

Innovative Technologies for the Use of Drilling Waste in Reclamation and Shaping the Physicochemical Properties of Degraded Soils

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ABSTRACT

This paper presents an evaluation of the use of drilling tailings in reclamation and development of physical and chemical properties of degraded soils. Composites with drilling wastes and other wastes, useful for the reclamation of degraded soils and their reconstruction in devastated areas, were used in the tests. Comparative objects were anthropogenic soil fertilized with NPK and manure at a dose of 30 Mg/ha + NPK. The research shows that the tested anthropogenic soil was characterized by very poor sorption properties, and especially the composites with the participation of the tested waste improved the described properties. The composite with sewage sludge from ZA Puławy had a more favorable (by approx. 20%) effect on the sorption capacity of the reclaimed anthropogenic soil than the composite with municipal sewage sludge from Lublin. The effect of the tested composites on reducing the hydrolytic acidity of the reclaimed soil was similar, but much more favorable than that of manure. The results of the conducted research confirmed that drilling wastes have environmentally valuable properties, but due to their unbalance, they should not be used alone, but in composites with properly selected other wastes, such as manure or sewage sludge.

Keywords: drilling waste, sewage sludge, degraded soils, soil reclamation, physicochemical properties

INTRODUCTION

In Poland, approx. 60% of industrial waste generated is mining waste, which is generated as a result of exploration and extraction of minerals [1]. According with the Regulation of the Minister of the Environment on the waste catalog [2] mining waste includes: waste from mineral extraction, waste from physical and chemical processing of metal ores, waste from the physical and chemical processing of minerals other than metal ores, used drilling fluids and other drilling waste. Drilling wastes constitute a geological cross-section of the drilled space of the earth's crust, which makes them natural rocks, partially modified by

additives that improve the drilling process [3]. According to the principle that „what has been taken from the environment should return to it”, it is necessary to develop rational methods of environmental management of drilling waste. In the light of the above, it is urgent to develop rational ways of drilling waste management, which can be achieved by conducting innovative scientific research. Such research was undertaken as part of this work. Their main objective was to assess the properties of drilling wastes for the need to produce a composite with other wastes, which would have the properties of an agent useful for: making biological cover at landfills; reclamation of soils in degraded areas and their reconstruction on soilless

formations, including drilling rig areas, as well as shaping the properties of poor-quality soils. The results of research on waste management so far indicate the need to optimize their properties in order to achieve the maximum ecological effect [4]. Waste is generally characterized by diverse and unbalanced properties, which in natural management may reduce their fertilization efficiency. In such a case, it is justified to combine the wastes, thanks to which the properties of the created composite are optimized [5-7]. Such activities regarding drilling waste were undertaken in the research carried out as part of the project „Optimization of drilling waste management methods”, the aim of which was to use drilling spoil to produce a composite that meets the properties of an agent improving the properties of poor quality soil and useful for soil reconstruction in devastated areas. A comprehensive analysis of the properties of drilling wastes showed that they are characterized by good, but varied properties, which in natural management may reduce their fertilization efficiency. This was the basis for developing the principle of their association with industrial sludge from the biological sewage treatment plant of Nitrogen Works Puławy S.A., municipal sewage sludge from Lublin, Carboniferous mining waste from the „Bogdanka” Hard Coal Mine and waste mineral wool from horticultural crops under covers from the Garden Plants in Niemce near Lublin. The impact of the developed composites on the formation of comprehensive properties of the degraded anthropogenic soil from the foreground of the „Kruszywa Niemce” Sand Mine was carried out in strict pot and field experiments, the results of which allowed the development of innovative technologies for their use for the reclamation of degraded soils and soil reconstruction on soilless formations. This technology has been implemented in the reclamation and natural development of the excavation after sand extraction in Undertaking „Kruszywa Niemce” S.A. The aim of this work was to evaluate changes in physicochemical properties in the created external reclamation layer.

