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Application of Drilling Waste in the Reclamation of Acidic Soils

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1. Introduction

The dynamic development of shale gas extraction observed in the recent years gives hope for inhibiting unfavourable changes occurring in the natural environment by lowering the CO_2 emissions from the energy sector. On the other hand, it also raises serious concerns related to the pollution of environment during the exploration, release, and exploitation of shale gas from shale deposits (Siemek et al. 2013, Cao et al. 2015). Both ground and surface waters, as well as soil constitute the most endangered elements of the natural environment. Additionally, huge amounts of drilling waste are produced in the course of shale gas exploration, including drill cuttings and spent drilling fluid (Babko et al. 2016). Drill cuttings consist in the excavated material produced during drilling, which is extracted along with the drilling fluid. According to the available data, 0.6 m^3 of waste is produced from 1 mb of wellbore drilled, which translates into 2500-6000 Mg of waste from one drilling. In the case of Poland, it is estimated that approximately 87500-210000 Mg of drilling waste is produced annually (Macuda 2010, Kronenberg 2014). The composition and properties of drill cuttings may vary greatly, depending on type of drilling fluid used, as well as on the geological and technological conditions of drilling. Drill cuttings are mainly made up of rocks and shales of autochthonous material in which drilling is performed and may be combined with varied amounts of drilling fluid. Variable and diversified composition of drill cuttings hinders the process of devising an efficient technology of managing this type waste and complicates the assessment of their toxicity, as well as the determination of environmental impact (Abbe et al. 2009, Reichetseder 2016, Pawłowski et al. 2016, Burri 2016). Drill cuttings management in Poland is most frequently limited to depositing on waste disposal sites. Simultaneously, research work on the possibility of managing and utilizing this type of waste is performed. Studies on the usefulness of drill cuttings as a soil additive for swamp plants which were conducted in the USA showed that there is no significant difference between cultivating plants in soil modified with drill cuttings and its unmodified variant (Ball et al. 2012, Bielińska et. al. 2016, Bielińska et. al. 2015, Pawłowski et. al. 2015). The Taranaki technical report showed the influence of drill cuttings on the productivity of sandy soils. Changes in the level of nutrients caused by the drill cuttings addition enabled to convert non-productive soils into pastures (Technical report 2011, Żukowska et. al. 2016, Lindzen & Sloan 2016).

Considering the properties of drill cuttings, the reclamation of acidic soils may prove to be an attractive method of managing this type of waste, which constitutes a problem both in Poland and other countries. Soil acidification is caused by natural and anthropogenic factors. The latter mainly consist of acid rains, intensive plant cultivation and application of high doses of physiologically acidic mineral fertilizers (Baran & Turski 1996, Karczewska 2008). Acidification of soils negatively impacts, among others, the quantitative proportions of soil microflora. In acidic soils fungi and actinobacteria are the dominant group, whereas bacteria decline. Therefore, various biochemical processes including, for instance, nitrification, organic matter decomposition and humus forming, slow down or – in extreme cases – stop altogether (Dobrzańska et al. 2008, Karczewska 2008).

In order to counteract the negative effects of soil acidification, liming is a commonly employed solution. Liming can be performed by means of various calcium fertilizers of natural or industrial origin (Baran et al. 2011). The work presents the results of preliminary studies on the improvement of acidic soil properties by means of drill cuttings.

2. Materials and methods

Acidic soil, drilling cuttings and ion exchange substrate Biona-312 were used as basic materials in the study.

Soil was taken from the field situated in Felin, near Lublin (Eastern Poland). It consisted of the following fractions: sand (2.0-0.05mm) – 22%; silt (0.05-0.002 mm) – 73%; clay (<0.002 mm) – 5%. The pH value of the soil in KCl solution was about 4.8. The contents of macronutrients available for plants in the soil were determined according to Polish standards (Lityński & Jurkowska 1982, Ostrowska et al. 1991). It was characterized by common (Mg), low (K, S), very low (P) or insufficient (N, Ca) contents of nutrients in terms of plants requirements – Table 1.

