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PETROLOGY AND DEPOSITIONAL ENVIRONMENT OF THE COAL FROM BOBOV DOL BASIN, BULGARIA

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ABSTRACT. Coal seams from Bobov Dol basin were sampled and analyzed using recent petrographic methods for establishing the coal petrography. The average huminite reflectance was measured on Eu-ulminite B to be 0.4 %. Petrographic study reveals that the coal is exceedingly rich in macerals from huminite group. Their average amount is 91% on mineral matter free basis. Texto-ulminite, eu-ulminite, densinite, gelinite and corpohuminite were counted in considerable amounts. Textinite was also established in small amounts as part of plant roots, along with phlobaphinite. The macerals from the Liptinite group were detected using fluorescent light, applied for the first time for these coals. Sporinite, cutinite, resinite, alginite, fluorinite, bituminous substances of bituminite and exsudatinite type and liptodetrinite were established. Bituminite, exsudatinite and fluorinite were identified for the first time in these coals. Inertinite macerals were found to be in amount less than 2 percents, thus suggesting continuously wet conditions during deposition of the organic material. The group is represented mainly by fungal remains, indicating weakly oxic conditions through the plant deposition, while the other macerals constitute not more than 0.5 %, or are missing at all. Petrographic properties reveal that the coals were subjected to advanced humification and strong gelification. The results of maceral analysis indicate deposition in continuously wet swamp or marsh type paleomire from mixed herbaceous and wood angiosperm type vegetation, as indicated by the low amount of resinite in the coal.
***Key words**: coal, macerals, maceral analysis, depositional environment, Bulgaria.*

ПЕТРОГРАФИЯ И УСЛОВИЯ НА ОТЛАГАНЕ НА ВЪГЛИЩАТА ОТ БОБОВДОЛСКИЯ БАСЕЙН, БЪЛГАРИЯ

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РЕЗЮМЕ. Опробвани са въглищни пластове от Бобвдолския басейн. Пробите са анализирани със съвременни методи за мацерална идентификация. Среднота стойност на отражението на хуминита, измерена на Еу-улминит В е 0.4 %. Въглищата са богати на мацерали от група Хуминит със средно съдържание на органична маса от 91%. Текстоулминит, еу-улминит, денсинит, гелинит и корпохуминит са установени в значителни количества. Текстинитът е установен в малки количества като част от корени, понякога с флобафинит. Липоидните мацерали са наблюдавани във флуорисцентна светлина за първи път за тези въглища. Установени са споринит, кутинит, резинит, алгинит, флуоринит, битуминит и липтодетрлинит. Битуминитът и флуоринитът са идентифицирани за първи път в тези въглища. Инертинитовите мацерали се установяват в количество под 2%, което свидетелства за обводняване на торфеното блато. Представени са предимно от останки на гъби, които са индикатор за слабо окислителни условия при отлагане на растителните останки, докато останалите мацерали са не повече от 0.5 %. Установените петроложки особености указват за напреднала хумификация и интензивна гелификация на тъканите. Резултатите от мацералния анализ свидетелстват за отлагане на смесена тревна и дървесна ангиоспермна растителност в продължително оводнено блато.

Introduction

The Bobov Dol coal basin is situated in South-West Bulgaria and belongs to the Pernik coal province (Minčev, 1961). The coal-bearing Paleogene sediments are deposited in NW-SE graben structure over denudated crystalline shists, Triassic and Jurassic sediments (Zagorchev et al., 1994). They are typical mollasse sediments, containing coal beds and bituminous shales, separated in four unofficial lithostratigraphic formations (Zagorchev et al., 1994). The lower part of the Paleogene profile is represented by polymictic conglomerates and sandstones with total depth of 250m. Above them thin-layered argillites and marls, containing organic matter in the form of thin stripes, were deposited. The formation has middle Oligocene age (Černjavska, 1970) and is followed by alternating conglomerates, sandstones and aleurollites. The following coal-bearing formation is represented by sandstones, sandy clays, aleurollites, argillites and up to 8 coal beds with varying thickness. It has up to 100m thickness and late Oligocene age (Černjavska, 1970). The upper sediments are thin-layered argillites with late Oligocene to early Miocene age (Černjavska, 1970). The Paleogene sediments were strongly folded by tectonic deformations into several sinclines and an anticline.

Methods

The present study is based on 21 samples, taken from the IV and IV^a coal seams. For microscopic investigations the samples were crushed to a maximum size of 1 mm, mounted with epoxy resin, ground and polished. Maceral analysis was performed by Single-scan method (Taylor et al., 1998) with Leica MPV microscope, equipped with reflected white and blue irradiation light and 20x/0.40, 50x/0.85 and 100x/1.25 objectives for oil immersion investigations. At least 300 points were counted, using automatic point counter "Prior G".

