

Preliminary petrographic assessment of the suitability of Bobov Dol coals, SW Bulgaria for fluidized-bed coal gasification

Alexander Zdravkov¹, Doris Groß², Dragana Životic³, Ivan Kojic⁴, Ksenija Stojanović⁵, Achim Bechtel²

¹ University of Mining and Geology “St. Ivan Rilski”, 1700 Sofia, Bulgaria; E-mail: alex_zdravkov@mgu.bg

² Montanuniversität Leoben, A-8700 Leoben, Austria; grossdoris77@gmail.com, achim.bechtelt@outlook.de

³ University of Belgrade, Faculty of Mining and Geology, 11120 Belgrade, Serbia; dragana.zivotic@rgf.bg.ac.rs

⁴ Innovative Centre of the Faculty of Chemistry, Belgrade, 11000 Belgrade, Serbia; ivankojic@chem.bg.ac.rs

⁵ University of Belgrade, Faculty of Chemistry, 11000 Belgrade, Serbia; ksenija@chem.bg.ac.rs

Предварителна петрографска оценка на пригодността на въглищата от Бобовдолския басейн, Югозападна България, за газификация в кипящ слой

Александър Здравков¹, Дорис Грос², Драгана Животич³, Иван Коич⁴, Ксения Стоянович⁵, Ахим Бехтел²

¹ Минно-геоложки университет „Св. Иван Рилски“, 1700 София, България

² Минен университет, А-8700 Леобен, Австрия

³ Белградски университет, Факултет по минно дело и геология, 11120 Белград, Сърбия

⁴ Иновативен център към Факултет по химия, 11000 Белград, Сърбия

⁵ Белградски университет, Факултет по химия, 11000 Белград, Сърбия

Резюме. Въглищни проби от пластове I^a, I, II^{a+b}, III, IV и V от находище Бобов дол в Югозападна България са изследвани с помощта на оптична микроскопия и технически анализ, за да се определи тяхната пригодност за газификация в кипящ слой. Умерените до високи пластово осреднени стойности на въглищната пепел и стойностите на отражателната способност на хуминита в диапазона 0,430–0,499 класифицират изследваните въглища като среден до много нисък клас, нисък ранг А (суббитуминозни) според Международната класификация на въглищата в пласта. Въз основа на органичния състав и пепелното съдържание, въглищата от пластове II^{a+b}, III, IV и V изглеждат по-подходящи за газификация в кипящ слой, но тяхната полезност ще зависи до голяма степен от предварителната обработка на въглищата.

Ключови думи: Югозападна България, Бобовдолски кафяви въглища, органична петрология, газификация.

Abstract. Coal samples from seams I^a, I, II^{a+b}, III, IV, and V from the Bobov Dol deposit in SW Bulgaria were studied using optical microscopy and proximate analysis in order to determine their suitability for fluidized-bed gasification. The moderate- to high seam averaged ash yields and the huminite reflectance values in the range 0.43–0.5 classify the studied coals as medium- to very low grade, low-rank A (sub-bituminous) according to the International classification of in-seam coals. Based on the organic composition and ash yields, the coals from seams II^{a+b}, III, IV, and V seem more suitable for fluidized-bed gasification, but their usefulness will depend largely on the coal pre-processing.

Keywords: SW Bulgaria, Bobov Dol sub-bituminous coal, organic petrology, gasification.

Introduction

With the development of the steam engine, coals have quickly become the driving force of the In-

dustrial revolution and were undoubtedly one of the main factors for the technological development of the mankind. Coals' greater importance is in the coke and steel making industries, as well as a rela-

tively low-cost power generation. Coal utilization in thermoelectric power plants, however, is largely related to environmental pollution with fine particulate matter, toxic metals, nitrogen and sulfur oxides, etc. Despite the current technological development in the construction of thermoelectric power plants, the release of significant quantities of CO₂ during coal combustion is currently considered the greatest ecological problem causing sharp climatic changes.

