and lithotype description of the samples, preparation of microscopic samples, quantitative maceral analysis, determination of the reflectance of the maceral ulminite.

The determination of the lithotypes was performed according to the lithotype nomenclature accepted in Bulgaria for soft brown coal (Šiškov, Valčeva, 1983). The

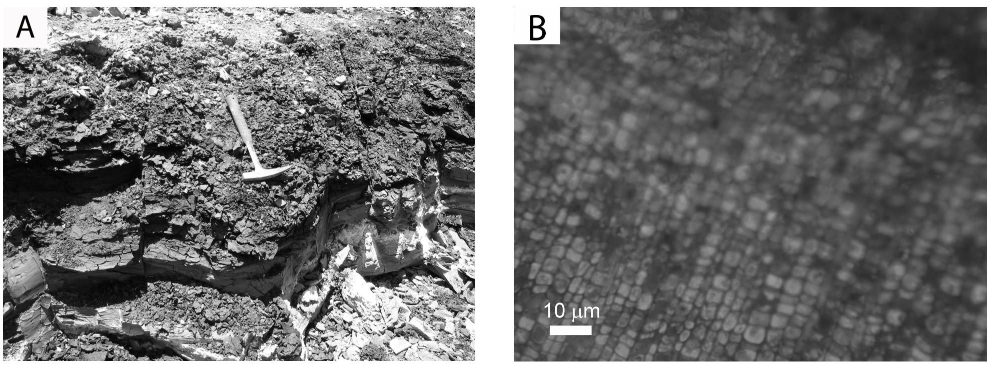

Fig. 3. Images of Berovo coal and textinite
(A) Photographic image of coal seam studied; (B) Microphotographic image of textinite

latter includes the homogeneous lithotypes (xylain, humovitrain, and semifusain) and the heterogeneous ones (humoclarain, humodurain, and liptain).

For microscopic investigation and preparation of particulate blocks the samples were crushed to an upper size of 1 mm, mixed with polyester resin binder and ground and polished of one end face (BDS ISO 7404-2). Fourteen particulate blocks were prepared according to the described procedure.

The quantitative maceral analysis was carried out in reflected white light and oil immersion using a microscope equipped with a point integrator. About 500 points were counted of organic and mineral matter on each particulate block. Identification of macerals followed the classification proposed by the International Committee for Coal and Organic Petrology (ICCP, System 1994, 1998; 2001; Sykorova et al., 2005).

The reflectance was determined by Standard Universal MPT "Opton" microscope-photometer with Antiflex Epi 40/0.65 oil objective in cedar oil immersion at monochromatic light (546 nm). Reflectance measurements were carried out at 50–100 points according to the procedure described in BDS ISO 7404-5.

The chemical investigations of the lignites include Proximate and Ultimate analyses. The Proximate analyses were conducted according to the requirements of the respect Bulgarian National Standards ISO (BDS ISO 5068, BDS ISO 1171, BDS ISO 562) and includes the determination of moisture (W^a, %), ash (A^d, %), yield of volatile matter (VM^{daf}, %). The Ultimate analyses include the determination of the percent contents of C^{daf} and H^{daf} according to ISO 609, N^{daf} according to Kjeldahl (BDS ISO 333) and according to BDS ISO 351.

The powder X-ray diffraction patterns were recorded on DRON 3M diffractometer with Fe-filtered CoKα radiation and using standard flat specimen holder.

RESULTS AND DISCUSSION

LITHOTYPE COMPOSITION

Considerable part of the seam in Berovo deposit is represented by dark-cinnamon brown to gray-black lignites with massive or earthy outlook, built predominantly of the heterogeneous lithotype humoclarain with inclusions of splinter-like or very big fragments of xylain (xylit), smaller pieces of liptain and lenses of semifusain and fusain. According to the lithotype classification of soft brown coal (adopted in 1993 by the International Committee for Coal and Organic Petrology) the lignites in Berovo deposit are related to xylite-rich coal.