Table 1. Contents of nutrients in soil
Tabela 1. Zawartość makroskładników w glebie

$N-NH_4$	$N-NO_3$	P_2O_5	K_2O	Mg	$S-SO_4$	Ca
[mg per kg]		[mg per 100g]				[mg per dm^3]
2.98	22.26	3.40	10.21	5.97	0.67	318.8

Drilling cuttings were obtained from the facility of mining waste utilization located in Luchów near Biłgoraj (Eastern Poland). Its granulometric composition was as follows: sand (2.0-0.05mm) – 75%; silt (0.05-0.002 mm) – 18%; clay (<0.002 mm) – 7%. The pH value of waste in KCl solution was about 9. The content of carbonates in waste equalled to 36.2% and the organic matter content was 8.5%. The metal contents in a solution obtained after waste mineralization with aqua regia (0.1 g of waste + 7 ml of the acid) was determined by ICP-OES JY 238 Ultrace (Table 2). Waste only contained high concentration of barium which was above the acceptable levels for agricultural and industrial lands, according to Polish regulations (Journal of Laws of the Republic of Poland No 165/2002 item 1359, Journal of Laws of the Republic of Poland No 175/2011, item 1048). However, barium was present in the cuttings in the form of sulphates insoluble in water (barite). Therefore, it would determine very low threat to the environment. The ion exchange substrate Biona-312 was prepared at the Institute of Physical Organic Chemistry of BNAS in Minsk. It was a mixture of 56% (mass) of ion exchange substrate Biona-112 and 44% of clinoptilolite. The Biona-312 substrate contained the following amounts of nutrients ($g \cdot kg^{-1}$): N – 11.21, P – 3.41, K – 17.60, Mg – 4.38, Ca – 22.24, S – 6.09, Mn – 0,22, Cu – 0,06, Zn – 0,06, Co – 0.02, Mo – 0.04, B – 0.11, Fe – 2.23, Na – 1.38, Cl – 3.9. The studies were performed using orchard grass (*Dactylis glomerata* L. cv.

Amera) as the test species. For the purpose of the pot experiment, six series of media were prepared including: soil with 2% (v/v) addition of Biona-312 substrate (the control series), four soil series with 2% addition of Biona-312 substrate and increasing drilling waste doses and soil alone (Table 3).

Table 2. Content of metals in drilling waste. Permissible content B – permissible content for agricultural lands (group B), permissible content C – permissible content for industrial lands (group C) according to Journal of Laws of the Republic of Poland 165/2002 item 1359
Tabela 2. Zawartość metali w odpadach wiertniczych. Dopuszczalna zawartość B – dopuszczalna zawartość dla użytków rolnych (grupa B), dopuszczalna zawartość C – dopuszczalna zawartość dla terenów przemysłowych (grupa C) zgodnie z Dziennikiem Ustaw nr 165/2002 poz. 1359

Metal	Content [ppm]	Group B	Group C
Mg	19407	–	–
Ca	76219	–	–
K	284443	–	–
Na	4811	–	–
Cu	99	150	600
Mn	469	–	–
Fe	14370	–	–
Co	0,08	20	200
Cr	63	50	500
Cd	0	4	1
Pb	21	100	600
Ni	40	100	300
Ba	1820	200	1000

Biona-312 substrate was used to supplement soil with macronutrients since it was characterized by poor richness in these elements. The drilling waste doses were established as: 2%, 5%, 10% and 15% (v/v) taking into account pH measurements of the composed media (Table 3).

