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Influence of physical and chemical characteristics of coal basin "Kolubara" on soil degradation and environment

Uticaj fizičko-hemijskih karakteristika uglja RB „Kolubara“ na degradaciju zemljišta i životnu sredinu

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Abstract: As part of the paper, an abbreviated technical analysis of coal in the Kolubara basin for three coal-bearing fields: "Veliki Crljeni Field", "Field B and C", "Tamnava-Western Field" was examined. The results of the technical analysis regarding its possibilities to obtain products with the highest possible calorific value through preparation and processing indicated that this coal is characterized by a high content of ash and coke, and lower values of UCV (upper caloric value) and LCV (lower caloric value). In addition to the high percentage of moisture, a larger amount of inorganic substances is present in some samples. The content of macroelements, of heavy metals, and other microelements were examined by "Veliki Crljeni Field". The chemical composition is dominated by Al, Si, Ca and Fe, and S. are more significant. In terms of geochemical characteristics of coal and complete chemical composition, significant differences are represented, both in their distribution in the vertical profile of the main coal seam and laterally. The results show that the differences are particularly pronounced in the contents of some elements, e.g. chromium, manganese, vanadium, arsenic, etc. An analysis of the impact of coal mining on soil degradation and the environment is given.

Keywords: Kolubara basin, coal, ash, moisture, microelements, macroelements, heavy metals, environment.

Sažetak: U okviru rada ispitana je skraćena tehnička analiza uglju u basenu Kolubara za tri ugljonosna polja: PK „Veliki Crljeni“, Polje B i C“, PK „Tamnava-Zapadno polje“. Rezultati tehničke analize u pogledu njegovih mogućnosti da se kroz pripremu i preradu dobiju produkti što veće kalorične vrednosti, ukazali su da se ovaj ugalj odlikuje velikim sadržajem pepela i koksa, a nižim vrednostima GTE (gornja energetska moć) i DTE (donja energetska moć). Pored visokog procenta vlage, u pojedinim uzorcima je prisutna veće količina neorganskih materija. Sadržaj makro-elemenata, teških metala i drugih mikroelemenata, ispitana je PK „Veliki Crljeni“. U hemijskom sastavu dominiraju Al, Si, Ca i Fe, a značajnije je zastupljen i S. U pogledu geohemijskih karakteristika uglja, i kompletnog hemijskog sastava zastupljene su značajne razlike, kako u njihovoj raspodeli u vertikalnom profilu glavnog ugljnog sloja, tako i lateralno. Rezultati pokazuju da su razlike naročito izražene u sadržajima nekih elemenata, npr. hroma, mangana, vanadijuma, arsena i dr. Data je analiza uticaja iskopavanje uglja na degradaciju zemljišta i životnu sredinu.

Ključne reči: basen Kolubara, ugalj, pepeo, vlaga, mikroelementi, makroelementi, teški metali, životna sredina.

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INTRODUCTION

Coal is a widely available, inexpensive, affordable and reliable source of energy. The geopolitical distribution of the world's reserves is in a much better position than the distribution of oil and gas. Europe's lignite production is still competitive with coal imports, but the economic reserves of major European producers are exhaustive. Efforts to more efficiently and thoroughly exploit coal reserves will reduce the EU's overall energy dependency and as a result, coal in Europe will remain the primary energy source by 2030 (Ćalić, Andrić, 2010).

Characteristics of coal

In genetic terms, coal can be humus, derived from the remnants of higher terrestrial plants or sapropellous which originates mainly from lower plants, primarily algae. Coal is almost never pure in the earth's crust; various matter is mixed with a coal substance that reduces its energy (ballast) or pollutes the environment. When decomposed by aerobic conditions, coal loses about 80% of its organic matter, and only 20% remains and participates in synthesis. The elemental composition of the organic mass of coal is characterized by a predominance of carbon (from 65% in brown coals to 98% in anthracite) and a lower amount of oxygen (from 30% to 1%) and hydrogen (from 6% to 1%) (Larry, 2013). Kolubara coal is mostly xylitic and earth-bar, woody structured and with significant content of fusite. Fusite is often mineralized, because its structural openings are filled with clay matter and pyrite. Characteristics of trench coal as a fuel are not the most favorable because, due to the higher content of moisture and minerals, uneven granulation and composition, and therefore low thermal power. Its use is limited to the immediate vicinity of the site. By technological or chemical processing, coal is therefore brought to such a state that it can completely replace the fuels used in high-temperature technological processes, as well as in direct combustion for electricity and heat production (Kleinhans et al., 2018).