The homogeneous lithotypes like xylain, humovitrain, semifusain and fusain form lenses and interbeds among the main mass of the coal seam. They are formed as a result of specific biochemical transformation of wood remnants during the peat stages (Šiškov, Valčeva, 1983).

Xylain forms lenses and fragments of monolith vegetation remnants with dimensions of several to tens of centimeters. It is characterized by light brown to cinnamon-brown color and well-preserved structure with clearly expressed rings. In the sampled section this lithotype displays higher contents in the second and third level.

Humovitrain is rarely observed and only in transitions to xylain. It is formed from smaller stems and twigs with structure partially converted to gel-like mass and is characterized by dark brown color, glassy luster and conchoidal fracture.

Semifusain displays dark brown to black color and silky luster. It is represented by wood remnants of pieces sized 2×6 cm.

Fusain looks like charcoal and builds pieces and fragments. It is characterized by black color, fibrous structure, silky to metal luster and ashy outlook. Easily disintegrates to smaller pieces. Together with semifusain they form bigger nests and lenses among the lignites from the third level.

The heterogeneous lithotypes humoclarain and humodurain are formed from detritus and completely dispersed vegetation remnants. They play the role of matrix for varying in size inclusions of the homogeneous lithotypes.

Humoclarain is dark brown to gray-black in color with earthy to massive outlook. It builds the main part of the lignites.

Humodurain is gray-black, mat and with greater content of mineral admixtures. It is characteristic for the uppermost levels of the coal layer.

Liptain is a specific lithotype, which builds vegetation fragments and lenses and is formed from coniferous vegetation during enrichments of lignin-cellulose tissue with tar substances (Шишков, 1976). The observed liptain is fragile with well-formed rings and characteristic yellow, ochre, or light-brown color. Most often it associates with xylain.

MACERAL COMPOSITION

During the microscopic studies of the lignites from Berovo deposit there have been observed almost all macerals of the three maceral groups – huminite, liptinite and inertinite, as well as some mineral admixtures (Table 1). From the data shown it is apparent that leading role in the composition of lignites is played by the macerals from the group of huminite followed by those of inertinite group, while liptinite and the minerals are considerably lower in content.

The total quantity of macerals from the huminite group in the seam varies from 63.2 to 91.2 vol. % of the total mass and from 68.4 to 93.2 vol.% on a mineral matter free basis (m.m.f.b). For the lithotypes of xylain, humovitrain and liptain the quantities vary, respectively, in the ranges of 73.1 to 87.5 vol.%, 78.9 to 97.2 vol.% (Table 2).

During the quantitative analyses it was established that all macerals from the huminite group, namely textinite, ulminite, atrinite, densinite, corpohuminite and gelinite are present. It is of interest to mention the quantitative relations of the macerals, which characterize the degree of decomposition and humification of the original vegetation. Widely represented are the macerals of the two end positions of the gel formation process, respectively the non-altered textinite and the partly altered ulminite combined in the maceral subgroup telohuminite, as well as the partially or fully decomposed atrinite and densinite from the subgroup of detrohuminite. More restricted is the presence of the subgroup gelohuminite, which includes the macerals gelinite and coprohuminite.

Textinite is represented by remnants of very well preserved, non-gelified wood and herbaceous vegetation. It is characterized with clear cell structure with opened lumens, sometimes filled with resinite or corpohuminite (Fig. 3B). Its content varies in wide ranges from 1.8 to 53.8 vol.% in the seam samples and from 5.1 vol.% in humovitrain up to 78.7 vol.% in the sample of xilain.

