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Study of the Possibility of Biorecultivation of Soils Contaminated with Brown Coal Waste

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ABSTRACT

Environmental pollution by industrial waste, including brown coal mining, is one of the environmental problems of many countries. To improve the ecological situation in the region, it is necessary to recultivate soils contaminated with brown coal waste. The goal of the study was to study the possibility of biorecultivation of soils polluted with brown coal waste from the Lenger deposit located in the south of Kazakhstan. It was found that the inorganic part of the brown coal waste is represented by minerals: Quartz SiO_2, Gypsum $CaSO_4 \times 2H_2O$, Kaolinite $Al_2Si_2O_5(OH)_4$, Cronstedtite $Fe_3((Si_{0.74}\ Fe_{0.26})_2O_5)(OH)_4$, Margarite $CaAl_2(Si_2Al_2)O_{10}(OH)_2$, Muscovite $H_2KAl_3(SiO_4)_3$, Calcite $CaCO_3$, Laumontite $CaAl_2Si_4O_{12}(H_2O)_2$, Lead Aluminium Sulfate Hydroxide $Pb_{0.5}Al_3(SO_4)_2(OH)_6$, with quartz content in samples in the range of 61.5–92.9%. The organic part of the waste is 90.0% made up of humic acids and fulvic acids. It has been established that heterotrophic, cellulolytic microflora and micromycetes are represented by the genera *Rhodococcus, Bacillus, Pseudomonas, Penicillium; Trichoderma, Dietzia*, promising for biorecultivation purposes. The phytocenosis of coal waste dumps is composed of toxicotolerant ruderal plant species of the local flora: *Centaurea scabiosa* L., *Centaurea iberica Trev., Cichorium intybus* Linn*., Cousinia cyrdariensis Kult., Achilleamille folium* L*., Thlaspiar vense* L*., Arctiumto mentosum* Mill*., Onopórduma cánthium L., Agropyron cristatum* L*., Phlum pratense* L*., Erytrigia repens* L*., Nevski/Agropyron repens* L*.,Cynodon dactylon* L*., Capparis spinosa* L*., Polygonum aviculare* L*., Dodartia orientalis* L*., Althaea officinales* L*., Alhagipseud alhagi* (Bieb.)Desv*., Peganum harmala* L. For biorecultivation of soils contaminated with brown coal waste, an algorithm of work has been developed, including the use of soil blocks with integrated seeds of toxicotolerant plants inoculated with microorganisms.

Keywords: brown coals, waste, biorecultivation, mineralogical composition, anthropogenic phytocenosis, toxicotolerant plant species.

INTRODUCTION

Coal has long been used as a fossil fuel and as a raw material for further chemical processing. Brown coal, as well as its waste, are cheap and easily accessible sources of energy for the electric power industry [Lehmann, 2007]. By its origin, brown coals are caustobiolites, a carbonaceous sedimentary rock of organic origin that has undergone carbon enrichment. According to Symonowicz and Toczko (2023), brown coals, by their origin, are caustobiolites, i.e. carbonaceous sedimentary rock that has undergone carbon enrichment. At the same time, the following stages of modification are monitored: the initial basis is vegetation as a carbon-forming material; then peat is formed, gradually passing into the stage of brown coal; the next stage is the formation of stony coal, ending with the formation of anthracite. To date, the most intensive research is being conducted on the conversion of coals into various liquid products. To date, the study of the peculiarities of modifications of brown coal and its waste is one of the important issues of environmental protection in the areas of mining and processing of minerals. The organic matter of coals is a set of valuable chemical products. Despite the changes occurring with the organic mass of coals in the process of carbon formation, a significant part of the structural units of coals retains the characteristics of the starting substances.