Kolubara basin

The Kolubara basin is one of the largest lignite basins in the Republic of Serbia, located south of Belgrade. It began to be exploited around 1910. The first thermal power plant was built in 1936 within the basin, more precisely in Veliki Crljen. The Kolubara part of the basin covers over 600 km^2 of surface. There are excavation fields A, B, C, D, as well as the mines of Vreoci, Veliki Crljeni, Kolubara, Kosmaj, Baroševac, Tamnava etc (Bogdanović i dr., 2007).

The aim of this paper is physico-chemical analysis of coal in three coal fields in the Kolubara basin and environmental impact assessment.

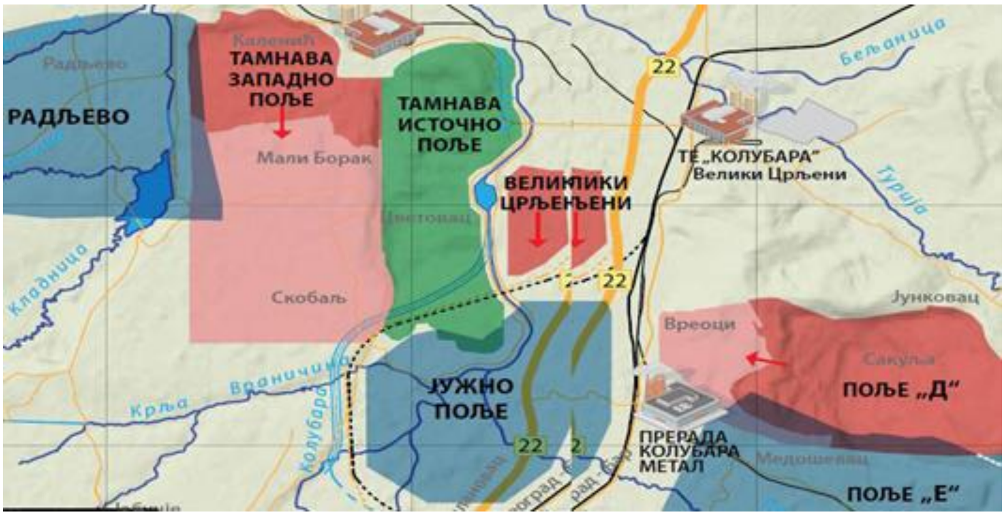

Figure 1- Surface mines in the area of SF „Kolubara" (Source: CINS)

MATERIALS AND METHODS

The coal of the main coal seam of Kolubara was analyzed for three coal-bearing fields: "Veliki Crljeni", "Field B and C", "Tamnava-Westen Field". Coal samples taken from wells represent the entire interval of the drilled layer in the well. A tabular overview of coal quality or analyzed parameters represents the weighted value by the depth of each deposit, all to cover the entire area of the Kolubara basin.

Coal samples from the field "Veliki Crljeni" were taken in the well Qo/Qqr-120, located in the western part of the field, where the exploitable part extends between the eastern border of the surface mine "Tamnava-eastern field" and the line Belgrade-Bar. The main coal seam was drilled in the well in the depth range from 8.90 to 26.10 m. The profile was tested with 4 samples, representing vertical intervals of lengths from 0.60 to 5.00 m.

Surface mine "Field B and C" covers the south-eastern parts of the Kolubara basin. Two coal samples from well S-6/05 were sampled from "Field B and C".