MATERIALS AND METHODS

Technology of excavation reclamation

The excavation after sand extraction, with a depth of approx. 30 m, was filled on an area of 0.5 ha with gangue – Carboniferous mining

waste from the „Bogdanka” Hard Coal Mine. A layer of mining waste with a thickness of 1 meter was spread on the bottom of the excavation, and then, in order to create a natural sealing barrier, it was compacted with a caterpillar tractor. The excavation was filled with Carboniferous mining waste to a height of 1.5 m below ground level. Two plots with an area of 0.25 ha each were marked out on such an area. On the designated areas, the 1.5-meter top reclamation layer was made in a way that will guarantee, in accordance with the Spatial Development Plan (PZP), forest management of the land after reclamation. To make the profile of the top reclamation layer with a thickness of 1.5, the following were used: Carboniferous mining waste from the Bogdanka Hard Coal Mine, drilling waste from Luchów Górny near Tarnogród, overburden of anthropogenic soil deposited in the foreground of the Sand Mine and sewage sludge:

- on one surface, sludge from the Biological Sewage Treatment Plant at ZA Puławy – a by-product,
- on the second surface, sludge from the mechanical and biological municipal wastewater treatment plant in Lublin.

Such action was aimed at confirming the knowledge obtained in exact experiments regarding the possibility of using composites in reclamation works drilling waste, local resources of organic waste – sewage sludge, without the need to transport them over long distances.

The arrangement of layers in the created reclamation layer took into account the target forest management of the reclaimed area.

- For surfaces using composites with sewage sludge from Puławy „P” it was as follows:
 - 1. Layer 0–25 cm: anthropogenic soil 2000 Mg·ha⁻¹ + „P” sediment 200 Mg DM·ha⁻¹ + NPK. 2. Layer 25–50 cm: anthropogenic soil 1000 Mg·ha⁻¹ + sewage sludge „P” 200 Mg DM·ha⁻¹ + mining waste 1000 Mg·ha⁻¹ + drilling waste 200 Mg·ha⁻¹.
 - Layer 50–75 cm: anthropogenic soil 500 Mg·ha⁻¹ + sewage sludge „P” 100 Mg DM·ha⁻¹ + mining waste 1500 Mg·ha⁻¹ + drilling waste 300 Mg·ha⁻¹.
 - Layer 75–100 cm: anthropogenic soil 300 Mg·ha⁻¹ + mining waste 2000 Mg·ha⁻¹ + drilling waste 300 Mg·ha⁻¹.
 - Layer 100–150 cm: mining waste 2500 Mg·ha⁻¹ + drilling waste 500 Mg·ha⁻¹.

- For surfaces using composites with municipal sewage sludge from Lublin, the „L” was as follows:
 - Layer 0–25 cm: anthropogenic soil 2000 Mg·ha⁻¹ + „L” sediment 200 Mg DM·ha⁻¹ + NPK. 2. Layer 25 - 50 cm: anthropogenic soil 1000 Mg·ha⁻¹ + „L” sediment 200 Mg DM·ha⁻¹ + mining waste 1000 Mg·ha⁻¹ + drilling waste 200 Mg·ha⁻¹.
 - Layer 50–75 cm: anthropogenic soil 500 Mg·ha⁻¹ + „L” sediment 100 Mg DM·ha⁻¹ + mining waste 1500 Mg·ha⁻¹ + drilling waste 300 Mg·ha⁻¹.
 - Layer 75–100 cm: anthropogenic soil 300 Mg·ha⁻¹ + mining waste 2000 Mg·ha⁻¹ + drilling waste 300 Mg·ha⁻¹.
 - Layer 100–150 cm: mining waste 2500 Mg·ha⁻¹ + drilling waste 500 Mg·ha⁻¹.