Due to the emission of pollutants as a result of meteorological processes and phenomena, coal dumps are sources of pollution of underground aquifers and surface water bodies. Land disturbance occurs during the development and storage of waste that is withdrawn from land use. The overburden rock produced during the development of deposits containing coal is placed in dumps throughout the entire period of operation of the deposits. According to statistics, there has been an increase in the volume of contaminated soils [Eremeeva, 2023]. On the other hand, the area of recultivated land, on the contrary, decreases. Waste accumulated as a result of coal mining is also growing. It was found that the volume of emissions of toxic substances increased by 12.5%, and the number of neutralized toxicants decreased by 55.4%. The same pattern was revealed for disturbed lands, the area of which increased by 154.0%, with an increase in the volume of accumulated waste in the areas of brown coal mining – by 30.0%, and the volume of reclamation decreased by 42%. Environmental pollution at the local level eventually turns into regional pollution. Soil contamination with various toxic elements poses a serious threat to humanity, affecting not only food security, but also human health, entering the body in different ways [Bećirović, 2015; Ngozi, Arihilam, 2019]. In the modern practice of the development of coal-mining countries, In the modern practice of the development of coal-mining countries, special attention is paid to the development of new technologies for the restoration of disturbed ecosystems. The use of modern reclamation technologies based on the use of unconventional meliorants is promising. The use of unconventional meliorants allows to increase the productivity of reclamation works and significantly reduce the cost of creating a nutrient layer.

For example, the effectiveness of foliar treatment of plants with a suspension of sapropel, vermicompost and diatomite has been shown [Gazizov et al., 2021]. It was found that the acid-hydrolysis process of decomposition of peat, sapropel and brown coal can simulate the native processes of their modification under the influence of biotic and abiotic factors [Sokolov, 2008]. It was revealed that the greatest nitrogen intake is characteristic of the fertilizer obtained on the basis of brown coal. It is shown that by its nature, the main hydrolysate of humic acid obtained from brown coal differs from soil humic acids [Allard, 2006]. Bound lipids isolated from brown coal consisted almost exclusively of aliphatic components, mainly with a predominance of long-chain alkane acids. According to the discovered fragments of lignin consisting of vanillin and 4-hydroxybenzoic acids, it was found that the processes of conversion of lignin into humic acid occur more intensively in brown coal waste. This efficiency of brown coal modification contributes to the recommendation of the use of humic acid obtained from brown coal waste in the agro-industrial complex, which reliably leads to a significant increase in potato yield, tuber yield and the quality of rhizobacteria such as *Bacillus megaterium* and *Bacillus subtilis*, which promote plant growth [Ekin, 2019, Eswaran, 2021]. Optimal doses of organomineral fertilizers obtained from brown coal waste to increase corn yield were determined [Symanowicz, 2022].

The research results [Mikos-Szymanska, 2019] tested the hypothesis that granulation of synthetic nitrogen fertilizer (urea) with a natural organic nitrogen source (brown coal) will reduce nitrogen losses from the soil system. Brown coal pellets enriched with urea were simultaneously molded and dried in an experimental dryer with superheated steam. After application to unseeded soil columns, granules of urea and brown coal reduced nitrous oxide emissions by 40%, reduced leaching of mineral nitrogen and maintained a higher nitrogen level in the upper soil layer compared to ordinary urea in its pure form.

It has been established that the influence of brown coal waste on the biochemical qualities of the soil is enhanced in combination with various groups of soil microorganisms. A number of microorganisms [Akimbekov, 2020] are able to secrete humic organic matter by biotransformation of coal. Micromycetes of the genera *Trichoderma, Cladosporium Penicillium, Epicoccum, Metarhizium* are known to be capable of transforming brown coal hydrocarbons [Pokorný, 2005]. The role of higher plants in creating and maintaining a microclimate for the augmented soil microflora during biorecultivation works is known [Chakravarty, 2019]. The search for ways to recultivate soils contaminated with brown coal waste, which represent a source of environmental pollution, is extremely important. The purpose of this study was to study the possibility of biorecultivation of soils in the areas of brown coal waste storage located on the territory of Southern Kazakhstan.

MATERIALS AND METHODS

The objects of the study were brown coal waste located on the territory of the city of Langer in the south of Kazakhstan (Fig. 1). Due to the sprawl of the city, some waste storage sites were either in the city or on the border with residential areas.

Brown coal waste storage sites are hills from 15.0 to 50.0 m, black or dark gray in color. Visually, the waste was divided into two groups: A-dense black waste, visually similar to coal, lamellar, with a characteristic luster; B-grayish, porous, medium-density waste, at a depth of 5–10 cm and deeper similar to slags.

Sampling was carried out according to the requirements of GOST 33770-2016 from the waste storage area and at certain distances from it. For X-Ray Fluorescence analysis (XRF) spectrometer Shimadzu IRPrestige was used [Longoni 2006].

Diffractometric analysis of the crystal part of the samples was carried out on the D8 Advance (Bruker) apparatus. The obtained diffractogram data were decoded using EVA software. The values of samples and phases were determined by the Search/match program using the PDF-2 database. The above phase analysis results relate only to the crystalline part of the sample under study.