"Tamnava-Western Field" covers the north-northwestern parts of the Kolubara basin. The coal of the main coal seam was tested on 2 wells: Olm-OO-147,5, in the depth interval from 37.00-65.00 m and the well Rgh-Pj/143.75 in the depth interval from 33.60 to 62.00 m. Coal testing was performed at intervals of variable thickness from 2.80 to 8.00 m. A total of 5 coal samples were taken. During sampling, all cores from the exploration wells had natural reservoir moisture, because they affect the determination of coal quality.

There are two ways to express the composition of coal; by technical analysis, where moisture, volatiles, carbon and ash are determined or by elemental analysis, which determines the chemical composition, content of macroelements, heavy metals and other microelements. Elemental analysis also allows the thermal value of some coal to be calculated, and the composition of the relays can to be determined (Yuan et al., 2021).

In the first part of the work, the samples were dried at room temperature, ground and pulverized. The content of coarse, hygroscopic moisture, ash content and thermal power were determined. Technical tests were performed in the laboratory of MB Kolubara, using the standard methods (Tomanec, 2000).

In the second part of the paper, macro and micro-elements in coal are analyzed. All analyzes were done by X-ray fluorescence spectroscopy, using the device ARL Optimix (Thermo Electron Corporation, USA). This method has also been recognized by the American Geological Society for Coal Analysis ("Methods for sampling and inorganic analysis of coal", U.S. Geological Survey Bulletin 1823). Due to very low concentrations, some microelements were determined by atomic absorption spectrophotometry (AAS): Hg, Cd, Th, U, Se, Be, Co, As.

RESULTS AND DISCUSSION

The results of the technical analysis of the coal seam in the "Veliki Crljeni Field", "Field B and C" and "Tamnava-Western Field", are given comparatively in Table 1.

Table 1 - Comparative technical analysis of coal seam in "Veliki Crljeni Field", "Field B and C" and "Tamnava-West Field"

Parameters	"Veliki Crljeni"		"Field B and C"		"Tamnava-Western Field"	
well depth, (m)	8,9-13,9	24,5-25,1	15,5-35,9	30,7-62,2	37,0-65,0	33,6- 62,0
moisture, %	60,80	53,80	53.35	41.46	29,00	17,33
ash, %	5,60	9,00	11.79	31,10	53,33	47,76
coke residue, %	54,26	53,81	48,11	47,32	48,91	*49,44*
UCV, (кJ/кg)	8890	10207	9435	12108	8447	9434
LCV, (кJ/кg)	7064	*8542*	7570	11052	7192	8172

Source: authors results

The amount of ash in coal varies from layer to layer. Coal from "Veliki Crljeni" is characterized by the lowest ash content (15.6% and 9.0%) and the lowest values of UCV (upper caloric value) (8890 kJ/kg) and LCV (lower caloric value) (7064 kJ/kg).

UCV and LCV in the vertical profile of the coal seam progressively grow from the highest to the lowest part and are opposite to the trend of changing the moisture content (negative correlation). UCV and LCV of landfill gas during the biodegradation of municipal solid waste decreases with decreasing humidity (Malešević et al., 2015). The moisture content in the coal from SM "Tamnava-Western Field" shows the lowest value. Free moisture, ash, and sulfur are not the ingredients of fuel. (Goodarzi et al., 2006; Ivković i dr., 2021). Coal from "Field B and C" and coal from SM "Tamnava-West Field" show very close values of coke content (48.11% to 49.44%).

From this, as well as from a number of other analyzed geological coal wells and the corresponding measured trench coal, discrepancies between geological measurements and measurements during exploitation can be observed. The cause of such phenomena is the secondary presence of tailings and this analysis once again confirmed the dependence of the decline in coal quality and its pollution by the presence of tailings (Ahmaruzzaman, 2010).

The forthcoming coal exploitation in this basin has to be viewed through increasingly complex geological conditions, primarily through the increasingly complex geological structure of the coal layers, which are built with an intensive participation of overburden layers with a thickness of more than 1 m. This, in exploitation terms, automatically means an increased participation of "bad lots" coal.