Strong regulation and the ever-increasing prices of greenhouse emissions in EU during the past decade have already made coals unattractive for power generation and they will soon become economically unprofitable. Therefore, we need to reconsider the possible utilization of the remaining coal reserves with a more environmentally friendly technology. Coal gasification is considered one of the promising cleaner coal technologies, which is already being actively developed for several decades. Nowadays, there are more than 100 different designs of coal gasification reactors, some of which are commercialized, and various fixed(moving)-bed, fluidized-bed and entrained-flow reactors are being operated worldwide for over half a century (Collot, 2006; van Dyk et al., 2006; National Energy Technology Laboratory). The process of coal gasification is essentially focused on the transformation of the solid organic matter under high temperature (>800 °C) into synthetic gaseous products (syngas – mostly composed of CO and H₂, and depleted in CO₂), which can further be used for power generation or production of liquid fuels by Fischer-Tropsch reactions (Wagner et al., 2008; Bielowicz, 2013). While the type of gasification agent (air, steam or oxygen) and the configuration of the gasification system (reactor design, temperature, pressure, residence time, etc.) certainly play an important role, the effectiveness of the gasification process is largely governed by the amount of reactive components (macerals) in coal and the ash yield (Collot, 2006; Bielowicz, 2013; Bielowicz, Misiak, 2020). Both parameters can be easily determined using standard laboratory techniques, and thus the petrographic analysis of the coal is essential for assessing its potential suitability for gasification or for selecting the correct processing technology.

In this short note, we present the organic petrographic data of Bobov Dol coals and following the approach of Bielowicz (2013) we provide a preliminary assessment of the suitability of the coal for fluidized bed gasification.

Geological settings

The Bobov Dol coal deposit is among the oldest known deposits on the territory of Bulgaria, which has been actively mined at least since the late 19th century. The coal has been used essentially for power generation in the nearby Bobov Dol thermoelectric power plant and for domestic heating. This is the largest sub-bituminous coal deposit in SW Bulgaria with 275 Mt total available coal resources as of the beginning of 2002 (Jordanov et al., 2002). Considering the rapid decline of coal mining since then and the gradual closure of all underground mines until 2018, it can be speculated that significant coal resources are probably still available in that deposit. Up to 14 coal seams are distributed within the 100 m thick Bobov Dol Fm. Most of them, however, have limited spatial distribution, low thickness (<1m) and/or high ash yields. Only 6 coal seams (numbered I to VI from base to top) were considered economically significant and were until recently extensively mined in numerous underground mines, mostly along the eastern margin of the basin (Kamenov, 1959). Seam I^a located beneath the first widespread coal seam (I) was locally also mined despite the poor quality of the coal. All seams are extensively folded due to syn- to post-depositional compressional events related to the Savian tectonic phase.

Methods

Maceral group composition of 88 sub-bituminous coal and carbonaceous shale samples representing seams I^a, I, II^{a+b}, III, IV, and V from the Bobov Dol Basin, was determined using Leica DM 2500 P under incident white and blue-excitation light. Maceral identification followed the official nomenclatures for low-rank coals (ICCP, 1998; Sýkorová et al., 2005; Pickel et al., 2017). Random vitrinite reflectance (Ro) was measured on each sample using Zeiss Axio Imager.M2m microscope, equipped with Hilgers FOSSIL MOT. Yttrium–Aluminum–Garnet reflectance standard (Ro = 0.899%) was used for calibration. Moisture and ash yield of each sample were determined following standard procedures (ISO 17246:2010) and ash was subsequently recalculated on dry basis.

Results and discussion

The detailed micropetrographic composition of Bobov Dol coals was recently investigated by

Zdravkov et al. (2024). In short, the studied coal seams are predominantly composed of huminite macerals (seam avg. 83.9–90.6 vol.%; mineral matter-free, mmf), followed by liptinite (seam avg. 7.4–15.2 vol.%, mmf), whereas inertinite is scarce (seam avg. 0.9–2.0 vol.%, mmf; Fig. 1).