Huminite reflectance was measured on minimum 50 point per sample, using Yttrium-Aluminum-Garnet standard with reflectance 0.899% and 546 nm monochromatic light.

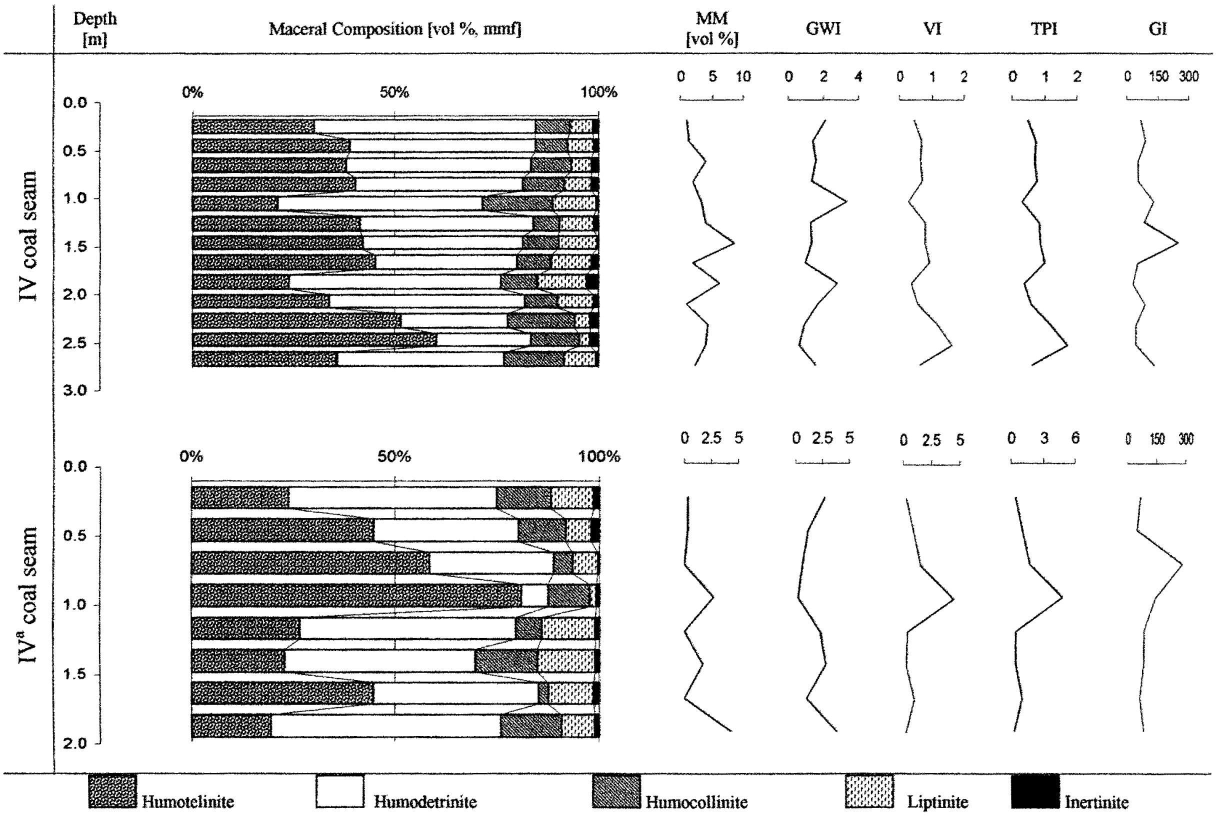


Fig. 1. Maceral composition and facies indices distribution in the IV and IV[a] coal seams, Bobov Dol basin

Results and discussion

Microscopically Bobov Dol coals are banded humic coals with bright and dull bands. The most common macroscopic components are clarain and vitrain with small amounts of fusain. The later occur as small isolated lenses within the groundmass.

The coal rank was determined by measuring huminite reflectance. The values achieved ranges from 0.34 to 0.45% Rr for the both coal seams, which according to the German, North American (Taylor et al., 1998) and Bulgarian coalification classifications correspond respectively to transition from Matt- to Glanz- Brown coals, sub-bituminous C-B coals or O_2-O_3 coalification stage. The average reflectance value, established by this study, is 0.4% Rr and correlates well with previous investigations of these coals (Valceva, 1985, Šiškov et al., 1989).