Table1

Petrographic composition (maceral analysis) of the lignite from Berovo basin, vol.%

Maceral and mineral	**55B**	**56B**	**59B+60B**	**61B**	**62B**	**64B**	**65B**	**66B**	**67B**	**68B**	**69B**	**70Ba**	**70Bb**
Textinite.	13.0	11.5	3.2	1.8	38.7	11.0	53.8	19.6	70.9	69.7	5.1	55.2	78.7
Ulminite	15.2	21.7	17.5	36.6	18.4	25.0	18.0	31.2	4.3	4.2	57.2	15.5	2.2
Dencinite + Attrinite	22.4	15.0	35.5	32.5	2.6	39.0	12.1	26.5	3.6	5.5	2.5	4.9	4.4
Corpohuminite	3.0	5.2	6.7	2.6	13.8	2.0	6.0	1.6	2.0	4.2	8.3	3.0	2.2
Gelinite	13.0	9.8	9.2	5.5	1.6	2.0	1.3	4.4	1.3	0.5	–	–	–
Total Huminite	**66.6**	**63.2**	**72.1**	**79.0**	**75.1**	**79.0**	**91.2**	**83.3**	**82.1**	**84.1**	**73.1**	**78.6**	**87.5**
Sporinite	1.9	3.8	2.2	3.9	–	–	–	2.2	–	1.6	0.7	–	–
Cutinite	3.6	3.1	3.8	1.0	0.6	1.0	0.3	1.3	–	–	1.8	0.7	–
Resinite	0.8	–	–		0.7	0.8	1.0	–	1.3	–	0.4	1.3	–
Suberinite.	–	0.7	1.3	0.3	–	1.3	1.0	–	–	0.5	0.7	–	–
Alginite	–	–	–	1.3	–	–	–	–	–	–	–	–	–
Total Liptinite	**6.3**	**7.6**	**7.3**	**6.5**	**1.3**	**3.1**	**2.3**	**3.5**	**1.3**	**2.1**	**3.6**	**1.8**	–
Funginite	9.6	10.1	5.0	2.6	15.4	2.0	2.9	3.1	–	5.5	5.5	2.6	3.1
Semifusinite	3.0	4.5	1.6	3.7	5.9	4.0	1.3	3.5	1.0	3.2	5.1	–	0.9
Fusinite	0.8	2.8	–	0.8	–	1.6	–	0.6	–	–	–	–	–
Macrinite	3.0	3.8	1.3	0.3	–	–	–	0.3	–	–	5.5	–	–
Inertodetrinite	–			4.7	–	2.8	–	1.6	–	–	–	–	–
Total Inertinite	**6.8**	**21.2**	**7.9**	**12.1**	**21.3**	**10.4**	**4.2**	**9.1**	**1.0**	**8.7**	**16.1**	**7.1s**	**4.0**
Clay mineral	4.4	0.3	5.7	1.6	–	5.5	0.7	4.1	4.9	1.2	–	3.4	1.4
Pyrite	6.3	7.3	7.0	0.8	2.3	2.0	1.3	–	10.7	3.9	7.2	9.1	7.1
Opal	–	–	–	–	–	–	0.3	–	2.3	0.5	–	–	–
Total Mineral	**10.7**	**7.6**	**12.7**	**2.4**	**2.3**	**7.5**	**2.3**	**4.1**	**15.6**	**5.1**	**7.2**	**12.5**	**8.4**

Table 2

Petrographic composition calculated on a mineral matter free basis (m.m.f.b.) of the lignite from Berovo basin, vol.%