Telohuminite (seam avg. 12.4–44.2 vol.%, mmf), represented mostly by gelified and homogenized leaf- and wood-derived ulminite and detrohuminite (seam avg. 30.3–69.2 vol.%; mmf), predominates, although proportions vary significantly between individual samples. Gelohuminite is present in sub-

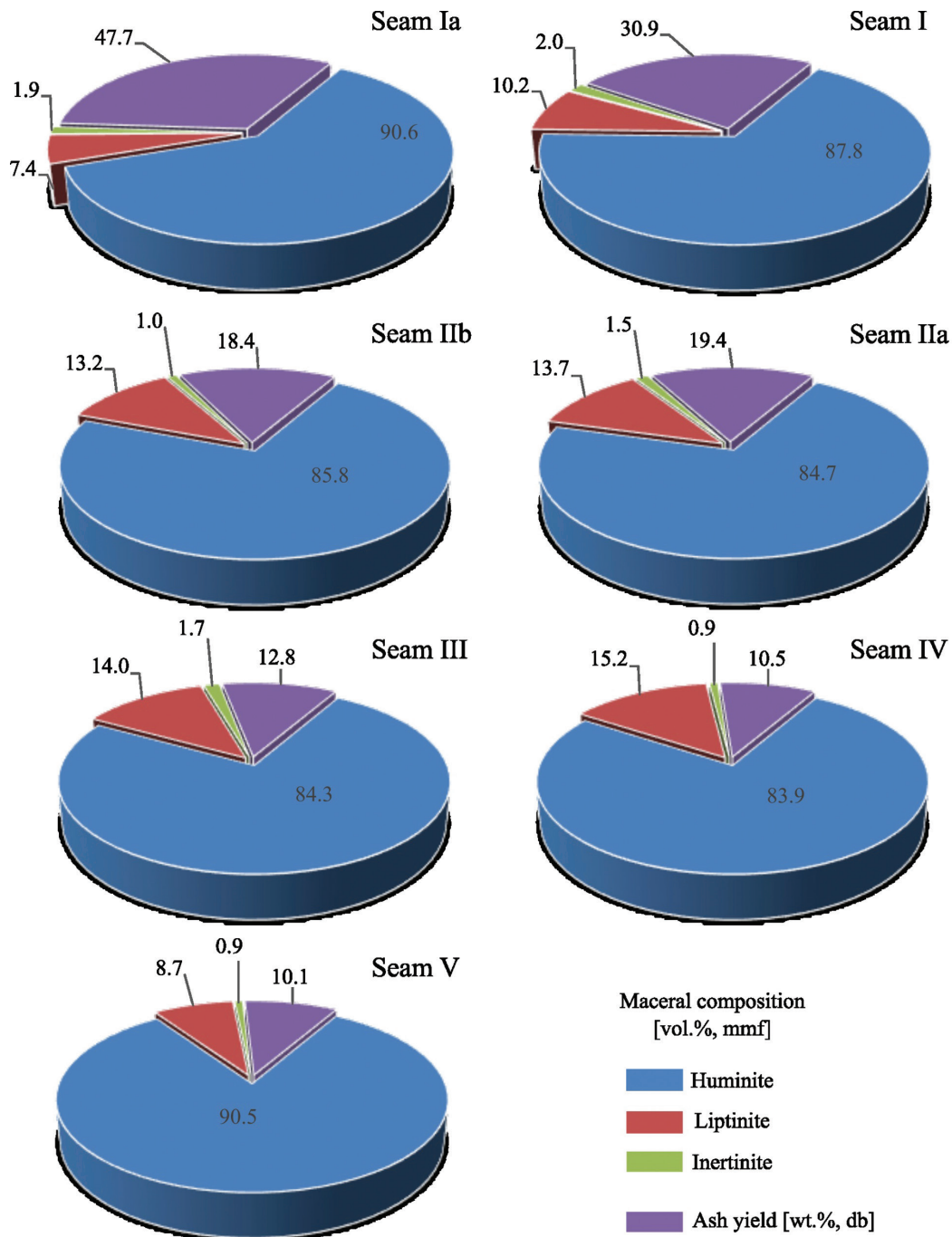


Fig. 1. Seam averaged maceral composition (vol. %, mmf) of Bobov Dol coals

Фиг. 1. Пластово осреднен мацерален състав (об. % на база суха безпепелна маса) на въглищата от Бобовдолския басейн

