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THE POTENTIAL FOR CARBON SEQUESTRATION IN RECLAIMED MINE SOIL

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ABSTRACT

Mining sector has been the central attention of the business and public policy sustainable development scheme for several years. Reclamation territories are with potential carbon sequestration capacity in degraded mining areas and can be an impeccable option for achieving sustainable development goal-13. This paper made an investigation about the presence of heavy metals in reclaimed area possibilities to improve reclaimed soils with biochar with idea to enhance ecosystem carbon pool and atmospheric CO_2 sequestration capacity to offset CO_2 emission and soil organic carbon losses. The results show that the carbon content in biochar is 46.6 % and would be classified in class 2 according to the International Biochar Initiative. H/C and O/C atomic ratios could be useful for restoration of degraded poor mine soils through enhanced carbon sequestration. O/C ratio is 0.085 mol mol^{-1} and half-life of biochar is 2795.65 years, which confirm the biochar stability. For each kilogram of biochar incorporated into the soil, 144 g of carbon were sequestrated 52,8 % CO_2 emission reduction. Hence, the biochar developed in this study can be used in carbon-deficient soils with the added benefit of long-term carbon storage.

Keywords: *biocarbon, biochar, recultivation, industrial biowastes, treatment technologies.*

INTRODUCTION

The mining industry is one of the significant industrial sectors. Regardless of the restructuring processes of the mining sector and the economic crisis, it continues to play a determinant role in Bulgaria. The mining industry is a major sector of the country's economy, and the value of the production is about BGN 2 billion [1, 2]. According to the data of the Bulgarian Chamber of Mining and Geology, there are 595 deposits established in the country [3]. Exploration, extraction and the initial mineral processing of underground resources leave negative consequences and pollute the earth's womb and the environment. A serious problem in the mining and initial mineral processing of underground resources is the generation of huge amounts of waste (earth-rock and slime-like materials) [4]. In contemporary dimensions of the mining production the problem of interaction of the environment is very important. The contradiction between the ever-increasing needs of raw materials and the limited reserves of natural raw materials is deepening [5 - 8].

Depending on the useful elements and the degree of their extraction from the raw material, as well as their chemical composition, the waste from ore raw materials accumulates in the natural objects and is transferred along the food chains. The total size of disturbed lands from exploitation in the country exceeds 237.9 thousand decares. The total number of tailing dams in Bulgaria is 33, 21 of which have been suspended from operation and partially recultivated. 34 % of the total disturbed areas have been reclaimed [1 - 4].

These disturbed areas, where reclaimed mine soils are developing, may play significant role for potential of carbon sequestration in new ecosystems, especially in local range on areas disturbed by human activity [9]. A potential approach to mitigating the rising CO_2 concentration is to enhance sequestration of carbon in terrestrial ecosystems CO_2 increasing biomass productivity and allocating the assimilated carbon into long-lived plant and soil organic matter CO_2 [10, 11]. Carbon sequestration of the mine soils is high in the first 20 years after reclamation in the topsoil depth [9]. In general, higher rates of carbon sequestration are observed for mine soils under pasture and grassland management than under forest land use [9 - 12]. Proper reclamation and post reclamation management may enhance carbon sequestration and add to the economic value of the mined sites [9]. Management practices that may enhance carbon sequestration include increasing vegetative cover by deep-rooted perennial vegetation and afforestation, improving soil fertility, and alleviation of physical, chemical, and biological limitations by fertilizers and soil amendments such as biosolids, manure, coal combustion by-products, and mulches [12 - 14]. When listing nutrients in disturbed terrains, carbon is listed first, and this is no accident. On average, plants contain about 45 % carbon. CO_2, which is the main source of carbon, contains 27.3 % carbon, or 1.6 kg of CO_2 is needed to build 1 kg of dry plant mass. In addition to carbon, oxygen, and hydrogen, which are obtained from CO_2 from the air and water, plants also need many elements for their development, which they take from the soil. The available amounts of these nutrients determine soil fertility. Carbon provides microorganisms with energy as well as aids growth. The principle of using biochar for carbon (C) sequestration is related to the role of soils in the C-cycle [10, 14, 15] This means that biochar allows carbon input into soil to be increased greatly compared to the carbon output through soil microbial respiration, and this is the basis behind biochar's possible carbon negativity and hence its potential for climate change mitigation Concomitant with carbon sequestration, biochar is intended to improve soil properties and soil functioning relevant to agronomic and environmental performance [9, 11, 15, 16].