Surfaces shaped in this way were fertilized with NPK mineral fertilization in the amount of: N – 80 kg·ha⁻¹ (ammonium nitrate); P₂O₅ – 80 kg ha⁻¹ (polifoska 8); K₂O – 120 kg·ha⁻¹ (potassium salt) and seeds of a mixture of grasses with the following species composition: Timothy (*Phleum pratense* L.) – (Obra C/1) – 20%; meadow fescue (*Festuca pratensis* Huds.) – (Ardenna C/1) – 20%; cocksfoot (*Dactylis glomerata* L.) – (Ambasador C/1) – 10%; red fescue (*Festuca rubra* L.) – (Kos C/1) – 20%; red fescue (*Festuca rubra* L.) – (Nimba C/1) – 10%; meadow grass (*Poa pratensis* L.) – (Balin C/1) – 15%; red clover (*Trifolium pratense* L.) – (Krynica C/1) – 5%, in the amount of 80 kg·ha⁻¹. In the reclaimed area, after a period of 3–5 years of use as grassland, it is planned to plant trees in accordance with the PZP. Such a solution increases the usefulness of reclamation with the use of woody plants, which was found during the reclamation and development of the area degraded by sulfur borehole mining in Jeziórko near Tarmobrzeg.

Comparative objects were anthropogenic soil fertilized:

- NPK in doses as above.
- Manure at a dose of 30 Mg·ha⁻¹ + NPK in doses as above.

Technical reclamation was carried out in May 2015. Biological recultivation – grass mixture sowing, due to the prevailing drought, was carried out in August 2015. The photos below show the growth of plants in the 2nd month after sowing.

In 2016/17, the properties of the soil formed on reclaimed areas were analyzed. From each

area, 6 soil samples were taken from 5 points spaced proportionally from the 0–25 cm layer.

Laboratory analysis

The following soil and waste samples were determined:

- Potentiometrically in H₂O and 1 mol·dm⁻³ KCl [8],
- Hydrolytic acidity (Hh) by the Kappen method in 1 mol·dm⁻³ CH₃COONa, [9-10],
- Basic cations (S) using the Pallmann method in the extract of 0.5 mol·dm⁻³ of ammonium chloride (pH 8.2) [11],
- The sorption capacity (T) and the degree of saturation of the sorption complex with basic cations (V) were calculated.
- The content of heavy metals – general forms were determined in the Central Agroecological Laboratory (accredited analyses) of the University of Life Sciences in Lublin – Cu, Zn, Pb, Cd, Cr and Ni – by Atomic Adsorption Spectrometry in an acetylene-air flame [12] and Hg according to the CLA/ASA/5/2013 procedure.

RESULTS AND DISCUSSION

Characteristics of the materials used in the experiment

Drilling waste is a sandy formation with a content of 41% of the sand fraction (2.0–0.05 mm), 34% of the dust fraction (0.05–0.002 mm) and 25% of the clay fraction (<0.002 mm), which qualifies it as ordinary clay (Table 1). This formation was characterized by an alkaline reaction: pH in 1 mole of KCl was 8.6; while in H₂O it was 9.0. Drilling wastes were characterized by a high content of alkaline cations (23.25 cmol(+)·kg⁻¹) and sorption capacity(23.70 cmol(+)·kg⁻¹), corresponding to very good quality soils and high (96.7%) saturation with basic cations. Hydrolytic acidity was low and amounted to 0.45 cmol(+)·kg⁻¹ [13]. The total content of heavy metals (Pb, Zn, Cu, Cr, Ni, Cd, Hg) in drilling wastes was low, characteristic of natural soils [14-16]. The barium content was 14900 mg·kg⁻¹, which indicates that these values are high compared to soils [17]. The mineralogical composition of drilling waste (clay minerals – 45.5%; quartz – 27.4%, dolomite – 10.7% [18]), and the argument for the reclamation use of drill cuttings are their physical and chemical properties, such as such as the