The experiment started on 15 May 2015. In each pot (of 360 cm^3 volume) 51 orchard grass seeds were sown. The experiment was carried out in a phytotron with a 13/11 light/dark regime. The daytime (7 am-8 pm) air temperature was 25°C while the night-time (8 pm-7 am) air temperature was 16°C. During the experiment the plants were watered with distilled water. The amount of water depended on the current needs of the plants. The experiment was terminated after 42 days from the time of seed sowing. The plant stems were cut down and roots were separated. The wet and dry (105°C) biomass of stems and dry (105°C) root biomass were weighed. The total dry biomass of plants was calculated as a sum of dry stem and root biomass. The results obtained were used for the calculation of mean values characterizing the experimental series (arithmetical mean values). The statistical significance of differences between mean values was assessed using Student's t-test at the confidence coefficient p = 0.95 (Czermiński et al. 1992, Zgirski & Gondko 1998).

Table 3. Composition and characteristics of media series in the pot experiment
Tabela 3. Skład i charakterystyka podłoży w doświadczeniu wazonowym

Media series	Soil	Biona-312	Drilling waste	pH	Pot number
	[ml per pot]				
S	300	–	–	4.77	5
S+2% B	294	6	–	5.70	5
S+2%B+2%DW	288	6	6	6.48	5
S+2%B+5%DW	279	6	15	7.07	5
S+2%B+10%DW	264	6	30	7.21	5
S+2%B+15%DW	249	6	45	7.47	5

S – soil; B – Biona-312; DW – drilling waste

3. Results and discussion

The study results are presented in Figures 1-4. It can be seen that the 2% addition of Biona-312 substrate to soil increased wet stem biomass, dry root biomass and total dry biomass of orchard grass by 7-24%. However, the differences in all vegetative parameters (including dry stem biomass) between soil alone and soil supplemented with Biona addition (the control series) were not statistically significant. Thus, it could be said that Biona addition to acidic soil did not affect the plant growth to a significant degree, which is inconsistent with the study results obtained by Wasąg et al. (2000), Chomczyńska and Pawłowski (2003), Zdeb et al. (2008). The above-mentioned authors found that the addition of Biona-type substrates to sandy soil or sand significantly elevated dry stem and root biomass of plants by 100-1300% and 30-600%, respectively. The lack of an effect of Biona addition on grass growth in the present study probably is connected with pH of medium. The value of pH for the soil-2% Biona mixture was 5.70 which is below the range of 6-7 (6.3-6.7 for medium-heavy soils) reported as the optimal one for cultivation of most plants (Lityński & Jurkowska 1982).

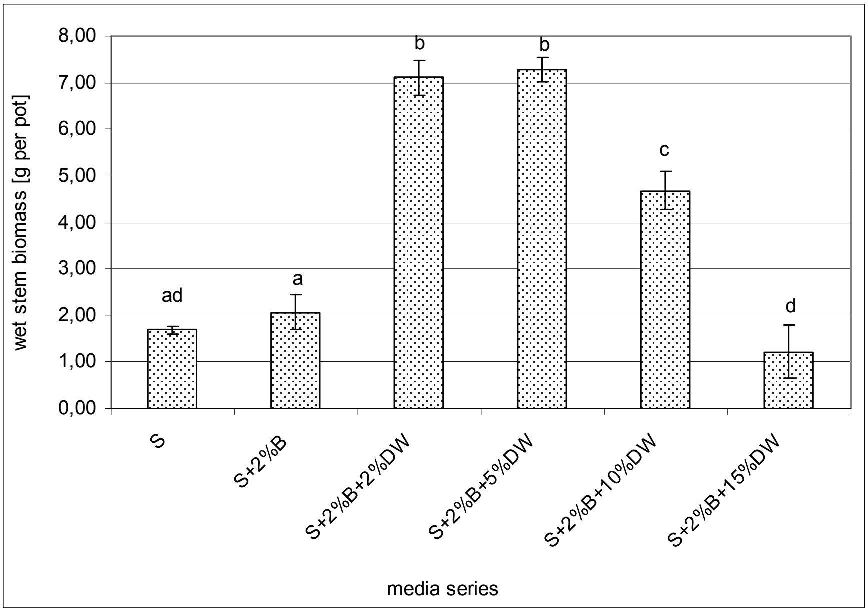

Fig. 1. Mean wet stem biomass of orchard grass in media series. S – soil; B – Biona-312; DW – drilling waste; different letters above bars indicate significant differences between mean values; I – standard deviation
Rys. 1. Średnia świeża biomasa pędów kupkówki pospolitej w seriach doświadczenia wazonowego. S – gleba; B – Biona-312; DW – odpad wiertniczy; różne litery nad słupkami wskazują istotne różnice między wartościami średnimi; I – odchylenie standardowe