Maceral \ Samples	55B	56B	59B+60B	61B	62B	64B	65B	66B	67B	68B	69B	70Ba	70Bb
Textinite.	14.5	12.5	3.6	1.9	39.6	11.8	55.0	20.5	83.8	73.5	5.5	62.8	85.9
Ulminite	17.0	23.5	20.0	37.5	18.8	27.4	18.4	32.7	5.4	4.4	61.7	17.7	2.4
Dencinite + Attrinite	25.0	16.3	40.7	33.2	2.7	42.4	12.4	27.7	4.3	5.8	2.7	5.6	4.9
Corpohuminite	3.4	5.7	7.6	2.7	14.1	1.9	6.0	1.6	2.4	4.4	9.0	3.5	9.0
Gelinite	14.5	10.6	10.5	5.6	1.7	1.9	1.4	4.6	1.6	0.5	–	–	–
Total Huminite	**74.4**	**68.4**	**82.4**	**80.9**	**79.1**	**85.4**	**93.2**	**87.1**	**97.2**	**88.6**	**78.9**	**88.6**	**97.2**
Sporinite	2.2	4.2	2.5	4.0	0.7	0.9	0.3	2.3	–	1.7	0.8	–	–
Cutinite	4.0	3.4	4.4	1.1	0.7	0.9	1.0	1.3	–	–	1.9	0.9	–
Resinite	0.9	–	–	–	–	0.6	1.0	–	1.6	–	0.4	1.3	–
Suberinite.	–	0.8	1.5	0.3	–	1.5	–	–	–	0.5	0.8	–	–
Alginite	–	–	–	1.3	–	–	–	–	–	–	–	–	–
Total Liptinite	**7.1**	**8.4**	**8.4**	**6.7**	**1.4**	**3.0**	**2.3**	**3.6**	**1.6**	**2.2**	**3.9**	**2.2**	–
Funginite	10.8	11.0	5.8	2.7	15.8	2.1	3.0	3.3	–	5.8	5.9	5.2	3.4
Semifusinite	3.4	4.8	1.8	3.8	6.0	4.7	1.4	3.6	1.2	3.4	5.4	3.0	1.0
Fusinite	–	–	–	0.8	–	1.7	–	0.7	–	–	–	–	–
Macrinite	3.4	4.2	–	0.3	–	–	–	0.3	–	–	5.9	–	–
Inertodetrinite	0.9	3.0	1.3	0.8	–	3.0	–	1.6	–	–	–	–	–
Total Inertinite	**18.5**	**23.0**	**8.9**	**8.4**	**21.8**	**11.5**	**4.4**	**9.5**	**1.2**	**9.2**	**17.2**	**8.2**	**4.4**

Most often ulminite represents stripes or particles with plant structure altered to various extents. Its content in the seam changes slightly in respect to both total and organic mass (Table 1 and 2). Very low such values (2.2 and 4.3 vol.%) were established in the samples of xylain and liptain, while in the sample of humovitrain, ulminite reaches up to 57.2 vol.% (Table 1).

The high contents of textinite and ulminite combined in the telohuminite maceral subgroup correspond to the presence of significant quantities of xylain and humovitrain on the studied coals.

The macerals atrinite and densinite, which are combined in the maceral subgroup detrohuminite, represent the main mass, which builds the lithotype humoclarain. The studied samples display that the transitions between atrinite (mixture of fine detritus particles sized below 10 μm) and densinite (gelified particles cemented in amorphous mass) are continuous and, therefore, in the maceral analysis they were counted together. Their quantities in the samples from the seam vary from 2.6 to 39.0 vol.% of the total mass and, respectively, from 2.7 to 42.5 vol.% of the organic mass. Lowest contents of the macerals from the subgroup of detrohuminite were found for humovitrain – 2.7 vol.%, liptain – 4.3 vol.% and xylain – 5.8 vol.%.

Corpohumite is found in the form of rounded, oval or elongated bodies, formed on the expense of rich in tannine secrets, which play protective role in the live vegetation. It was observed that it fills the cell openings of textinite and ulminite or is isolated among atrinite and densinite. It was found in all samples in quantities from 1.6 to 13.8 vol.% (Table 1).

Gelinite represents non-structured organic matter fully decomposed to the formation of humus gel. It is either not found in the samples from the selected lithotipes, or is met in insignificant quantities (from 0.5 to 1.3 vol.%). The subgroup of gelohuminite that includes gelinite and corpohuminite is most weakly represented.