ordinate amounts (seam avg. 1.1–6.4 vol.%, mmf). The detrital groundmass of the coal is composed of varying proportions of attrinite (seam avg. 17.4–40.8 vol.%, mmf) and densinite (seam avg. 12.7–28.5 vol.%, mmf), depending on the contents of the mineral matter. In accordance with the abundance of leaf- and wood-derived tissues, the liptinite macerals are mostly dominated by cutinite associated

with fluorinite in phyllohuminite, and resinite/exsudatinitite occurring as cell infillings. The inertinite macerals are mostly represented by funginite and locally inertodetrinite. Only in two samples from seams I and III, inertinite is more abundant (up to 13.6 vol.%, mmf).

The measured ulminite reflectance displays significant scatter because of the various degree of tis-

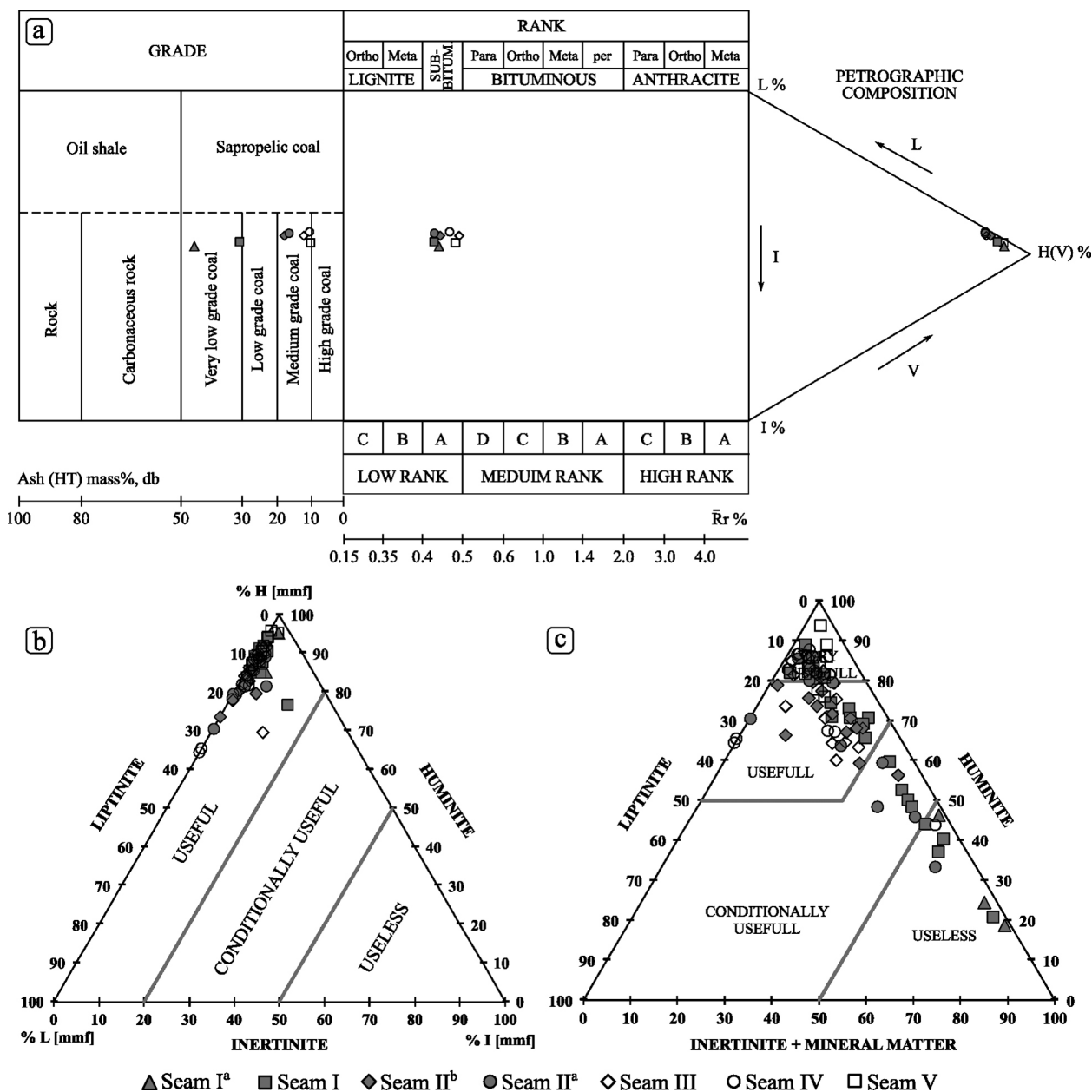


Fig. 2. Seam averaged technological classification of Bobov Dol coals (a, modified after UN. ECE. Committee on Sustainable Energy, 1998; Bielowicz, 2012), triangular diagrams denoting suitability of the coals for gasification based on maceral composition (b) and mineral matter (c, after Bielowicz, 2013)

Фиг. 2. Пластово осреднена технологична класификация на бобовдолските въглища (а по UN. ECE. Committee on Sustainable Energy, 1998; Bielowicz, 2012), триъгълни диаграми въз основа на мацералния състав (b) и минералните примеси (c по Bielowicz, 2013), отразяващи пригодността на въглищата за газификация

sue impregnation with resinous substances (Zdravkov et al., 2024). Seam averaged values, however, vary between 0.43 and 0.5%, confirming the sub-bituminous coalification rank of the coals (Fig. 2a). This data is compatible to the previous technological classification of the coals from seams I, II^{a+b} and IV (Kortenski et al., 2006) and in combination with the average ash yields (10.1–47.7 wt.%; Fig. 1) denotes that the coals from all seams are of medium (seams II^{a+b}, III, IV, V) to very low grade (seams I^a and I) and low coalification rank (Fig. 2a).

From an organic point of view, Bobov Dol coals are dominated by reactive components rich in carbon and oxygen (huminite) and hydrogen (liptinite), suggesting that the coals are suitable for fluidized bed gasification (Fig. 2b) and the low amounts of less reactive to inert macerals (inertinite) will not significantly hamper the gasification process. However, mineral matter should also be taken into account