Isolation of microorganisms was carried out on the following nutrient media: cellulolytic bacteria – on the Getchinson medium, g/l: distilled water – 1.0 l; $CaCl_2$ – 0.1; $NaNO_3$ – 2.5; K_2HPO_4 – 1.0; $FeCl_3$ – 0.01; $MgSO_4 \times 7H_2O$ – 0.3; NaCl – 0.1; pH of the medium is adjusted to 7.2, adding 20% Na_2CO_3 solution, agar-20.0; heterotrophic bacteria on medium MPA, g/l: meat broth – 1.0 l, peptone – 10.0; NaCl – 5.0; agar–20.0; pH 7.3±0.2; micromycetes on medium Chapek, g/l: distilled water to 1.0 l, sucrose – 30.0; peptone – 5.0; yeast extract – 2.0; $NaNO_3$ – 3.0; K_2HPO_4 – 1.0; KCl – 0.5; $MgSO_4$ – 0.5; $FeSO_4 \times 7H_2O$ – 0.01; agar – 20.0.

Identification of bacteria to the species was carried out with determinants [Bergey, 2001]. Microscopy of microbiological preparations was carried out using microscopes "Mikmed-5" (Russia), "Tayda" (Japan) at magnification ×40, ×600, ×1000, electron scanning microscope at magnifications up to ×3300 times.

The taxonomic analysis of flora was carried out using the plant determinant "Flora of Kazakhstan" (1969). The assessment of the abundance of vegetation in the studied areas was carried out according to the Drude scale in the modification, where the abundance of the species was determined by the following criteria: Soc-entirely, Sor1-quite-abundantly, Sor2-abundantly, Sor3-veryabundantly, Sp-rarely, Sol-singly, Un-in one copy.

Statistical processing of the obtained research data was carried out to determine the arithmetic mean with standard deviations. The studies were

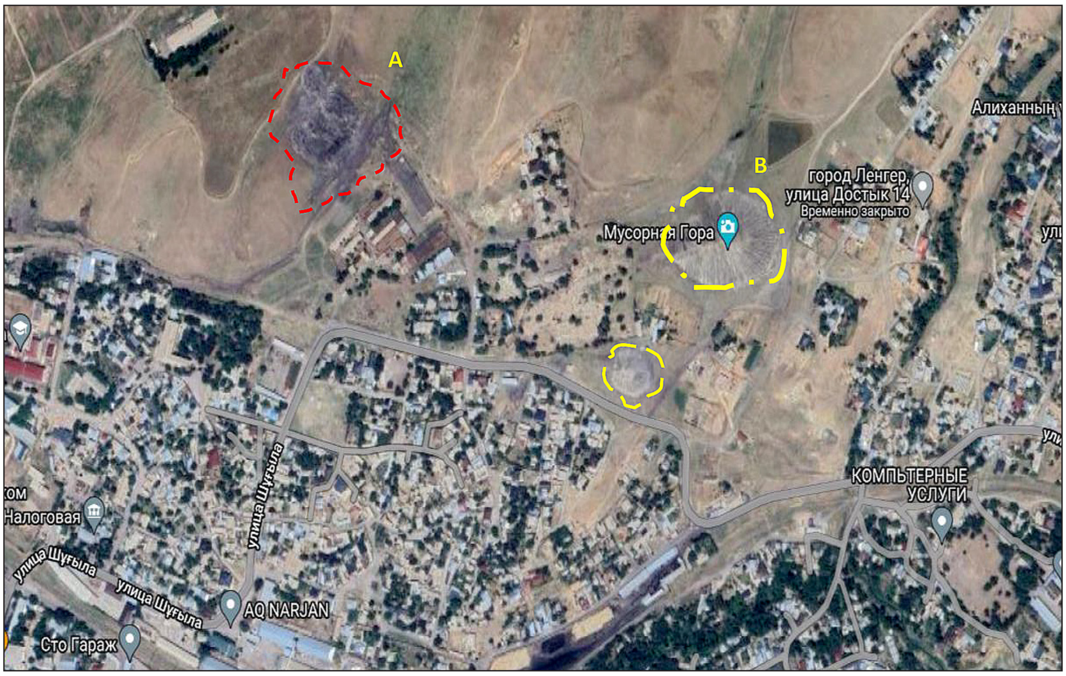

Figure 1. Waste storage sites: A. anthracite-like (42011'23" N, 69052'06"E, Google Earth), B. Slag (42012'11" N, 69052'06"E, GoogleEarth)

carried out in 3 and 5 multiple repetitions. The obtained data were processed using the Excel computer application program.