The liptinite group includes coal remnants of resistant parts of the live vegetation with characteristic morphological features, color and functions. The quantity of macerals from this group varies from 1.3 to 7.6 vol.% of the total mass and from 1.6 to 8.4 vol.% of the organic matter (Tables 1 and 2). The macerals sporinite and cutinite are prevailing in some of the samples with contents of up to 3.9 vol.%, while in other samples there are found resinite and suberinite with much lower contents (from 0.3 to 1.8 vol.%). Remnants of algae – alginite, were observed in some single samples (Table 1).

The group of inertinite is represented by macerals with plant cell structure like: funginite, semifusinite, fusinite and rarely macrinite and inertodetrinite. The contents of inertinite vary from 1.0 vol.% for liptain to 21.3 vol.% for the average samples. Funginite is maceral consisting of mainly high reflecting single or multi-celled fungal spores, sclerotia, hyphae and mycels and other fungal relics with cell structure. Funginite in the studies lignites is the most spread inertinite maceral with contents varying from 2.0 to 15.4 vol.% in the total mass and from 2.1 to 15.8 vol.% in the organic mass, while for xylain and humovitrain this range is 5.8–5.9 vol.%.

Semifusinite occupies an intermediate position in respect to reflection and well-preserved structure to the macerals of the subgroup of humotellinite and fusinite. Its content in the samples changes from 0.9 to 5.9 vol.% (Table 1) and from 1.2 to 6.0 vol.% (Table 2).

The fusinite in the studied lignites is weakly represented displaying quantities of 0.3 to 5.5 vol.%. Semifusinite and fusinite are formed in the peat stage at extreme conditions, which are characterized with presence of high redox potential, dehydration of the medium, active work of aerobic bacterial, as well as a result of forest fires.

Macrinite is found in insignificant quantities (from 0.3 to 3.8 vol.%) mainly in the samples from the uppermost parts of the coal seam. It was observed as not big stripes and lenses of non-structured amorphous mass with gray-white color and high reflection. It has been probably formed from humine substance, subjected to dehydration and oxidation still in the peat stage.

Inertodetrinite is a maceral represented by discrete particles with various form and with sizes ≤10 μm. It was found in single samples from the uppermost part of the layer and is present in low quantities – from 0.8 to 2.8 vol.%.

The mineral admixtures are represented by clay minerals (kaolinite, smectite and halloysite), pyrite and opal. Their contents vary in the range from 2.3 to 12.7 vol.% for the samples from the seam, and from 5.1 to 15.6 vol.% for the separate lithotypes. The quantity of the clay minerals is not high and varies from 0.9 to 5.7 vol.%. High contents of pyrite were established – up to 7.3 vol.%, in the samples from the first level and up to 10.6 vol.% in the samples from the chosen lithotypes. Opal is found predominantly in the samples of xylain and liptain, where it is present in the form of droplets in the cell lumens.

It is well known that the maceral composition very well describes the conditions of deposition and burial of the organic matter, as well as the processes in the peat stage, which are connected with its preservation and stages of transformation. The maceral subgroups telohuminite and detrohuminite are reasonably accepted as determinative for the facial type of the peat swamps. The significant quantities of macerals from the subgroup of telohuminite (20.7÷71.8 vol.%), which are characterized with well-preserved structure, show optimal conditions of preservation of the organic matter — predominantly of wood type. From another point of view, the presence of the macerals from the group of detrohuminite (up to 39.0 vol.%) show that in the peat marsh there have been taking place conditions for accelerated mechanical, chemical and biochemical disintegration predominantly of the bush-like, cane and grass vegetation. Macerals like alginite, semifusinite and fusinite are indicators for the changeable water level during peat genesis. In the peat processes the role of the mineral associations and especially that of the clay minerals is not far below. The facial analysis in a row of coal basins showed that coals rich in well-preserved macerals of the subgroup of telohuminite as is the case with Berovo deposit have been formed in submerged marshes of forest type.