Biochar, produced from biomass, which is in abundance, fit to this management while being a form of waste disposal and recycling [17, 18]. Biochar has a number of specific functions in the natural environment that will be beneficial to prevent global warming and also to increase the functionality of soils by an adaptation of the current biological carbon cycle, the biochar is produced from biomass and half is returned to the soil as charcoal [10, 14, 15, 19]. While biochar is being applied to soils for the conditioning and fertilization purposes, this application can also be beneficial in reduction of toxic components. Studies have shown that biochar is also capable of absorbing metals, and organics that contaminate soils which harm people, plants and animals [12]. This is because biochar as an additive to a soil can be expected to improve its overall absorption capacity impacting toxicity because there is a decrease in transportability and depletion of the presence of metal or organic compounds [10, 11].

To achieve climate neutrality by 2050, it will be necessary to use the full potential of soils to mitigate and adapt to climate change. Increasing soil carbon through techniques is a relatively inexpensive solution. Research estimates that carbon farming costs 10-100 \$ t^{-1} CO_2 removed, compared to 100-1000 \$ t^{-1} CO_2 removed for technologies that mechanically remove carbon from the atmosphere. The restoration of soil organic carbon by implementing integrated practices for sustainable soil management and creating conditions for mitigating the causes and consequences of climate change will contribute to the final achievement of this ambitious goal [5 - 8, 11].

EXPERIMENTAL

Materials

Samples were taken from tailings dams, which are a consequence of the activities of a leading company in the mining industry of Bulgaria. The object is divided into two subobjects and includes the reclamation of about 2500 decares. Tailing dam 1 has a volume of 150 million tons; it is closed and reclaimed and sampled (B1-1, B1-2, B1-3, B1-4, B1-5 illustrated in Fig. 1), while tailing dam 2 is in operation, but 290 decares of its air slope were also reclaimed, from which samples were also taken (B2-1, B2-2, B2-3, B2-4, B2-5, illustrated in Fig. 2). It has a project volume of 258 million m^3.

Biochar production

Biochar was produced from poultry litter feedstock and wood ash from biomass in 1:1 ratio. The product was thermally treated by pyrolysis process at 450°C.

Fig. 1. Tailings dam - 1.

Fig. 2. Tailings dam - 2.

Methods

Analyzes were performed on an ICP-Atomic emission spectrometer (High Dispersion ICP-OES Prodigy of the Teledyne Leeman Labs USA company with dual plasma monitoring (axial and/or radial)). The construction of the optical system is "Eshelle" and the detector "L-PAD", with high resolution (0.007nm), continuous spectral range (from 165 to 1100 nm). The spectrometer is equipped with a so-called "free running" RF generator, which provides power up to 2 kW at 40.68 MHz.

Quantitative elemental analysis of dry soil samples for the content of carbon, nitrogen and hydrogen was carried out with an automatic analyzer EA 3000 of the Italian company EuroVector. The method of analysis includes the sample burning at a high temperature (980 - 1100°C) and determining the component by gas chromatography.

The shape and size of particles in the samples were studied using a scanning electron microscope (SEM) Philips PH Model 515, operating in regime of secondary electron emission (SEE).

Biochar Stability

Based on the proximate analysis data, the H/C and O/C atomic ratios for the biochar were estimated by using eqs. 1 and 2 [17]:

$$H/C = 0.397 \times VM / FC + 0.251 \quad (1)$$

$$O/C = 0.188 \times Vm / FC + 0.035 \quad (2)$$

where *FC* is the percentage of fixed carbon content and *VM* is the percentage of volatile matter content in the biochar.

Mean residence time and the percent of carbon that would remain in the soil after100 years (BC+100) was calculated according to eqs. 3 and 4 [18 - 20]:

$$MRT = 4501 \times e^{-3.2\times(H/C)} \quad (3)$$

$$BC+100 = 1.05 - 0.616 \times (H/C) \quad (4)$$

where *MRT* and BC + 100 are mean residence time (*MRT* expressed in years) and the % of carbon that would remain in the soil after 100 years *(BC + 100)*, respectively. *H/C* stands for the atomic ratio of the biochar. The letter 'e' represents the term exponential.