Macro and microelements in coal

The chemical composition of coal, that is, the content of macroelements in coal, in the well profile Qo/Qqr-120 is shown in Table 2.

Table 2 - Chemical composition-content of macronutrients in the coal in the “Veliki Crljeni Field”

Parameters	1	2	3	4	Medium value
depth, (m)	8,90 - 13,90	13,90 -18,50	19,50 - 24,50	24,50 - 26,10	
SiO_2	11.6	22.65	10.42	3.26	11.98
TiO_2	0.14	0.11	0.25	0.01	0.13
Al_2O_3	3.93	5.12	2.87	1.24	3.29
Fe_2O_3	1.47	1.31	2.48	1.33	1.65
MgO	0.18	0.20	0.14	0.11	0.16
CaO	2.65	1.43	1.66	3.30	2.26
K_2O	0.40	0.70	0.23	0.07	0.35
P_2O_5	0.01	<0.01	0.01	0.01	0.01
S	1.09	0.945	2.207	1.602	1.460

Source: authors results

The most abundant component is SiO_2 (3.26-22.65%), and in the lowest part of the coal bed profile it grows fairly progressively and then decreases significantly.

If the highest interval of the layer is excluded, the aluminum content shows the same trend of down enrichment as for silicon (Figure 2).

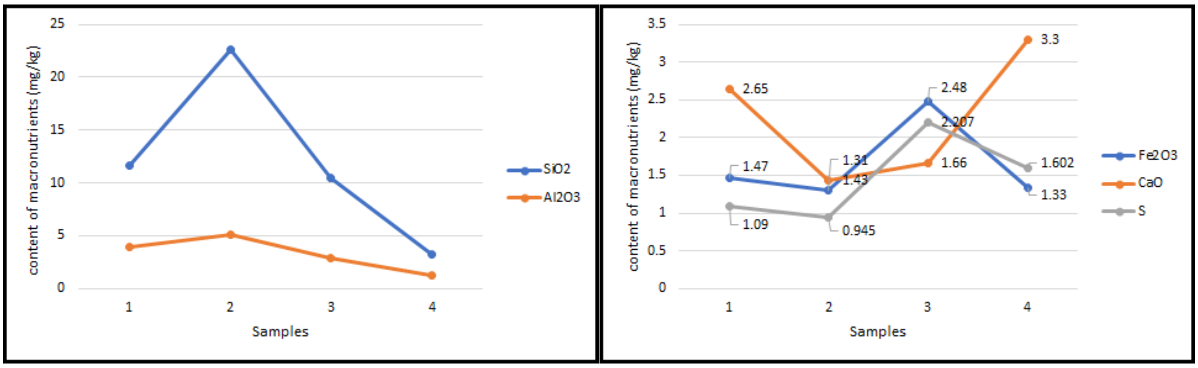

Figure 2 - Change of trace elements concentration with dept
Source: authors results

The contents of S, Fe_2O_3 and CaO correlate very well and they are in smaller quantities in the higher part compared to the lower part. The concentrations of MgO, K_2O, TiO_2 and P_2O_5 are low in all samples (Study, 2010).

The contents of heavy metals and other microelements in the profile of the Qo/Qqr-120 well are shown in Table 3. The values of the microelements by depth show a parabolic arrangement for all elements except Ni and Zn which show linear dependence with respect to depth. Most elements (As, V, Zr, Hg, Pb) have maximum values at 19.50-24.50 m, except Mn, Ni and Zn which have the highest values at 24.50-25.10 m (Pandey et al., 2011).