REFLECTANCE

The reflectance was determined on macerals from the telohuminite subgroup mainly on the ulminite maceral. The mean incident reflectance of huminite varies from 0.20 to 0.28 R_r % for the maceral ulminite and from 0.14 to 0.19 R_r % for the maceral textinite (Table 3). The standard deviation varies in narrow boundaries from 0.02 to 0.06 (Table 3). The increase of the reflectance from textinite to ulminite confirms the acceptance that in case of low rank coal this indicator increases in relation to the degree of homogenization and tightening during gel transformation of the original plant material (Jakob, 1969; Šiškov, 1971; Valčeva, 1979). When comparing the data for the reflectance of ulminite from Berovo deposit with the mean values (0.13–0.22 R_r %) of

Table 3

Reflctance of the huminite group macerals

Sample	Maceral	Reflectance $R_r \pm \sigma$ %	Variation range
55B	ulminite	0.27 ± 0.06	0.10 ÷ 0.40
56B	ulminite	0.28 ± 0.06	0.15 ÷ 0.40
59B-60B	ulminite	0.28 ± 0.05	0.15 ÷ 0.45
61B	ulminite	0.25 ± 0.03	0.20 ÷ 0.30
64B	ulminite	0.26 ± 0.05	0.15 ÷ 0.40
65B	ulminite	0.20 ± 0.03	0.15 ÷ 0.25
66Ba	ulminite	0.23 ± 0.05	0.15 ÷ 0.30
67B	textinite	0.24 ± 0.04	0.15 ÷ 0.30
68B	textinite	0.20 ± 0.04	0.15 ÷ 0.30
69B	ulminite	0.26 ± 0.06	0.15 ÷ 0.45
70Ba	textinite	019 ± 0.02	0.15 ÷ 0.25
70Ba	textinite	014 ± 0.02	0.10 ÷ 0.15

some Bulgarian lignites (Šiškov, 1997) and with the values for eu-ulminite (0.15–0.39 R_r%) in lignites from Ptolemais basin, Nortehn Greece (Georgakopoulos, Valčeva, 2000), it was found that they belong to one and the same low rank coal – lignite C.

CHEMICAL CHARACTERISTICS

The chemical parameters of the Berovo lignites are represented in Table 4. The moisture (W^a %) varies from 10.5 to15.0 wt.% for the samples from the seam, and from 7.2 wt.% for liptain to 11.4 wt.% for fusain. The samples are characterized by low to relatively high ash (5.3–21.5 wt.%). For the separate lithotypes it is: 2.4 wt.% for liptain, 4.9 wt.% for xylain, 6.6 wt.% for semifusain and 6.8 wt.% for fusain (Table 4).

Generally, the lignites under study are of high volatile matter (VM^{daf}) varies from 56.0 to 63.8 wt.% (on a dry ash-free basis). Sample 64B is characterized with highest contents of such substances (63.8 wt.%) and lowest contents of carbon (C^{daf}, 54.5 wt.%). Liptain is among the lithotypes with highest yield of volatile matter – 72.5 wt.%. Slightly lower values are displayed by sample 70Bb (69.9 wt.%), which represents a transition between xylain (64.4 wt.%) and liptain. At the same time fusain display lowest yield of volatiles (44.3 wt.%) and highets content of carbon (64.6 wt.%). The same tendency is for semifusain (52.7 wt.% VM^{daf} and 60.0 wt.% C^{daf}). The shown data for the two last lithotypes are predetermined by the conditions of formation of the peat marsh – microbial-oxidation and/or thermo-oxidation processes.

The total carbon content in the lignites is very low and does not exceed 60.0 wt.% C^{daf}, both along the upper part of the seam and in depth along the section. The carbon content in the xylain samples is also low and varies from 56.3 to 59.0 wt.%. In the case of xylain carbon reaches 62.7 wt.% and the highest values of 64.6 wt.% were measured for the lithotype fusain (Table 4). The obtained values are lower than the ones found for lignites of other Bulgarian basins – Maritsa-East, Maritsa-West, Chukurovo, Stanintsi, Beli breg, Balsha (Markova, 2002).