Biochar characterization, Carbon sequestration and CO_2 reduction potential

The final stage in the characterization after conversion of the biomass to biochar was calculated the amount of carbon that would be captured by each biomaterial if the experimental conversion was applied. The equations below were used for the calculation of total biochar yield (eq. 5), fixed carbon content (eq. 6), total potential carbon in biochar (eq. 7), carbon sequestrated (eq. 8) CO_2 reduced (eq. 9) and the CO_2 reduction potential, respectively [18 - 21]:

$$Yield\ of\ biochar = \frac{Weight\ of\ biochar\ produced}{weight\ of\ initial\ pyrolysis\ feedstock} \quad (5)$$

$$Total\ Potencial\ Carbon = Total\ biochair\ produced \times \times \%Fixed\ carbon \quad (6)$$

$$Carbon\ sequestrated - Total\ Potential\ carbon \times \times Carbon\ stability\ of\ 80\% \quad (7)$$

CO_2 Reduction Potential = Carbon sequestered amount × 44/12 (8)

RESULTS AND DISCUSSION

Soil characteristic analysis

Table 1 presents the obtained results from chemical analysis of micro- and macro-nutrients, including heavy metals. The chemical analysis shows that all soils samples contain sufficient concentrations of potassium and calcium. The measured concentration of heavy metals in the mixture is much lower than that of limit value [6].

Although the soil materials from the reclaimed sites are very poor in humus and nitrogen, they still play a role in protecting the surrounding areas from contamination with heavy metals and other aggressive ions from the embankments. It should be noted that the soils in the area are naturally enriched with copper, chromium, and arsenic. The mobile forms of heavy metals do not exceed the control values for the protection of soil microorganisms, plant growth and soil filtration waters [4 - 6].

Soil health assessments. Along with nitrogen, phosphorus is a limiting nutrient in soil. Soils limited in phosphorus like this case reduce plant growth and development. Phosphorus availability generally increases with increasing soil organic matter since phosphorus is released through mineralization of organic matter. It is therefore advisable to introduce a mechanism to nourish this soil with organic matter and increase the possible mobility of phosphorus. In the samples there are not enough concentrations of aluminum, iron, and calcium to have high phosphorus retention capacity. The other problem is pH which is low and the amounts of aluminum and iron form very

Table 1. Results of chemical analysis.

Component, mg kg^{-1}	Sample No									
	B1-1	B1-2	B1-3	B1-4	B1-5	B2-1	B2-2	B2-3	B2-4	B2-5
Al	19.93	20.02	23.99	23.04	13.28	18.56	22.04	18.49	15.47	14.52
Ba	0.11	0.07	0.15	0.12	0.05	0.10	0.15	0.10	0.10	0.07
Ca	5.92	5.60	4.43	4.05	3.34	5.11	5.49	6.92	10.60	5.83
Cr	0.03	0.08	0.04	0.03	0.03	0.03	0.03	0.03	0.03	0.03
Cu	0.04	0.03	0.04	0.05	0.27	0.01	0.03	0.01	0.02	0.06
Fe	25.97	25.85	35.33	36.30	24.06	26.61	39.06	24.61	24.04	24.15
Mg	8.98	12.14	9.63	10.57	8.40	9.73	10.73	7.18	13.05	10.23
Mn	0.50	0.47	0.48	0.44	0.42	0.81	0.52	0.52	0.37	0.38
Ni	0.03	0.03	0.03	0.03	0.02	0.02	0.03	0.03	0.02	0.02
K	2.23	1.79	4.12	5.83	2.97	1.89	6.48	1.28	2.65	2.12
Na	1.09	0.88	0.84	0.73	0.75	0.56	0.46	0.78	0.84	0.51
Zn	0.09	0.06	0.10	0.09	0.04	0.05	0.15	0.10	0.14	0.07
Mo	0.02	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01
P	0.05	0.09	0.55	0.42	0.44	0.40	0.72	0.15	0.41	0.44
V	0.07	0.09	0.07	0.07	0.07	0.07	0.07	0.08	0.07	0.07
Co	0.01	0.02	0.02	0.02	0.01	0.02	0.02	0.01	0.01	0.01
S	0.51	0.45	0.49	0.46	0.43	0.37	0.36	0.41	0.38	0.33
B	0.13	0.08	0.11	0.08	0.14	0.14	0.03	0.30	0.44	0.09
Re	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1

strong bonds with phosphate. Calcium, magnesium, and potassium are essential plant nutrients since they are involved in a variety of plant functions and metabolic processes but again are not with optimal concentrations to have health soil.

Some trace metals are essential micronutrients (e.g. boron, zinc, manganese, copper, molybdenum) but in low concentration they are important for soil biota. On samples there is very low concentration of these elements which is not good for soil structure.