RESULTS AND DISCUSSION

Technogenic waste, in addition to the negative impact on the environment, has "positive" sides, they can serve as raw materials for the production of biological products and commercial products. Brown coal wastes are widespread low-grade coals lying close to the earth's surface, which makes it easy to extract. Brown coal waste can be recommended for recultivation of industrially polluted territories and degraded soils near highways, as a sorbent of gases released during compost preparation, such as NH3, H2S, mercaptans, various volatile fulvic acids; for adsorbing additives in wastewater treatment [Symanowicz, 2023]. It is known that the inorganic part of carbon-containing waste is represented by various minerals based on silicon, calcium, magnesium, etc. According to the results of X-ray phase analysis, it was found that the brown coal waste of the Lengersky deposit consists of minerals such as gypsum, kaolinite, quartz and illite. It was revealed that the coal content in coal mining waste is 15.2±1.5%.

Diffractometric analysis revealed that the mineralogical composition of brown coal waste is represented by such minerals as Kaolinite – $Al_2Si_2O_5(OH)_4$, Quartz – SiO_2, Gypsum – $CaSO_4×2H_2O$, Cronstedtite – $Fe_3((Si_{0.74}Fe_{0.26})_2O_5)(OH)_4$, Margarite – $CaAl_2(Si_2Al_2)O_{10}(OH)_2$, Muscovite – $H_2KAl_3(SiO_4)_3$, Calcite – $CaCO_3$, Laumontite – $CaAl_2Si_4O_{12}(H_2O)_2$, Lead Aluminium Sulfate Hydroxide – $Pb_{0.5}Al_3(SO_4)_2(OH)_6$. The mineralogical composition of the waste varies depending on the sampling site, so the content of the dominant mineral quartz ranges from 61.5 to 92.9% (Figure 2).

Coals contain a large amount of organic compounds, including up to 90% of humic acids and fulvic acids [Anemana, 2019], which were also found in the composition of brown coal waste used in studies (Figure 3). They contain many different functional chemical groups that contribute to physical modification and improvement of soil quality and stimulation of plant growth.

It was revealed that peaks of organic compounds in samples taken from different depths of carbonaceous waste increases depending on the depth of the sampling site (Figure 4), which suggests the processes of microbiological transformation of the organic part of carbonaceous waste.

Microbiological examinations show the presence of heterotrophic, cellulolytic microflora and micromycetes in carbon-containing waste. At the same time, the titer of microorganisms ranges from 10–10^2 CFU/g in the upper horizon, with an increase in the sampling depth to 20 and 30 cm, the titer of microorganisms increases to 10^3–10^4 CFU/g. In the horizon of 30–40 cm, the titer of microorganisms gradually decreases to 10^2 CFU/g. This can be explained by favorable conditions for the vital activity of microorganisms in the horizons of 10–20 and 20–30 cm. This can be explained by favorable conditions for the vital activity of microorganisms in the horizons of 10–20