Macerals from the Huminite group

The petrographic analysis reveals that the Bobov Dol coals are rich in huminite macerals (fig. 1). Although the amount of the individual macerals differs within the samples, it can easily be seen (fig. 1) the large amount of Huminite macerals, which ranges from 88 to 95 % for IV seam and from 84 to 97 for seam IV[a]. The distribution of the Huminite subgroups is shown on figure 1. The amount of the macerals from the Humotelinite subgroup is usually less than 50% of the sample. However 2 samples, taken from the bottom of the IV seam, as well as 2 samples from the middle of IV[a] seam show increased amounts of Humotelinite macerals (fig. 1) as a result of enhanced contribution of wood. Within this subgroup textinite has very small amount, with average value 2.3 vol. %. It has very low reflectance and forms plant roots (Fig. 2a). The cell cavities are empty and the cell walls are highly deformed (Fig. 2a) due to mechanical deformations and the softening of the tissue, caused by the biochemical gelification. The maceral has dark grey color and occurs only in the A variety. Transitions to texto-ulminite can rarely be seen (Fig. 2b). The amount of the later usually does not exceed 6-7 %, except in the samples from 0.4 to 1.2 m depth in IV[a] seam, where the amount of the texto-ulminite ranges from 25 to 34 %. Within the samples from the IV seam slightly increased values was observed at the 0.4, 1.2, 2.4 and 2.6 meters down from the top of the seam. The maceral occurs as B variety, having reflectance, higher than the groundmass. It is characterized by partially closed cell openings (fig. 2b). The most resistant to gelification tissue of the cell walls appears as thin curved line with low reflectance (fig. 2b). Rarely medium reflecting resinite with low fluorescence could be found within the cells, along with bright yellow fluorescing bituminous product, forming from the resinite (fig. 4a, b) (Taylor et al., 1998). Texto-ulminite usually shows transitions to eu-ulminite (Figs. 2b). The later is the most abundant maceral of this subgroup in the coal. Its amount is around 30 % for seam IV and around 26% for the IV[a] seam. It occurs as B variety and forms bands and lenses of more or less homogenous gelified tissue (fig. 2b, c).

Humodetrinite subgroup is represented by both attrinite and densinite macerals. Their average amount for the both coal seams is about 40%. Although the two were not separated quantitatively it appears the later is the most abundant, forming the strongly humified and gelified groundmass of the coal (fig. 2g). Attrinite was observed in transition to densinite (fig. 2e, f) or exists as small detritic particles within the mineral containing bands in the coal (Fig. 2a-lower part). Both densinite and attrinite does not show any fluorescence, which is an indication of the absence of the easily decomposing cellulose.

Humocollinite subgroup is represented mainly by phlobaphinite. The later occurs as small round or elongated bodies, filling the cell openings of former suberinitized tissues (fig. 2d) or texto-ulminite. Its presence indicates contribution from tannin-impregnated woods. The maceral predominates over the pseudo-phlobaphinite, which can also be observed as small bodies within the groundmass (fig. 2e). Gelinite in the form of porigelinite, filling cell-openings in phyllo-vitrinite and rarely in suberinite, was also established in the coal (fig. 2h).

Macerals from the Liptinite group

The amount of the macerals from this group is shown on figure 1. Their color in fluorescence light is orange in accordance to the medium rank of the coals (Taylor et al., 1998). Sporinite was established as flattened elongated and thread-like microspores and pollen (Figs. 3a, b, c). They possess in most cases intact sporopollenin layer suggesting short transportation or none at all. The later indicates autochtonous or hypoautochtonous origin of the sporinite. Sporangium with numerous spores has also been found in these coals (Fig. 3a). The maceral is easily identified in fluorescent light, as well as in reflected light, due to its specific shape and very low reflectance and dark grey color (Fig. 2e, f, g). The amount of microsporinite rarely exceeds 2 %. In addition a megasporinite particle was also established in one sample (fig. 2f).

Cutinite has been identified as leaf protection (Fig. 2h; 3d, e, f). It is usually thin and forms thread-like bodies around phyllo-vitrinite (fig. 2h; 3d, f). The color in fluorescent light is yellow to orange (fig. 3d), but dark green cutinite with very low fluorescence can also be seen (fig. 3f). In reflected light it shows dark grey to black color (Fig. 2h), and can be easily identified, because of its specific shape. The phyllo-vitrinite (an informal name for describing tissues, derived from leafs (Stach et al., 1982)) it covers has different degrees of tissue preservation (Fig. 2h). Single thick cutinite (Fig. 3e) can also be found. The maceral usually associates with fluorinite. The later forms round bright fluorescing bodies within the phyllo-vitrinite (fig. 3e, f) or scattered within the humodetrinite (fig. 3d). Along with the liptodetrinite the maceral is abundant in coal in amount from 2.2 to 2.6 % on average of the liptinite macerals.

Suberinite was identified only in few samples. The cell walls are thin and the cell-openings are filled with phlobaphinite (fig. 2d). The color in reflected light is grey, but in fluorescent light the maceral does not fluoresce, due to the medium rank of the coal. The distinction in reflected light between textinite A and suberinite was based on the shape of their cell-walls, which for the later have elongated form. In addition the cell lumens of the suberinite are always filled with tannin-derived humic substances.