The measurement of carbon, nitrogen and hydrogen aims to analyze the distribution of structural elements in the soil, as well as the C/N and C/H ratios, which are indicative of the ongoing processes in the soil (Table 2) and have a great influence on the rate of degradation of the organic material. Depending on what is the ratio between carbon and nitrogen in these organic materials, in addition to mineralization, biological fixation or immobilization of mineral nitrogen in the soil can occur. In the results obtained, critical values of nitrogen and ratios are observed and cannot fully satisfy the needs of microorganisms, and hence the proper breakdown of nutrients for plant nutrition. The low content in the C/N ratio can be related to the higher content of alkynes, alkenes, alkanes and other compounds. C/N is a dynamic ratio that explains the co-utilization of C and N by soil microbes. In this case, this ratio is much lower, which can be explained by the mineralization of the organic material included in the soil. To normalize the balance, it is necessary to feed the soil with carbon-containing improvers such as biocarbon, for example.

Biochar characterization

The presence of biochar in the soil system affects structure, porosity, bulk surface area, pore distribution, and particle size. The characteristics of biochar (chemical composition, surface chemistry, particle distribution and pore size) as well as physico-chemical stabilization determine its effect on soil functions. Introducing a well-grounded system for sustainable development at the regional level and creating economically stimuli for the utilization of waste biomass leads to a positive assessment and reduction of the consequences of droughts, erosion, and destruction. The outlined trends of losses in the content of organic carbon, humus, and highly disturbed soil microbiology, despite the reclamation, will manifest in the future as a trend of loss of soil fertility. If periodic feeding of reclaimed land is introduced, using an organic waste improver, waste will be significantly reduced, and bio-carbon will be utilized to be put into such threatened areas. Fig. 3 shows such an object - biochar obtained from the utilization of bird excrements and fly ash [7, 8]. The microscopic picture clearly shows the high porosity and internal surface of the developed soil improver. The development and introduction of integrated measures and practices to

Table 2. Results from elemental analysis.

Sample	Element, %			Ratios	
No	Nitrogen, N	Carbon, C	Hydrogen, H	C/N	C/H
B1-1	0.19	0.56	0.71	2.94	0.78
B1-2	0.24	0.84	1.06	3.5	0.79
B1-3	0.33	1.54	0.73	4.6	2.11
B1-4	0.26	1.00	0.88	3.84	1.14
B1-5	0.18	0.48	0.42	2.08	1.14
B2-1	0.23	0.36	0.93	1.57	0.39
B2-2	0.19	0.56	0.75	2.95	0.75
B2-3	0.20	0.47	1.08	2.35	0.44
B2-4	0.16	0.33	0.62	2.06	0.53
B2-5	0.17	0.25	0.67	1.47	0.37

Table 3. Results from biochar analysis from feedstock (litter and wood biomass) in pyrolysis condition (350 - 500°C).

Sample	Element, %			Ratios			
	Nitrogen, N	Carbon, C	Hydrogen, H	C/N	C/H	O/C	VM/FC
Biochar	2.38	46.6	6.9	19/1	7/1	0.085	0.266

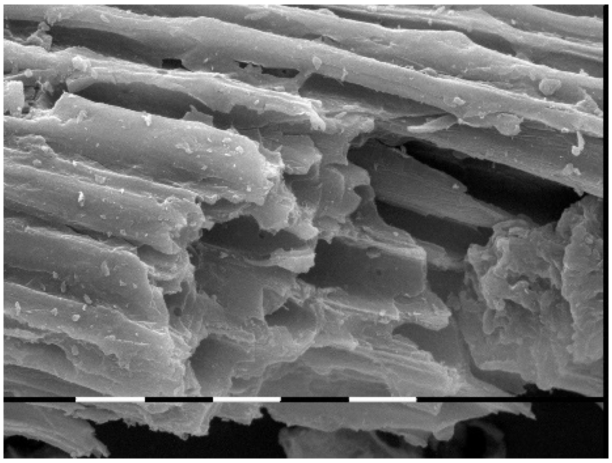

Fig. 3. SEM of biochar on base of poultry litter and biomass ash.

create conditions for sustainable management of the main components of the environment to guarantee soil fertility, stable moisture storage, and maintenance of soil microflora, including improving the well-being of recultivated tailing dams is of special importance. So, the introduction of decentralized practices and systems for using waste and obtaining biochar will provide a basis for the accumulation and storage of organic carbon in soils, to reduce on the one hand the amount of generated carbon oxide, methane and nitrous oxide in the atmosphere and on the other hand to creating an opportunity for rapid restoration of disturbed soils.