high content of dust and clay particles, the presence of biogenic elements, e.g. K, Ca, Mg. Moreover, drilling wastes are characterized by alkaline pH and high buffering capacity, which prevents pH changes in the substrate due to the impact of acidifying agents and the release of heavy metals into the environment [1]. Sewage sludge from the Biological Sewage Treatment Plant (BOŚ) of Nitrogen Works Puławy S.A. has the status of a by-product, with a slightly acid reaction (pH H₂O 6.5; KCl 6.0). The sewage sludge used in the experiment was characterized by a high content of alkaline cations (25.17 cmol(+) \cdot kg⁻¹) and sorption capacity (26.75 cmol(+) \cdot kg⁻¹), corresponding to very good quality soils and high (94.09%) by saturation with basic cations (Table 1). The saturation with H⁺ ions was low (6%). The content of heavy metals was low and compared to municipal sewage sludge, the values were 2–3 times lower.

Sewage sludge from the municipal wastewater treatment plant in Lublin was characterized by a slightly acid reaction, with a pH value of 6.4 in H₂O and 6.1 in 1 mole of KCl (Table 1). The hydrolytic acidity was 4.73 cmol(+) \cdot kg⁻¹, and the content of basic exchangeable cations was 29.14 cmol(+) \cdot kg⁻¹, which means that its sorption capacity was high (33.87 cmol(+) \cdot kg⁻¹) and high (86.0%) saturation with basic cations. The sanitary properties of sewage sludge met the requirements of the Regulation of the Minister of the Environment on municipal sewage sludge [15]. As shown by numerous research results [19-25], sewage sludge used for fertilization and soil reclamation is a source of nutrients (N, P, secondary nutrients and

microelements) for plants, improves the physical and physicochemical properties of the soil and increases the content of organic matter.

Mining waste has the following fractions: coarse-grained (20–200 mm) 40-50%, fine-grained (0.5–20 mm) 30–40% and mud-clay (<0.5 mm) approx. 20%. The mineralogical composition consists of (50–60%) clay minerals (kaolinite, illite, chlorite), quartz, muscovite (10–35%), organic substance (7–10%), siderite (2–5%) and trace amounts of feldspar and pyrite. These wastes were characterized by a neutral pH of 6.71 in 1 mole of KCl (Table 1). The hydrolytic acidity was 0.61 cmol(+) \cdot kg⁻¹, while the content of exchangeable basic cations was 10.59 cmol(+) \cdot kg⁻¹, which determines the high (94.5%) degree of saturation with basic cations. The content of heavy metals was comparable for soils with their natural content and did not exceed the reference levels for agricultural soils [14-15].

Anthropogenic soil is sandy clay with a granulometric composition: 50% sand fraction (2.0-0.05 mm), 47% dust fraction (0.05–0.002 mm) and 3% clay fraction (<0.002 mm). It was characterized by a strongly acid reaction, the pH in 1 mole of KCl was 4.0 (Table 1). The hydrolytic acidity was 2.80 cmol(+) \cdot kg⁻¹, while the sum of basic cations was only 3.40 cmol(+) \cdot kg⁻¹. The degree of saturation of the sorption complex with alkalis was 60.7%. The content of heavy metals: Pb, Zn, Cu, Cd, Cr, Ni, Hg and Ba was low and characteristic of soils with their natural content [14-16].

The manure was alkaline, the pH in 1 mole of KCl was 8.3 (Table 1). The content of basic