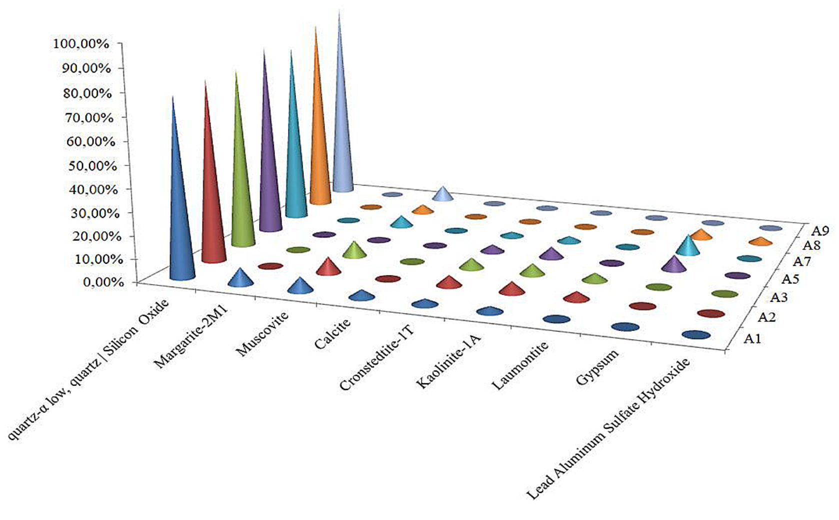

Figure 2. Mineralogical composition of brown coal waste

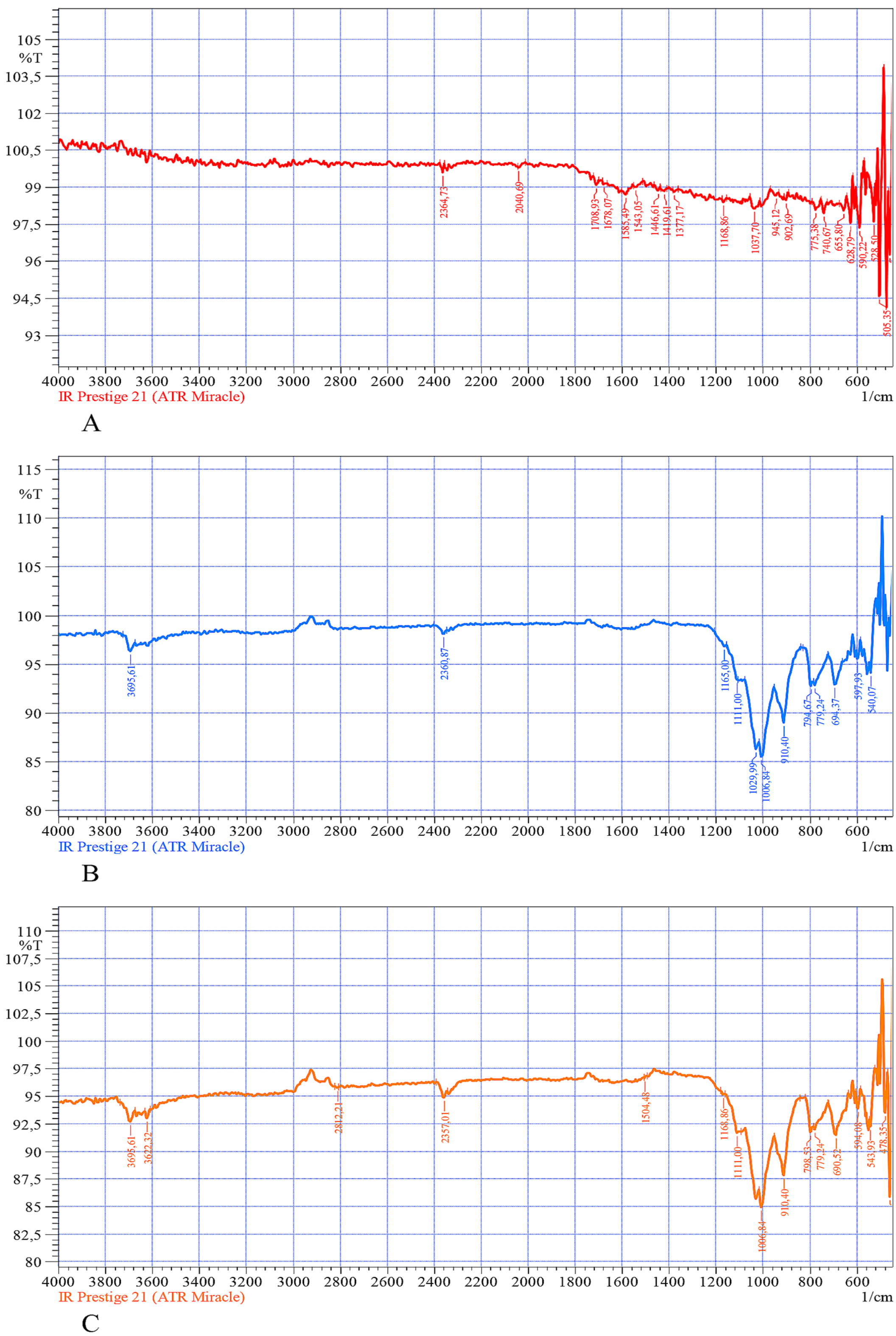

Figure 3. FTIR peaks of carbon-containing waste: A. Sample 1, B. sample 2, C. sample 3

and 20–30 cm: longer periods of humidity, gas-air regime, stable temperature regime. Limiting factors for the growth of microorganisms in the horizon of 0–10 cm are ultraviolet insolation and rapid drying of the surface, in the horizon of 30–40 cm – anaerobic conditions and lack of humidity. Taxonomic analysis revealed that the dominant groups of microorganisms of brown coal waste are represented by *Rhodococcus, Bacillus, Pseudomonas, Penicillium; Trichoderma, Dietzia*.