Hydrogen displays quite high and similar as well values between 5.4 and 5.7 wt.%, excluding sample 66B, where its content is 7.5 wt. %. A little bit higher are the hydrogen values for the separate lithotypes – up to 5.9 wt.% for xylain and to 6.1 wt.% for

Table 4

Chemical characteristics of lignite from Berovo basin, Macedonia

Sample	Proximate analyses (wt. %)			Ultimate analyses, daf (wt.%)					Atomic ratios		Oxidation level	
	Moisture W^a	Ash A^d	Volatile matter VM^{daf}	Carbon C^{daf}	Hydrogen H^{daf}	Nitrogen N^{daf}	Sulfur S^{daf}	Oxigen O^{daf}	H/C	O/C	Grüner	Vesselovski
55B	14.1	21.5	59.0	59.7	5.6	2.2	5.6	26.9	1.13	0.34	5.5	–0.2303
56B	13.4	15.9	56.0	–	–	–	–	–			5.6	–0.2234
56Ba	13.6	21.0	–	58.4	5.7	2.0	3.4	30.51.1	1.17	0.39	–	–
56Bc	10.5	9.2	–	58.7	5.6	1.2	3.3	31.2	1.15	0.40	5.7	–0.2306
57B	12.9	17.7	–	59.1	5.7	2.2	3.3	29.7	1.16	0.19	5.8	–0.2280
59B+60B	11.9	20.6	58.7	58.5	5.6	1.9	5.4	28.6	1.15	0.36	5.2	–0.2180
61B	14.9	13.6	60.9	56.4	5.4	2.3	1.0	34.9	1.15	0.46	6.9	–0.2540
62B	15.0	9.9	57.7	56.5	5.5	1.8	1.3	34.9	1.17	0.46	6.7	–0.2521
64B	12.8	21.2	63.8	54.5	5.5	1.6	1.6	36.8	1.21	0.51	7.0	–0.2752
65B	12.2	16.3	62.0	56.3	5.6	1.7	1.1	35.3	1.19	0.47	6.6	–0.2537
65Ba	13.2	5.3	61.8	–	–	–	–				–	–
66B	13.6	19.2	60.5	54.5	7.5	1.1	0.7	36.2	1.65	0.49	1.9	–0.2614
66Ba	13.2	11.7	58.1	–	–	–	–				–	–
67B-Liptain	7.2	2.4	72.5	62.7	6.1	0.5	1.0	29.7	1.17	0.34	5.0	–1.8841
68B-Xylain	8.2	4.9	64.4	59.0	5.8	0.9	1.3	33.0	1.18	0.42	5.9	–0.2237
70Ba-Xylain	9.0	3.0	64.4	56.3	5.9	0.8	1.1	35.9	1.26	0.48	6.2	–0.2517
70Bb Xylain	8.0	2.6	69.9	56.6	5.9	0.5	1.1	35.9	1.25	0.38	6.2	–0.2485
70Bc Fusain	11.4	6.8	44.3	64.6	6.1	1.0	0.6	27.7	1.13	0.32	4.7	–0.1700
70Bd Semifusain	8.8	6.6	52.7	60.0	6.2	0.9	0.7	32.2	1.24	0.40	5.3	–0.2113

liptain and fusain. In principle the content of nitrogen in the lignites is low and in our case there are no exceptions (Table 4). The percents of hydrogen and nitrogen are almost the same with the proved ones for Bulgarian lignites (Markova, 2002).