as it does not gasify and thus the overall content of minerals is a limiting factor. Research of Bielowicz (2013) indicates that coals with more than 50% of ash yield are completely unsuitable for gasification and optimal results are achieved for coals with <20% ash. Therefore, considering the higher ash yields in many of the seams I^a and I samples, these will likely not be suitable for gasification (Fig. 2c). Seams II^{a+b}, III, IV and V, on the other hand, contain significantly lower amounts of inorganic matter (<20 wt.%), which renders them more suitable for fluidized-bed gasification. However, it should be noted that the reported average seam ash yields for those seams from the geological exploration of the coal deposit are significantly higher (~35–46 wt.%; (Kamenov, 1959), indicating that the coals will most probably need to be processed in order to reduce the ash content and their gasification will likely strongly depend on the effectiveness of the coal processing. Moreover, the abundance of highly gelified components (i.e., ulminite + densinite) might also have negative effect due to their more homogenous structure and low porosity. It can, therefore, be suggested that the coals might require increased residence time to gasify effectively.

Conclusions

The maceral composition, huminite reflectance and ash yields of seams I^a, I, II^{a+b}, III, IV, and V from the Bobov Dol coal deposit were investigated

and interpreted in order to assess the usefulness for fluidized-bed gasification. The coals from all seams are composed predominantly of huminite and liptinite macerals, whereas inertinite is rare. Based on the average huminite reflectance (0.43–0.49%) and the moderate to high ash yields (10.1–47.7 wt.%), the coals can be classified as medium- to very low grade, low-rank A (sub-bituminous) according to the International classification of in-seam coals. The low rank of the coal, together with their organic composition, renders them suitable for gasification in fluidized-bed reactor. However, when the ash contents are also taken into account, only the coals from seam seams II^{a+b}, III, IV, and V appear to be suitable for this purpose.

Acknowledgments: Financial support from the Bulgarian National Science Fund through project KP-06-H64/5 is greatly acknowledged. Sincere gratitude is expressed to the anonymous reviewers for their critical reviews.