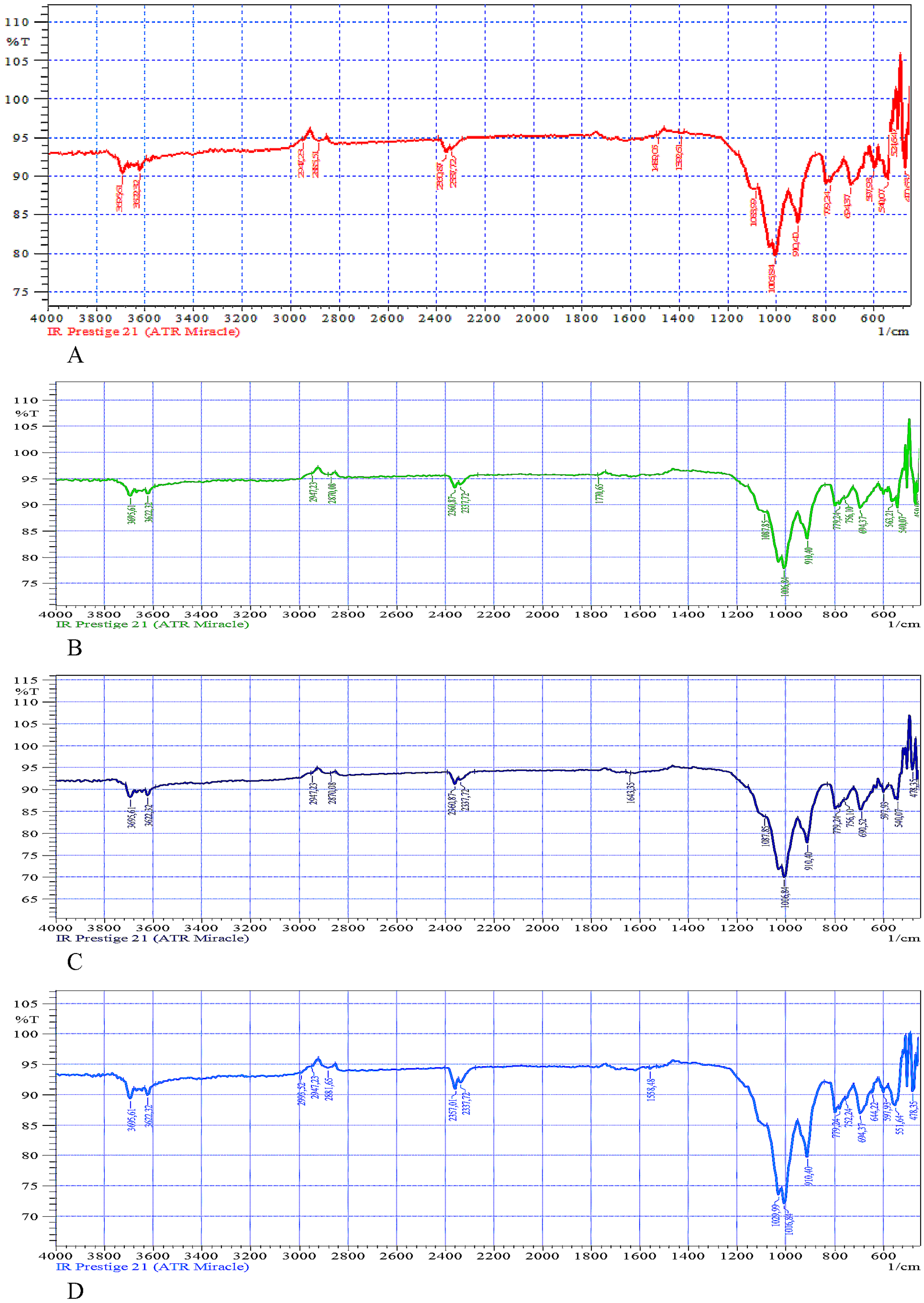

Figure 4. FTIR peaks of A7 carbonaceous waste samples taken from different horizons: A. 0–10 cm, B.10–20 cm, C. 20–30 cm, D. 30–40 cm