Resinite was identified as round bodies (Fig. 4e, f) filling resin ducts within the plant roots (fig. 2d; 3g, h). Its color in reflected light is dark gray (Fig. 2d; 3h) with strong internal reflections, and in fluorescent light dark yellow-green (fig. 3g). In addition small amount of medium reflecting and very low fluorescing resinite was also established, filling the cell openings of texto-ulminite (fig. 4a). This kind of resinite is known to produce large amounts of hydrocarbons at the boundary between lignite and sub-bituminous coals (Taylor et al., 1998). The later show strong fluorescence in yellow (fig. 4b) and partially or entirely substitutes the resinite.

The resinite amount is usually less than a percent. However few samples are characterized by slightly higher resinite content - at depth of 2.6m. (IV seam) and in the middle and lower part of the IV^a seam.

Bituminous substances of bituminite and exsudatinite type were established in amount less than 2%. They form small round bodies with low brown fluorescence within the groundmass or infill the cells of fungal spores (fig. 3c). These substances are known to form in the beginning of the bituminization process (Taylor et al., 1998) from alginite (bituminite) or the other lipoid macerals (exsudatinite).

All unidentifiable particles of the liptinitic macerals were counted as liptodetrinite (Fig. 3c). Its amount in the samples on average is around 2.5% and along with the sporinite and fluorinite is one of the main contributors of the Liptinite group. Increased amount of liptodetrinite indicate hypoautochtonous to allochtonous origin of part of the liptinite in these coals.

Macerals from the Inertinite group

Bobov Dol coals were found to have very low amounts of macerals from this group. Fusinite, semifusinite and inertodetrintie (Fig. 2e) were established only in few samples in amount less than 0.5 %, thus indicating enhanced humidity of the climate and absence of temperature-controlled destruction of the organic matter during the deposition of the examined coal seams. Fungal remains (ICCP, 2001) are common in all samples, but their amount does not exceed 2%. Exception is made for the sample from 1.8m from the IV seam, where the amount of fungal remains reaches 2.5%. The later are represented by single- (fig. 2f), twin- (fig. 2c) and multi-celled spores (fig. 2h) and roundish sclerotia relicts (Fig. 2g). Plectenchyma tissues can be determined too (Fig. 2g). The cell openings of the remains are usually filled with exsudatinite (fig. 3c).