Biochar Stability

The mineral content of the feedstock is largely retained in the resulting biochar, where it concentrates due to the gradual loss of C, hydrogen (H) and oxygen (O) during processing. Table 3 provides the elemental composition of representative feedstocks mixture.

The carbon content in biochar is 46.6 %. According to International biochar initiative [21], biochar with organic carbon content between 30 % and 60 % would be classified in class 2. Atomic ratios (mainly O/C) (eq. 2) are used to estimate the biochar content of carbon stability. It was found relationship between biochar half-life and O/C ratio with an O/C ratio of < 0.2 mol mol^{-1} the half-life would be > 1000 y [19]. In this case O/C ratio is 0.085 mol mol^{-1} and half-life of biochar is 2795.65 years (eq. 3), which confirm a biochar stability. The estimated MRT and the mass fraction of carbon that would remain after 100 years were greater than 1000 years and 80 % (96 % - eq. 4), respectively, for biochar with a H/C ratio of 0.14 (eq.1) [19]. As per European biochar certificate [20] and IBI Guidelines [21] biochar with H/C < 0.7 and O/C < 0.4 will be effective. In both cases the result has coincidence with the requirement. Hence, the biochar developed in this study could be used in carbon-deficient soils with the added benefit of long-term carbon storage.

Carbon sequestration and CO_2 reduction potential

The percentage of biochar yield after pyrolysis is 49.9 %. (eq. 5) This result is dependent on the pyrolysis temperature and thermal decomposition of organic matter [18]. The results in Table 4 shows that for each kilogram of biochar incorporated into the soil, 144 g (eq. 6) of carbon were sequestrated 52.8 % CO_2 emission reduction. Total potential carbon in biochar - 268 g of C/kg of feedstock (eq. 6) and CO_2 reduction potential - 771,84 CO_2 eq. kg^{-1} of biochar (eq. 8), has a stable form of carbon and could be stable for microbial

Table 4. Results from biochar characterization, stability, carbon sequestration and CO_2 reduction potential.

Parameters	Biochar results
Biochar stability	
Mean resident time (years)	2795.65
BC_{+100} (%)	96
Statistical data [10,12]	
Manure (million tone/year)	900
Total biochar (million tons)	330.21
Total potential carbon in biochar (million tons)	108.90
CO_2 reduction (million tons)	319.64
Carbon sequestration and CO2 reduction potentia*l*	
Biochar for carbon sequestration kg biochar/kg feedstock (litter + wood biomass)	0.494
Total biochar yield potential (kg of biochar/kg feedstock)	0.498
Fixed carbon content (kg of fixed carbon/kg of biochar)	0.539
Total potential carbon in biochar (g of C/kg of feedstock)	268
Carbon sequestered (g of C sequestered/kg of feedstock)	144
CO_2 reduced (%)	52.8
Total potential carbon	15387.786
CO_2 reduction potential (CO_2 q.kg^{-1})	771.84

decomposition and store atmospheric CO_2 in soil. Otherwise in plant would have been rapidly mineralized to carbon dioxide, that has emitted in the atmosphere. In this investigation has proven that the biochar is stable, with resistant to degradation carbon, and could be exploited as an agent for long-term carbon storage in soil as a climate change mitigation option.

CONCLUSIONS

This study presents the feasibility of producing biochar who has a good property and fulfilled key quality criteria for soil carbon sequestration, The carbon content in biochar is 46.6 %. and would be classified in class 2 according to the International Biochar Initiative. H/C and O/C atomic ratios could be useful for restoration of degraded poor mine soils through enhanced carbon sequestration. O/C ratio is 0.085 mol mol^{-1} and half-life of biochar is 2795.65 years, which confirm a biochar stability. The results show that for each kilogram of biochar incorporated into the soil, 144 g of carbon were sequestrated 52.8 % CO_2 emission reduction. Total potential carbon in biochar - 268 g of C/kg of feedstock and CO_2 reduction potential – 771.84 CO_2 eq. kg^{-1} of biochar, has a stable form of carbon and could be stable for microbial decomposition and store atmospheric CO_2 in soil. Hence, the biochar developed in this study could be used in carbon-deficient soils with the added benefit of long-term carbon storage.

Bulgaria is actively working on plans to achieve climate neutrality. A significant effect of the introduction of resource utilization through carbonization is the use of the amounts of bio-carbon obtained for the implementation of the future environmental policy and system for mitigating the causes and adapting to expected climate changes. Proper reclamation and post reclamation management may enhance carbon sequestration and add to the economic value of the mined sites. The proposed practice could be the key to increasing the storage of carbon, which can be preserved for a long time in the soil.

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