It is of interest to note the distribution of the contents of total sulfur in the studied lignites. High values (3.3 to 5.6 wt.% S_t) were established in the highest part of the seam. The quantity of sulfur in the second and third level is in the range 0.7–1.3 wt.% S_t. The same range of value is characteristic for sulfur content in the separate lithotypes. The quantity of sulfur in the lignites from Berovo deposit is higher than the mean values found for some Bulgarian lignite deposits (Markova et al., 2006; Markova et al., 2008).

Oxygen was determined by the difference to 100% and its values change from 28.6 to 36.8 wt.% for the seam and from 27.7 wt.% for fusain to 35.9 wt.% xylain. These results show that the Berovo lignites have significantly higher content of oxygen compared to other Bulgarian lignites (Маркова и др., 1990; Маркова, Кортенски, 1995; Markova, Kortenski, 2001; Markova et al., 2002; Markova, 2002).

The atomic ratio H/C in the first level along the seam has close values, mean 1.15. With depth it increases to 1.65 (Table 4).

The atomic ratio O/C in horizontal direction of the seam has mean value of 0.34. Such value has been found for the lignites in Beli breg deposit, Bulgaria. In depth of the seam the mean values of the same ratio are 0.48 or are much higher from the proved ones for some Bulgarian lignites (Markova, 2002).

The degree of oxidation according to Grunner in direction SE-NW along the seam, is almost equal – mean 5.5. The same index according to Veselovskii is on the range –0,2180 to –0,2303 (Table 4). The results for the oxidation degree according to Grunner for the lignites in Berovo deposit are relatively close to those obtained for some lignites in Bulgaria, while those for the same degree according to Veselovskii are considerably higher (Маркова, Кортенски, 1995; Markova, Kortenski, 2001; Markova, 2002; Markova et al., 2002).

The obtained elemental composition for the lithotypes (Table 4) confirms the conclusions made by Šiškov (1988), that the content of C and H increases from xylain to liptain. In the genetic row semifusain-fusain there is observed a tendency towards increase of carbon, which is a result of the thermo-oxidation transformations of the organic matter. The portion of oxygen lowers with the succession of fusenization, i.e. the thermo-oxidation processes lead to formation of structures that contain less oxygen. During liptinization there are formed groups poorer in oxygen than those found in xylain, but at the same time richer in oxygen compared to fusain (Markova, 2002).

The lithotypes that build the lignites of Berovo deposit are richer in oxygen compared to those in Stanyantsi, Chukurovo and Maritsa-East (Markova, 2002). The ratios H/C for liptain and xylain are close and for semifusain and fusain are higher in comparison to those for the same lithotypes from the above mentioned Bulgarian lignite deposits.

CONCLUSIONS

The petrographic composition of the studied lignite samples is represented by the macroscopically differentiated lithotypes: humoclarain, humodurain, xylain, humovitrain, liptain, semifusain and fusain. The main mass of the lignites is built by cinnamon brown, dense and humoclarain with earthy to massive outlook and there are also present xylain, humovitrain, liptain, semifusain and fusain in varying quantities.

Almost all macerals from the three maceral groups were found during the quantitative maceral and mineral analysis. The macerals of huminite group are dominating followed by those of inertinite, liptinite and the mineral admixtures. The structural macerals textinite and ulminite from the subgroup of telohuminite and the macerarls atrinite and densinite from the subgroup of detrohuminite are most widely spread and in almost equal quantities.

A low degree of coalification of the studied coals is determined basing on the yield of volatile matter, carbon, hydrogen, nitrogen, the atomic ratios H/C and O/C, as well as on the mean reflectance of the maceral ulminite.

Considering the high contents of oxygen and the greater degree of oxidation according to Veselovskii we proved that the studied lignites are richer in oxygen that the lignites of other Bulgarian deposits.

According to the International classification (ISO 11760) applied to the coals from the seam in mine Brik, Berovo deposit, we determine them as low rank C – lignites, with mean reflectance R_r<0.4%, low to relatively high yield of ash (5.3 to 21.5 wt.%), and with relatively high to high content of huminite (68.4 to 93.2 vol.%).

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