Table 1. Properties of materials used in reclamation

Properties	Unit	Sewage sludge Puławy	Sewage sludge Lublin	Drilling waste	Mining waste	Anthropogenic soil	Manure
pH	H ₂ O	6.50	6.40	9.0	7.61	4.50	8.90
pH	KCl	6.00	6.10	8.6	6.71	4.00	8.30
Hh	cmol(+) \cdot kg ⁻¹	1.58	4.73	0.45	0.61	2.80	0.75
S	cmol(+) \cdot kg ⁻¹	25.17	29.14	29.25	10.59	3.40	43.80
T	cmol(+) \cdot kg ⁻¹	26.75	33.87	29.70	11.19	5.60	44.55
V	%	94.09	86.00	96.70	94.50	60.70	98.32
Cu	mg \cdot kg ⁻¹	13.30	301.00	26.50	54.20	20.00	12.40
Zn	mg \cdot kg ⁻¹	55.40	874.00	66.10	57.20	24.50	151.0
Pb	mg \cdot kg ⁻¹	21.80	39.50	34.50	62.50	27.20	18.30
Cr	mg \cdot kg ⁻¹	12.50	89.20	18.80	17.20	19.50	5.40
Ni	mg \cdot kg ⁻¹	7.40	37.50	36.30	44.50	5.60	5.60
Cd	mg \cdot kg ⁻¹	0.51	3.52	0.72	1.01	0.34	1.00
Hg	mg \cdot kg ⁻¹	0.015	0.982	0.036	0.111	0.015	0.026
Ba	mg \cdot kg ⁻¹	51.30	110.00	14900.0	305.00	41.30	96.00

cations was 43.80, and the sorption capacity was 44.55 $\text{cmol}(+)\cdot\text{kg}^{-1}$, which indicates high (98.3%) saturation with basic cations. The total content of heavy metals was low.

Changes in physicochemical properties in reclaimed anthropogenic soil

Reaction. The addition of the tested composites to the anthropogenic soil resulted in the liquidation of its acidification from the strongly acidic level to the neutral level (Table 2). The soil fertilized only with NPK ($\text{pH}_{\text{H}_2\text{O}} = 5.0$; $\text{pH}_{\text{KCl}} = 4.2$) and additionally with NPK with farmyard manure ($\text{pH}_{\text{H}_2\text{O}} = 5.1$; $\text{pH}_{\text{KCl}} = 4.9$) was highly acidic, while the addition of a composite with sewage sludge from Puławy and with the use of municipal sewage sludge from Lublin, it reduced acidification by approx. 2.6–3.1 pH units to a neutral level. The impact of the composite with the sewage sludge from Puławy was more favorable by 0.3–0.6 pH than the composite with the municipal sewage sludge from Lublin. The pH of the soil is considered one of the most important indicators of soil fertility. The soil’s physical, chemical and biological properties depend to a large extent on the acidity of the soil. Acidity affects the durability of the lumpy structure and the air-water relations associated with it [26]. Other authors in their research, in order to reduce soil acidity, fertilized it with manure and sewage sludge [14, 27-28]. Gondek [29] and Skowrońska and Filipek [30], analyzing the fertilizing use of sewage sludge, showed its acidifying, but insignificant, effect, especially in the second year after application. It should be emphasized that the favorable shaping of the soil reaction found in the

conditions of the presented research should be associated mainly with the impact of drilling waste and mining waste, as well as sewage sludge.

Sorption properties. The very low sorption capacity of the anthropogenic soil increased under the influence of the tested waste composites 6.3–8.0 times compared to the soil fertilized with NPK and 6.4–7.9 times compared to the soil fertilized with manure (Table 3, Fig. 1). The obtained values of sorption capacity corresponded to very good quality soil. The composite with sewage sludge from Puławy had a more favorable (by approx. 20%) effect on the sorption capacity of the reclaimed anthropogenic soil compared to the composite with municipal sewage sludge from Lublin. The effect of the tested composites on reducing the hydrolytic acidity of the reclaimed anthropogenic soil was similar and much more favorable than that of manure (Table 3).

It is also valuable that more than 90% of the soil sorption capacity was due to Ca and Mg, elements having a positive impact on the initiation and course of the soil-forming process. The content of exchangeable cations in the sorption complex ranks them in the following order: $\text{Ca} > \text{Mg} > \text{K} > \text{Na}$ (Fig. 2). Also, the saturation of anthropogenic soil with alkaline cations corresponds to very good quality soil (Table 3).