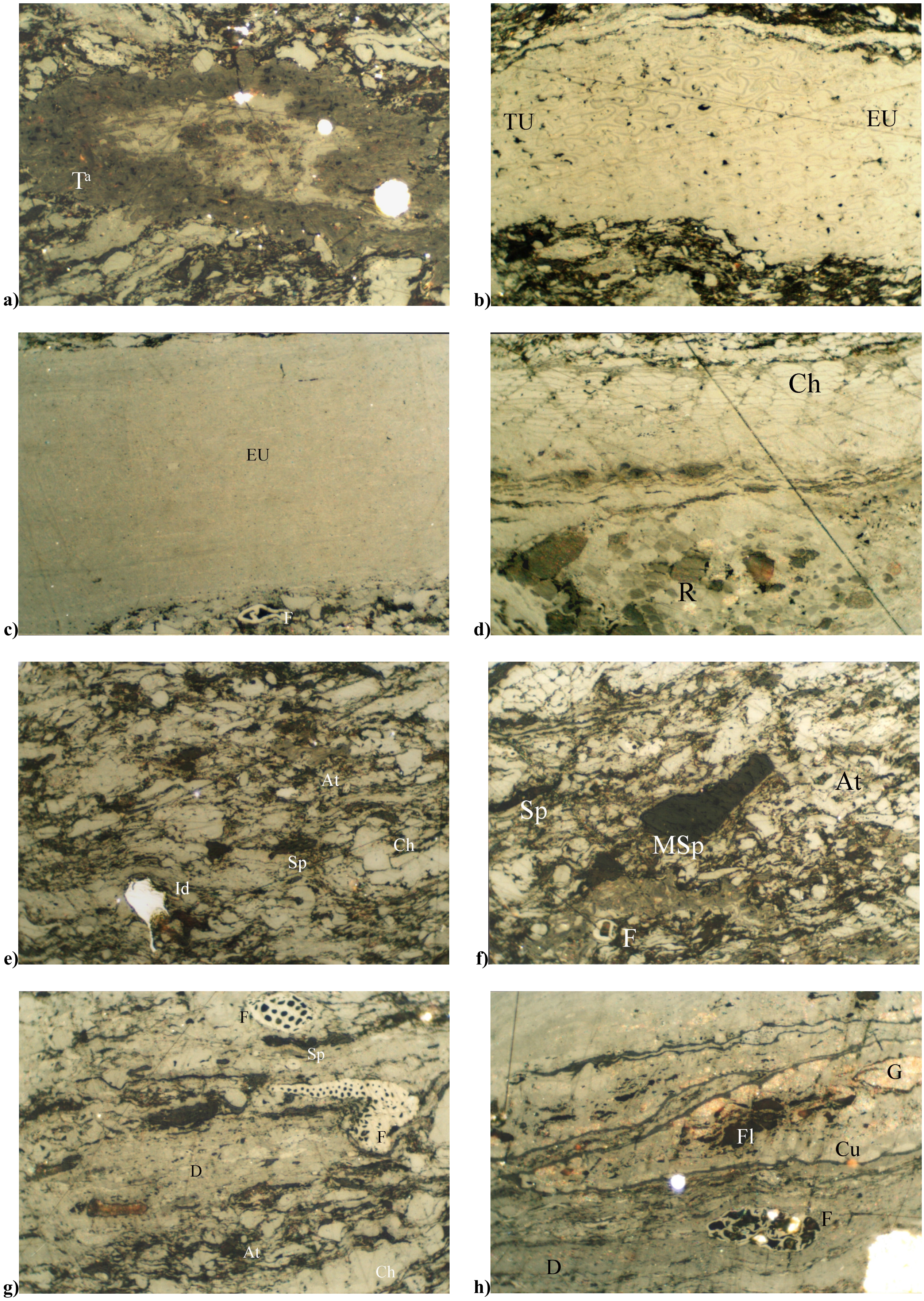


Fig. 2. Macerals from Bobov Dol coal basin – Huminite group: a) Textinite A (T^a), 500x; b) Transition from texto-ulminite (TU) to eu-ulminite (EU), 500x; c) Eu-ulminite (EU), 500x; d) Former bark tissue with corpohuminite (Ch) in the cell walls and resinite (R), 500x; e), f) Transition of attrinite (At) to densinite, along with sporinite (Sp), corpohuminite (Ch), inertodetrinite (Id) and funginite (F), 500x; g) Densinite groundmass (D) with plectenchima tissues (F), 500x; h) Densinite (D) with phyllo-vitrinite, cutinite (Cu), fluorinite (Fl), gelinite (G) and fungal sclerotia (F), 500x. All photos were made with reflected white light under oil immersion