*Table 3 - Content of heavy metals and other trace elements in co*al in the “Veliki Crljeni Field”*

Parameters	1	2	3	4	medium
depth (m)	8,90-13,90	13,90-18,50	19,50-24,50	24,50-25,10	value
As	55	45	65	45	53
Ba	175	160	170	55	140
Cr	1070	4540	850	195	1664
Cu	85	90	75	65	79
Mn	225	95	85	430	209
Ni	25	20	30	35	28
Sr	95	55	50	45	61
V	35	30	85	20	43
W	< 1	< 1	< 1	< 1	< 1
Zn	65	60	75	80	70
Zr	8	6	9	7	8
Hg	0.31	0.32	0.28	0.27	0.30
Cd	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Pb	48	44	39	38	42
Th	< 1	< 1	< 1	< 1	< 1
U	< 10	< 10	< 10	< 10	< 10
Se	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Be	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Co	7	12	5	9	8
Cl	200	200	200	300	225

Source: authors results

Mercury, chromium and manganese, although occurring in low concentrations, pose a threat to the environment (Wei et al., 2011). All elements except the Mn, Ni and Zn have minimum values at the greatest depth. If the results in depth are viewed solely through minerals, uneven granulation and composition, then the changes in microelements in depth should be constant growth curves (except for Pb) (Životić i dr., 2021).

The existing programs for testing the coal characteristics of the Kolubara mining basin need to be supplemented by testing the specific activity and content of natural radionuclides of uranium (^{238}U and ^{226}Ra) and thorium series (^{232}Th) and potassium (^{40}K) (Kisić i dr., 2013).

Sources of harmful substances in coal exploitation on the environment

Having in mind the characteristics of Kolubara lignite (moisture, ash, LCV, sulfur content) and annual consumption (over 20 million tons), there are increased emissions of gases as well as the impact of deposited ash and slag on aeolian erosion, groundwater, and surface water. In contact with groundwater and the environment, microelements reach the natural water system. By entering the cycle of water movement, they affect the flora, fauna, and humans (Ilić-Stamenković i dr., 2018). The increased emission of gases and particles into the air is contributed by the insufficient efficiency of the system for the separation of particles from the flue gas (electrostatic precipitators), as well as the absence of a flue gas desulphurization plant.

In addition to sulfur dioxide and particles, coal combustion produces other harmful pollutants: nitrogen oxides, carbon monoxide (as a result of incomplete combustion), carbon dioxide, and other gases.

Many years of massive coal exploitation created large quantities of tailings that were landfilled in inappropriate locations. In addition to the depletion of natural resources and pollution of water, air and soil, this fact has also led to significant destruction and degradation of land. Landfills in Serbia are estimated to have between 1.4 and 1.7 billion tonnes of overburden tailings and about 700 million tonnes of flotation and separation tailings.

About 40,000 acres of land have been degraded by surface mines and tailings dumps in large mining basins. Recultivation covered less than 20% of the degraded land area (Đukičin et al., 2014; Krgović, 2015). Considering the technology of coal mining on surface mines, sources of harmful substances or environmental hazards may be considered (Plan, 2017).

CONCLUSION

The data of technical analysis of coal of the main coal seam of Kolubara for three coal-bearing fields are presented and analyzed: "Veliki Crljeni Field", "Field B and C, "Tamnava - Western Field". The test results regarding its ability to obtain products with the highest possible calorific value through preparation and processing indicated that this coal is characterized by the highest content of ash and coke, and the lowest values of UCV and LCV. In addition to the high percentage of moisture, a larger amount of inorganic substances is present in some samples.

Chemical analyzes were performed and the contents of heavy metals and other microelements from the well Qo/Qqr-120 were analyzed, for the "Veliki Crljeni Field".

The chemical composition of coal is dominated by Al, Si, Ca, and Fe, and S. is more significantly represented. In higher concentrations are B, Cr and Mn, St and Cl, and somewhat less Cu, Zn, Pb, As, Ni and others. It is these elements that, with a change in the pH of the environment, can become mobile and bioavailable, where their concentration can increase, become toxic and dangerous to the environment and endanger human health.

Analytical results are important for assessing the possibility of using certain coals as raw materials for obtaining artificial fuels (coal powder, coke, semi-coke, and others). These tests will be of particular importance for the design of block coal mining on the floors of surface mines as well as for assessing the impact of coal combustion and combustion products on the environment.

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