A number of studies have shown the possibility of using brown coal waste for recultivation of disturbed soils. For the first time, work using sewage sludge components was carried out in Poland [Amoah-Antwi, 2020]. The use of unconventional meliorants makes it possible to increase the efficiency of reclamation and significantly reduce the cost of creating a fertile layer. Inefficient use of nitrogen fertilizers due to nitrogen losses from the soil is an urgent problem in agricultural systems with large areas. Brown coal, as well as its waste, which is not very popular due to its low calorific value, as an alternative, can be used as a raw material for the production of organo-mineral fertilizers.

Humic and low molecular weight organic acids influence the process of bioremediation of soils contaminated with high molecular weight hydrocarbons and heavy metal ions. Brown coal derivatives can form organometallic ligands with heavy metal ions [Wang et al., 2009]. Bioremediation has a number of advantages, which makes it possible to consider it as an effective method of combating various pollutants, as well as restoring soil microbiocenosis. Compared with traditional methods of remediation, bioremediation is an economical analogue. According to expert estimates, the average cost of bioremediation is no more than 20% of the cost of chemical purification methods. Currently, three main methods of bioremediation in situ are known: natural attenuation, biostimulation and bioaugmentation [Dzionek, 2015]. The method of natural attenuation is based on detoxification of pollutants by native microorganisms. The advantage of this method is its safety for the habitat. However, its implementation may take a long time, because microorganisms capable of decomposing toxic substances make up no more than 10% of the total mass of the soil microbiota. To increase the efficiency of natural attenuation, various approaches of biostimulation and bioaugmentation are being developed [Pimmata, 2013]. For biostimulation of the native microflora, the living conditions are optimized by introducing biogenic elements, maintaining humidity and gas-air regime. In the case of bioaugmentation, microorganisms are introduced in the form of biomass or an immobilized biological product. For the process to be effective, microorganisms must be resistant to environmental conditions and use toxic elements [Simarro, 2013]. For this purpose, native microorganisms are isolated and adapted to increased concentrations of toxicants, the necessary biomass is grown, which is subsequently augmented into the soil [Garbisu, 2017: Issayeva, 2017]. The effectiveness of bioaugmentation is affected by the level of antagonism or symbiosis between introduced and native microorganisms.

The choice of the bioremediation method and the bioremediators used depends on the characteristics of environmental pollution: quantitative and qualitative parameters of the pollutant, pollution history, soil type, weather and climatic conditions, etc. Microorganisms used in bioremediation can purify pollution in three ways: by converting toxicants into less toxic or harmless substances; by extracting pollutants from the environment with subsequent detoxification; suppression of the vital activity of harmful microorganisms. The first method of detoxification of pollutants is associated with the possibility of synthesis of various enzymes by microorganisms (hydrolytic enzymes and oxidoreductases), the second is due to metabolic processes that inactivate various toxins, and the third is antagonistic repression.

In the methods of landfill recovery, the dump rock itself, containing all the mineral salts necessary for the vital activity of microorganisms, is a good substrate for microbiocenosis. The metabolism of microorganisms leads to the accumulation of organic matter in the rock, accelerating the process of soil formation. In addition, microorganisms in the process of their vital activity, releasing various vitamins and growth hormones into the environment, stimulate the development of plants. Inoculation of the active microflora of the surface of coal mine dumps stacked with toxic rocks excludes the application of a fertile soil layer.

Analysis of the literature data shows that various types of meliorant plants are used for recultivation of overburden rocks [Maiti, 2007]. It was found that planting trees on mine soils contributes to improving soil quality by maintaining moisture, nutrients and density.

Similar studies were carried out with phytoconservation of the surface of toxic waste dumps, ash dumps and tailings of mining and metallurgical enterprises [Issayeva, 2015; Issayeva, 2023], where in order to form a fertile layer on the surface of toxic and dusty waste, it was necessary to form a layer of blocks consisting of soil with seeds of toxicotolerant species of ruderal herbaceous flora: *Crambe orientalis* L.*, Centaurea scuarrosa, Plantago lanceolata, Gallium verum* L.*, Agropyron repens, Psoraleae drupaceae, Polygonum aviculare* L.*, Cynodon dactylon, Dodartia orientalis, Cappers herbacea* L.*, Latuca tatarica* L.*, Peganum harmala, Alhagi pseudoadans* L.*, Bromus tectorum* L.. The plant species used for phytoconservation are perennial, which eliminates the need to maintain their vital activity in subsequent years. The results of the research have shown that a stable cover with the formation of turf is formed within 2–3 years. At the same time, an increase in the diversity of soil microflora in the formed soil layer was revealed.