References

- Bielowicz, B. 2012. A new technological classification of low-rank coal on the basis of Polish deposits. – *Fuel*, 96, 497–510; <https://doi.org/10.1016/j.fuel.2011.12.066>.
- Bielowicz, B. 2013. Petrographic composition of Polish lignite and its possible use in a fluidized bed gasification process. – *Intern. J. Coal Geol.*, 116–117, 236–246; <https://doi.org/10.1016/j.coal.2013.07.003>.
- Bielowicz, B., J. Misiak. 2020. The impact of coal's petrographic composition on its suitability for the gasification process: The example of Polish deposits. – *Resources*, 9, 111; <https://doi.org/10.3390/resources9090111>.
- Collot, A. G. 2006. Matching gasification technologies to coal properties. – *Intern. J. Coal Geol.*, 65, 191–212; <https://doi.org/10.1016/j.coal.2005.05.003>.
- ICCP. 1998. New vitrinite classification (ICCP system 1994). – *Fuel*, 77, 349–358; [https://doi.org/10.1016/S0016-2361\(98\)80024-0](https://doi.org/10.1016/S0016-2361(98)80024-0).
- ISO 17246:2010. n.d. Coal – proximate analysis.
- Jordanov, J., J. Kortenski, H. Kazandgiev. 2002. Energy potential of coal basins in Bulgaria. – In: *Intern. Conf. "Corporate Finance Management in Mining Industry"*, Varna, 7–11.10.2002.
- Kamenov, B. 1959. Die Geologie des Bobowdoler Braunkohlenbeckens. – *Ann. Direct. générale des rech. géol., Ser. A*, 8, 1–26.
- Kortenski, J., A. Zdravkov, D. Pinalova. 2006. Applying the international classification of in-seam coals and the international codification system to the coals from Pernik Province. – *Annual of the University of Mining and Geology "St. Ivan Rilski"*, 49, 41–46.
- National Energy Technology Laboratory. n.d. 5.2. Commercial Gasifiers [WWW Document]. <https://netl.doe.gov/>

- research/Coal/energy-systems/gasification/gasifipedia/types-gasifiers.*
- Pickel, W., J. Kus, D. Flores, S. Kalaitzidis, K. Christanis, B. J. Cardott, M. Misz-Kennan, S. Rodrigues, A. Hentschel, M. Hamor-Vido, P. Crosdale, N. Wagner. 2017. Classification of liptinite – ICCP System 1994. – *Intern. J. Coal Geol.*, 169, 40–61; <https://doi.org/10.1016/j.coal.2016.11.004>.
- Sýkorová, I., W. Pickel, K. Christanis, M. Wolf, G.H. Taylor, D. Flores. 2005. Classification of huminite – ICCP System 1994. – *Intern. J. Coal Geol.*, 62, 85–106; <https://doi.org/10.1016/j.coal.2004.06.006>.
- UN. ECE. Committee on Sustainable Energy. 1998. International classification of in-seam coals. New York, Geneva: UN, 41 p.
- Van Dyk, J. C., M. J. Keyser, M. Coertzen. 2006. Syngas production from South African coal sources using Sasol-Lurgi gasifiers. – *Intern. J. Coal Geol.*, 65, 243–253; <https://doi.org/10.1016/j.coal.2005.05.007>.
- Wagner, N. J., M. Coertzen, R. H. Matjie, J. C. van Dyk. 2008. Coal gasification. – In: Suarez-Ruiz, I., J. C. Crelling (Eds.). *Applied Coal Petrology: The Role of Petrology in Coal Utilization*. Academic Press, 408 p.
- Zdravkov, A., A. Bechtel, D. Groß, I. Kojić, K. Stojanović, D. Životić. 2024. Paleovegetation and environment during deposition of the Late Oligocene sub-bituminous coal in the Bobov Dol Basin (SW Bulgaria) as deduced from petrographic and geochemical characteristics. – *Intern. J. Coal Geol.*, 104489, <https://doi.org/10.1016/j.coal.2024.104489>.

Постъпила на 17.05.2024 г., приета за печат на 28.05.2024 г.

Отговорен редактор Йоцо Янев