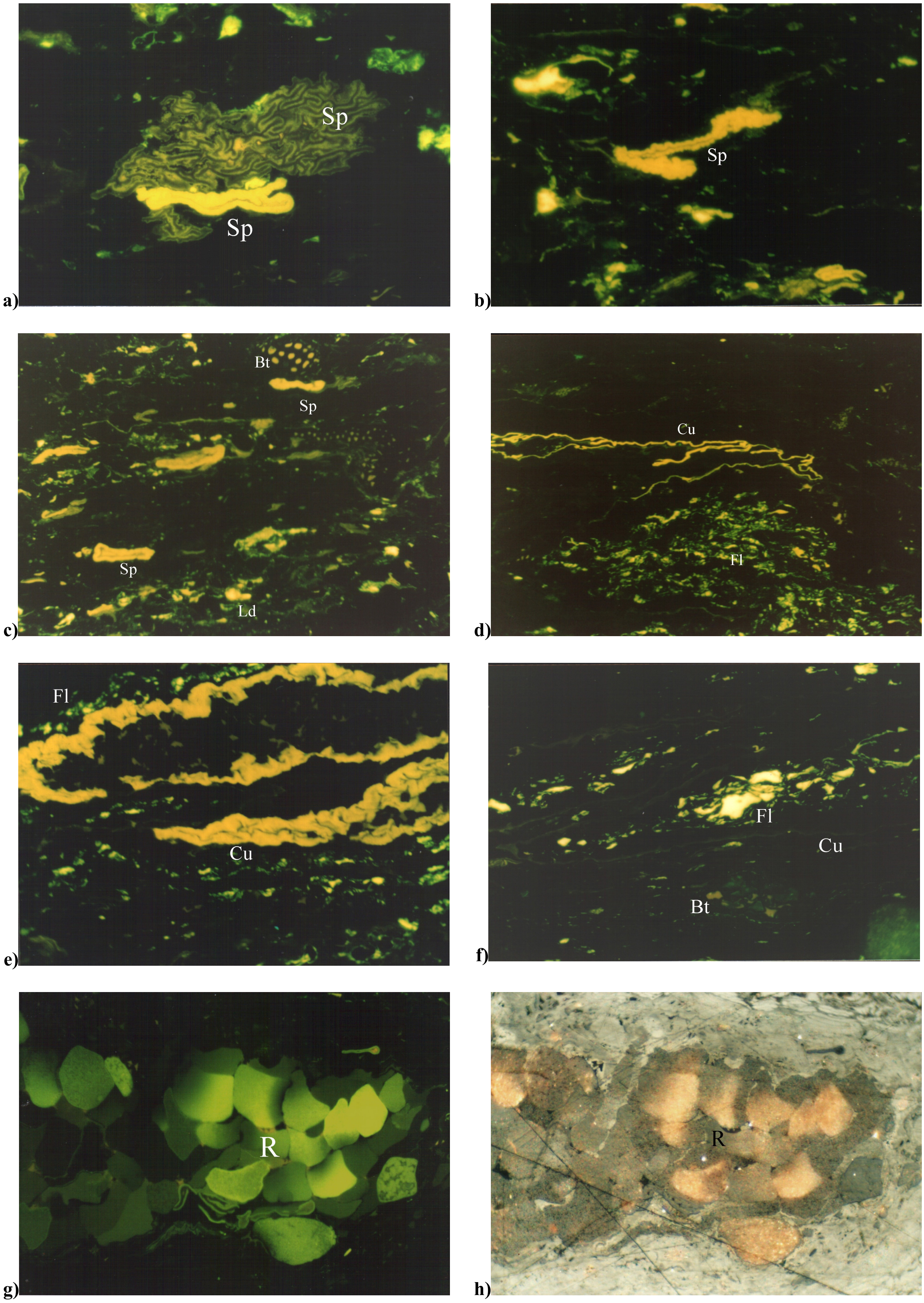
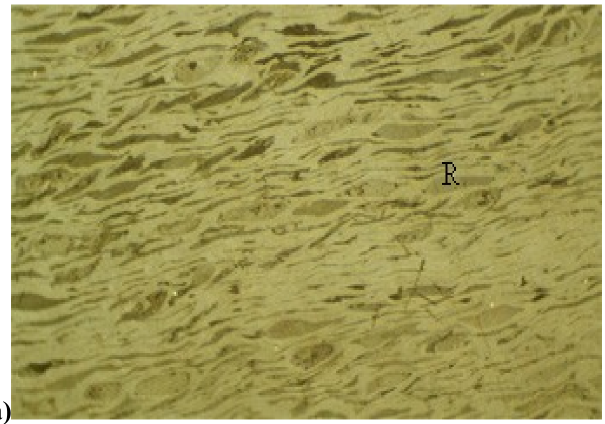
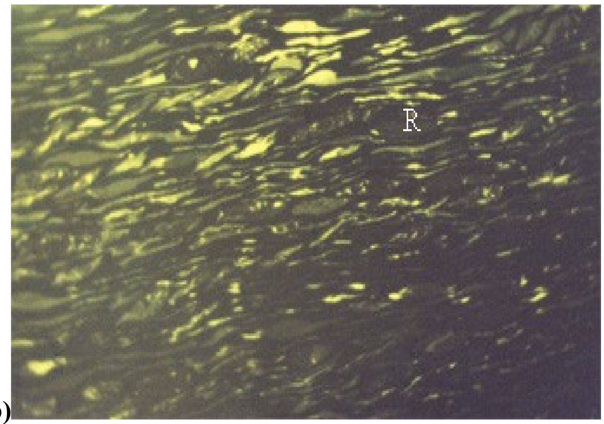


Fig. 3. Macerals from Bobov Dol coal basin – Liptinite groups: a) Sporangium (Sp), 500x; b) Sporinite (Sp), 500x; c) Sporinite (Sp), liptodetrinite (Ld) and exsudatinite (Bt) in the cells of funginite, 500x; d) Cutinite (Cu) and fluorinite (Fl), 500x; e) Thick cutinite (Cu) band, 500x; f) Fluorinite (Fl), 500x; g) Resinite (R), 500x; h) Same as g), 500x reflected light All photos (except h)) were made with fluorescent light under oil immersion



a)



b)

Fig. 4. Macerals from Bobov Dol coal basin – Bituminous substances: a) Texto-ulminite with resinite (R) and low reflecting bituminous substances in the cell openings, 500x, reflected light; b) Same as a), 500x, fluorescent light