Research by Baran et al. [20] on long-term soil contamination with sulfur showed that it contributes to the deterioration of their sorption properties due to the loss of exchangeable calcium and magnesium. This is due to the excessive concentration of H^+ ions. The sulfuric acid formed in the conditions of sulphation weakens the protective effect of the sorption complex against the leaching of mineral components, which is the basis for the chemical

Table 2. pH of the reclaimed anthropogenic soil

Properties	Unit	Soil + NPK reference object	Soil + manure 30 Mg/ha + NPK reference object	Composite with sewage sludge from Puławy	Composite with municipal sewage sludge from Lublin
pH	H ₂ O	5.0	5.1	7.4-7.6	6.5-7.2
pH	KCl	4.2	4.9	7.0-7.3	6.4-7.0

Table 3. Average values of sorption properties of reclaimed soil

No.	Reclamation variant	Hh	S	T	V
		$\text{cmol}(+)\cdot\text{kg}^{-1}$			%
1	Soil + NPK	2.67	4.08	6.75	60.4
2	Soil + manure + NPK	1.95	4.90	6.85	71.5
3	Composite with sewage sludge from Puławy	1.54	52.51	54.05	97.2
4	Composite with municipal sewage sludge from Lublin	1.55	41.93	43.48	96.8

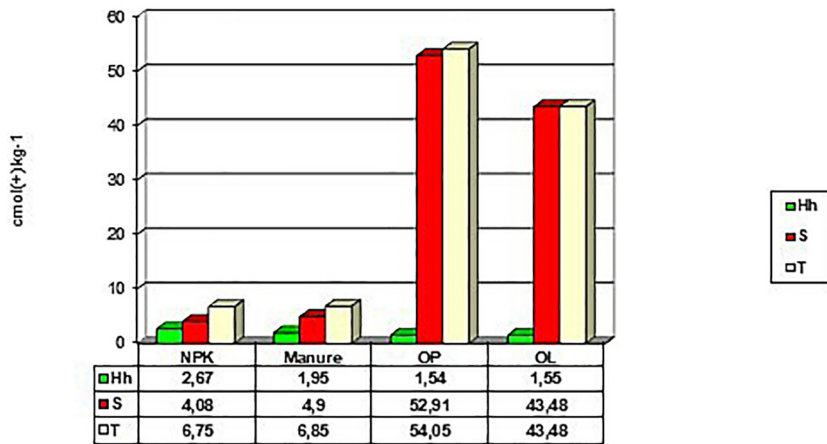


Fig. 1. Influence of reclamation methods on sorption properties in reclaimed anthropogenic soil. Average values. Symbols: NPK - soil + NPK; Manure - soil + manure 30 Mg/ha + NPK; OP - soil + composite with sewage sludge from Puławy; OL - soil + composite with municipal sludge in Lublin

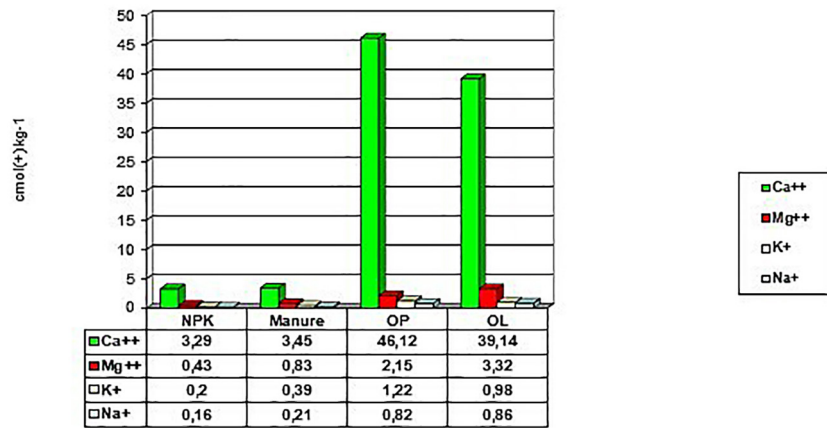


Fig. 2. Influence of reclamation methods on the content of exchangeable cations in reclaimed anthropogenic soil. Average values. Designations as in Figure 1