As a result of a floristic survey of the territory around the storage site of brown coal waste, plant species promising for phytoconservation of the waste surface were identified such as *Centaurea scabiosa* L.*, Cynodon dactylon* L.*, Centaurea* *iberica* Trev., *Cichorium intybus* L., *Cousinia cyrdariensis* Kult., *Achillea millefolium* L., *Thlaspi arvense* L., *Arctium tomentosum* Mill., *Onopórdum acánthium* L., *Agropyron cristatum* L., *Polygonum aviculare* L., *Phlum pratense* L., *Erytrigia repens L., Agropyron repens* L., *Capparis spinosa* L., *Dodartia orientalis* L., *Althaea officinales* L., *Alhagi pseudoadans* (Bieb.), *Peganum harmala* L. It was found that, depending on the level of transformation of brown coal waste, local phytocenoses with different plant species structure are formed. The most stable species were *Dodartia orientalis* L., *Polygonum aviculare* L., *Erytrigia repens* (L.) Nevski, *Centaurea pseudosquarrosa* Mikheev ex Gabrieljan et Mikheev, *Capparis spinosa* L., single specimens of which were found even on the surface of unchanged waste. The absence of tree forms is explained by the location of the object of study in the steppe zone.

The algorithm of biorecultivation works in the brown coal waste storage area involves the preparation of soil lumps or blocks consisting of different parts of native soil (in this case, loamy gray soil), expanded vermiculite (0.4–0.7 mm in size), biogenic elements (NPK), microorganisms and seeds of toxicotolerant plants. Due to the absence of a soil layer on the surface of the waste, it is covered with a layer of soil blocks with introduced components. After moistening, the seeds of plants germinate, form a vegetation cover, where, together with inoculated microorganisms, they will participate in the process of biorecultivation of contaminated soils.

CONCLUSIONS

As a result of the study, it was found that the mineralogical part of the waste of brown coal stored in the south of Kazakhstan is represented by: Gypsum $CaSO_4 \times 2H_2O$, Kaolinite $Al_2Si_2O_5(OH)_4$, Cronstedtite $Fe_3((Si_{0.74}Fe_{0.26})_2O_5)(OH)_4$, Margarite $CaAl_2(Si_2Al_2)O_{10}(OH)_2$, Muscovite $H_2KAl_3(SiO_4)_3$, Calcite $CaCO_3$, Laumontite $CaAl_2Si_4O_{12}(H_2O)_2$, Lead Aluminium Sulfate Hydroxide $Pb_{0.5}Al_3(SO_4)_2(OH)_6$, while the quartz content in the samples ranged from 61.5 to 92.9%. Humic acids and fulvic acids make up up to 90% of the organic part of the waste. Saprophytic microflora of waste consists of heterotrophic, cellulolytic microflora and micromycetes belonging to the genera *Rhodococcus, Bacillus, Pseudomonas, Penicillium; Trichoderma, Dietzia*. Ruderal plant species of the local flora resistant to soil contamination by brown coal waste were identified, such as: *Centaurea scabiosa* L., *Centaurea iberica* Trev., *Cichoriumin tybus* Linn., *Cousinia cyrdariensis* Kult., *Achillea millefolium* L., *Thlaspi arvense* L., *Arctium tomentosum* Mill., *Onopórdum acánthium* L., *Agropyron cristatum* L, *Phlum pretense* L., *Erytrigia repens* L., *Nevski, Agropyron repens* L., *Cynodon dactylon* L. Pers., *Polygonum aviculare* L., *Capparis spinosa* L., *Dodartia orientalis* L., *Althaea officinales* L., *Alhagi pseudalhagi* (Bieb.)Desv., *Peganum harmala* L., *Dodartia orientalis* L., *Polygonum aviculare* L., *Erytrigia repens* L. Nevski, *Centaurea pseudosquarrosa* Mikheev ex Gabrieljan et Mikheev showed the ability to grow on soils with a high content of waste. *Capparis spinosa* L. The algorithm of biorecultivation works has been developed, including the use of soil blocks with integrated plant seeds and microorganisms.

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