Indices of the coal facies

Maceral ratios used for paleoenvironmental interpretations are based on the assumption that coal macerals are plant and environment-dependant and thus can be used to assess the characteristics of the paleomire. There have been various suggestions to characterize the coal facies on the basis of the maceral ratios (Diessel, 1986; Diessel, 1992; Calder et al., 1991). Diessel (1986) introduces two indices – TPI and GI for interpretation of paleoconditions for Permian bituminous coals. These indices were later also applied for lignites and sub-bituminous coals (Kalkreuth et al., 1991, Kolcon et al., 1999; Singh et al., 2000; Gmur et al., 2002 and many others). The Tissue preservation index can be used to determine the degree of humification of plant tissues, because it is contrasting less humified structured and strongly humified unstructured tissue-derived macerals (Diessel, 1992). In addition TPI is also an indirect indicator of the type of the vegetation. In this way high TPI values suggest predominantly wood-derived tissues or high rate of subsidence, resulting in preservation of the structure of the macerals. Low TPI values indicate either predominantly herbaceous vegetation in the paleomire or very low subsidence rate, accompanied with advanced humification, leading to strong decomposition of the plant material (Diessel, 1992). The Gelification index has been defined as a measure of the degree of wet conditions (Diessel, 1992), because it is taking into account the presence of inertinite macerals, which are indicators for dry conditions. There has been a discussion whether fusinite, semifusinite and inertodetrinite are appropriate indicators, because they could be washed or blown into the mire system, or can be a result from crown fires, which are not necessary a consequence of dry conditions (Calder et al., 1991; Scott, 2000). However these macerals were calculated in the formula, because usually their contents are very low and their presence does not change the trends in GI values, which suggests continuously wet conditions during deposition of the coals from the sampled seams. The funginite content was also taken into account, as the later is known as an indicator for oxic conditions. The indices were calculated using formulas 1 and 2.

Similar approach to assess the paleoenvironmental conditions in the mire was made by Calder et al. (1991). They are describing two maceral indices – GWI and VI in respect to the classification of the mires proposed by Moore (1987). The Ground water index is a ratio of strongly to weakly gelified macerals. Detritic mineral matter content is used in the numerator of the formula to determine the type of the mire. The Vegetation index contrasts macerals of forest affinity with those of herbaceous and marginal aquatic affinity (Calder et al., 1991) and thus be an indicator of the vegetation type. Calder's indices were calculated for Bobov Dol coals using the formulas 3 and 4.

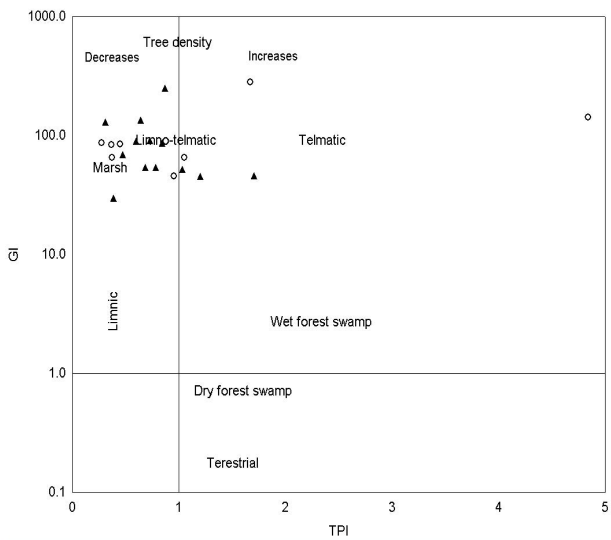
$$\mathrm{TPI} = \frac{\text{textinite} + \text{ulminite} + \text{semifusinite} + \text{fusinite}}{\text{humodetrinite} + \text{gelinite} + \text{macrinite} + \text{inertodetrinite}} \qquad (1)$$

$$\mathrm{GI} = \frac{\text{huminite}}{\text{inertinite}} \qquad (2)$$

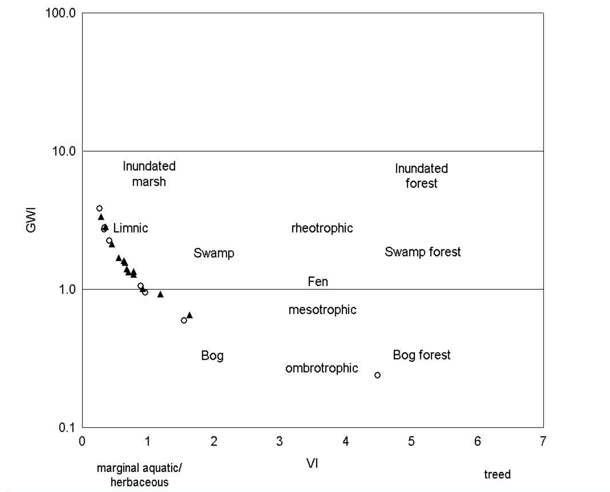
$$\mathrm{GWI} = \frac{\text{humodetrinite} + \text{gelinite} + \text{corpohuminite} + \text{mineral matter}}{\text{textinite} + \text{ulminite}} \qquad (3)$$

$$\mathrm{VI} = \frac{\text{textinite} + \text{ulminite} + \text{fusinite} + \text{semifusinite} + \text{resinite}}{\text{humodetrinite} + \text{inertodetrinite} + \text{alginite} + \text{liptodetrinite} + \text{sporinite} + \text{cutinite}} \qquad (4)$$