degradation of soils contaminated with sulphur. Other authors [31-32] report that the reclamation of post-mining areas in sulfur mining (borehole and open pit) is a complex and difficult issue, because this type of mining activity causes a number of transformations of the natural environment with a very wide scope. The results of Baran et al. [20] indicate a significant, 3.5–9-fold improvement in the sorption properties of the devastated soilless formation, which depended on the method of reclamation, especially the addition of sewage sludge [20]. Also, a highly positive effect on increasing the sorption capacity of soils degraded by strong acidification was found after applying sewage sludge and sawdust compost [6, 34-35]. The value of the sorption capacity of the reclaimed soil under the influence of the tested waste should be assessed as high, corresponding to good quality soils [36]. In the study of Baran et al. [20], in addition to a significant increase in the sorption capacity,

the saturation of the sorption complex with cations was improved. The dominant cation was calcium, the share of which corresponds to good quality soils. The share of the remaining cations forms a series: $Mg_2^+ > Na^+ > K^+$. The tested methods of reclamation with the use of mineral wool and sewage sludge had a beneficial effect on the content of calcium, magnesium, potassium and hydrogen cations in comparison to the use of only mineral fertilization with NPK [20]. From the studies conducted by other authors [37-41] shows that the increase in the dose of the sewage sludge used resulted in an increase in hydrolytic acidity. Only doses of 10 Mg per hectare of sludge did not cause a statistically significant increase in acidity in relation to control objects. On the other hand, manure fertilization caused a decrease in soil acidification. These studies also prove that sewage sludge causes a greater increase in the sum of alkaline cations in fertilized soils than cattle manure [37-41]. An expression of

the optimization of the sorption properties of the reclaimed soil was the improvement of its chemical and biological properties.

CONCLUSIONS

Drilling and mining tailings and sewage sludge used in reclamation were characterized by alkaline reaction, high content of alkaline cations and sorption capacity as well as high saturation with basic cations and low hydrolytic acidity. The total content of heavy metals in the waste was low. Anthropogenic soil is a strongly acidic sandy loam with low sorption capacity and content of basic cations, and high hydrolytic acidity. The content of heavy metals was low and characteristic of soils with their natural content. The addition of waste composites to highly acidic anthropogenic soil improved its reaction to a neutral level. The very low sorption capacity of the anthropogenic soil increased 6.3–8.0 times under the influence of the tested composites, compared to the soil fertilized with NPK and 6.4–7.9 times fertilized with manure. The composite with sewage sludge from Puławy had a more favorable (by approx. 20%) effect on the sorption capacity of the reclaimed anthropogenic soil than the composite with municipal sewage sludge from Lublin. The effect of the tested composites on reducing the hydrolytic acidity of the reclaimed soil was similar, but much more favorable than that of manure. The results of the research confirmed that drilling wastes have environmentally valuable properties, but due to their unbalance, they should not be used alone, but in composites with properly selected other wastes. This principle results from the present research, in which composites with varying amounts of drilling waste, Carboniferous mining waste, sewage sludge from the biological sewage treatment plant of Nitrogen Works S.A. were used for the reclamation of degraded anthropogenic soil in Puławy (having the status of a by-product) and sewage sludge from the municipal sewage treatment plant in Lublin.

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The changes in physical and chemical properties in the reclamation layer were tested on a reclaimed excavation after sand extraction in Undertaking „Kruszywa-Niemce” S.A., according to the technology developed as part of the project „Optimization of drilling waste management methods”.

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