For interpreting the paleoenvironment a cross-plot diagram of TPI versus GI and VI versus GWI (Fig. 5) were used. In addition the indices values were plotted versus depth (Fig. 1) in order to establish their distribution within the coal seams. Most of the samples from Bobov Dol basin are characterized by low TPI and VI values, suggesting either increased contribution of herbaceous vegetation, which is usually easily decomposing through the humification process, or strong decomposition of the plant material, due to severe humification of wood tissues. The later is more probable as indicated by the palynological analysis of the coals and coal-bearing sequence. The spore-pollen content, established by Kitanov (1938), Chernyavska et al. (1962), Černjavska (1970) and Palamarev et al. (1998) indicate predominance of angiosperm species from the Juglandaceae family – *Sabal major* Ung., *Myrica lignitum* Ung., *Salix longa* Al. Br., *Carya serraefolia* (G.) Kraeusel, *Carpinus grandis* Ung., *Betula macrophylla* Heer, *Quercus aff.*Ber. Along with them conifer (*Taxodium dist.* (Z.) Rich.mioc.H., *Sequoia langsdorfii* (Brongn.)H., *Alnus kefersteinii* Goepp.), herbaceous (from the Stratoites, Nymphaea, Nelumbo, Nuphar families) and fern (*Pteris parschlugiana* Ung., *Osmunda lignitum* Gieb.) species were also developed. Based on the occurrence and distribution of the paleoflora within the coal-bearing sequence Palamarev et al., (1998) established that the paleoflora was dominated by trees and shrubs with restricted distribution of herbaceous species. This fact agrees well with the conclusions, established by the present study, which indicate that the deposition of the Bobov Dol coals was processed in limno-telmatic to telmatic environment (fig. 5a) from predominantly wood vegetation. However the absence of alginite, which is usually formed in open-water limnic to limno-telmatic environments (Taylor et al., 1998) restricts deposition in such paleomire. Thus the coals were most certainly deposited in a telmatic forest swamp. This is in contrast with the previous investigations of these coals (Konstantinova, 1956) where it was reported the presence of sapropellic layers within the IV coal seam. However in the present study such layers were not established. These layers most probably have restricted distribution and indeed indicate deposition in transitional limno-telmatic environment in that part of the paleomire. The coals are characterized by relatively high humodetrinite content, which is a result of severe humification, caused by enhanced bacterial activity of wood destroying bacteria. The presence of framboidal pyrite, which is thought to have bacterial origin as a result of the activities of sulfate reducing bacteria, could be interpreted in maintenance of this suggestion.



a)



b)

Fig. 5. Cross-plot diagrams of TPI versus GI (a) and GWI versus VI (b);

IV seam IV^a seam

Similar conclusions could be made when analyzing the Calder's diagram (Calder et al., 1991) (fig. 5b). According the plot the coals were formed under preferentially rheotrophic conditions with high ground water input.

The inertinite content in the samples is very small, resulting in high GI values (Fig. 1, 5). The later suggest continuously wet conditions during deposition of the organic matter. A typical constituent of the inertinite group is funginite, which contributors are the fungi, attacking the wood tissue. Because the fungi exist only in the upper oxygenated peatigenic layer, the presence of funginite (despite of its low amount) could be considered as an indication of oxic (though weakly oxic) conditions during plant deposition. In addition the climate was established (Palamarev et al., 1998) to be warm tempered during the Late Oligocene. Climatic changes towards warming of the climate were processed in the Early Miocene, which result in increasing the role of the thermophilous species of Lauraceae family.

Conclusions

Petrographic investigations were performed on two coal seams from Bobov Dol deposit in order their properties to be established. The results reveal that the coals from IV and IV^a seams are rich in macerals from the Huminite group. Among them the contents of humodetrinite prevail, which suggest severe humification of the plant tissues, as a result of enhanced bacterial activity. Microsporinite and liptodetrinite are the main contributors of the Liptinite group. The contents of inertinite macerals are very low. Among them funginite is common in small amounts in all samples, suggesting oxic conditions during deposition of the organic matter. For reconstruction of the paleoenvironment the maceral contents were arranged in four petrographic indices, which along with the features of the macerals, established by the petrographic analysis were used to determine the depositional environment. These indices reveal that the organic material was deposited under rheotrophic continuously wet telmatic to limno-telmatic conditions. The coals are a result of deposition of predominantly angiosperm wood vegetation, mixed with ferns, herbs and conifers, forming forest swamp paleomire.

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