The addition of drilling waste in the range of 2-10% (v/v) to soil influences growth of the test species advantageously, significantly increasing nearly all values of vegetative parameters. The wet stem biomass of plants growing in series with 2 or 5% waste addition was over four times higher than that obtained on the studied soil (Fig. 1). The dry stem biomass obtained in series S+2%B+2%DW and S+2%B+5%DW was almost three times grater than the one for plants growing on soil alone (Fig. 2). The dry root biomass of orchard grass on soil enriched with 2 or 5% waste addition exceeded that found for series S by 126-141% (Fig. 3). The total dry biomass obtained in series S+2%B+2%DW and S+2%B+5%DW was over 2.5 times higher than the one of plants growing on soil alone (Fig. 4). It should be noticed here that there were no statistically significant differences in values of vegetative parameters between media series S+2%B+2%DW and S+2%B+5%DW.

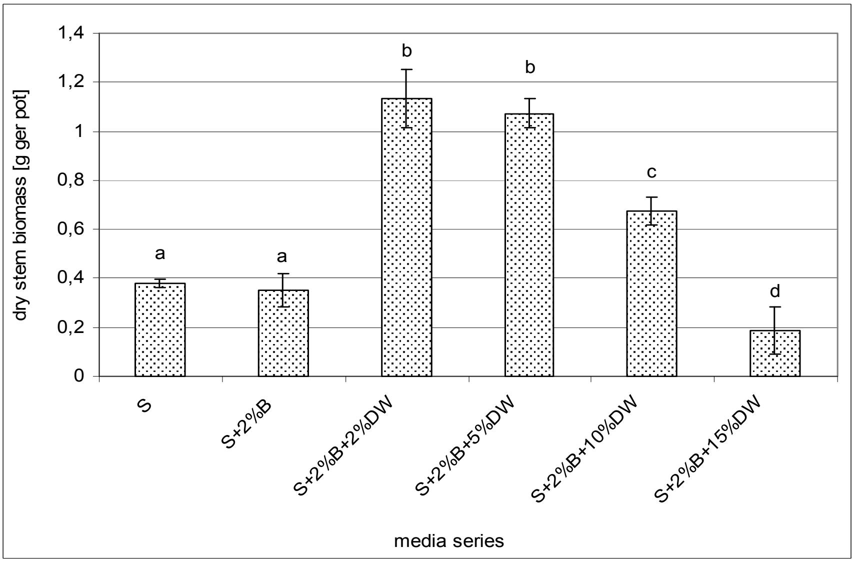

Fig. 2. Mean dry stem biomass of orchard grass in media series. S – soil; B – Biona-312; DW – drilling waste; different letters above bars indicate significant differences between mean values; I – standard deviation
Rys. 2. Średnia sucha biomasa pędów kupkówki pospolitej w seriach doświadczenia wazonowego. S – gleba; B – Biona-312; DW – odpad wiertniczy; różne litery nad słupkami wskazują istotne różnice między wartościami średnimi; I – odchylenie standardowe

As mentioned above, 10% waste addition to soil also positively affected grass growth, although the increase in values of vegetative parameters attributable to this addition was not as high as the one observed for 2 or 5% doses of drilling waste. Namely, wet and dry stem biomass, dry root biomass and total dry biomass of plants growing on soil supplemented with 10% waste addition were higher than those obtained on soil alone by 177%, 76%, 9% and 44%, respectively. At the same time all vegetative parameters for test species growing in series S+2%B+10%DW were significantly lower than those determined for media series supplemented with 2% Biona addition and 2 or 5% waste dose (Figs 1-4).

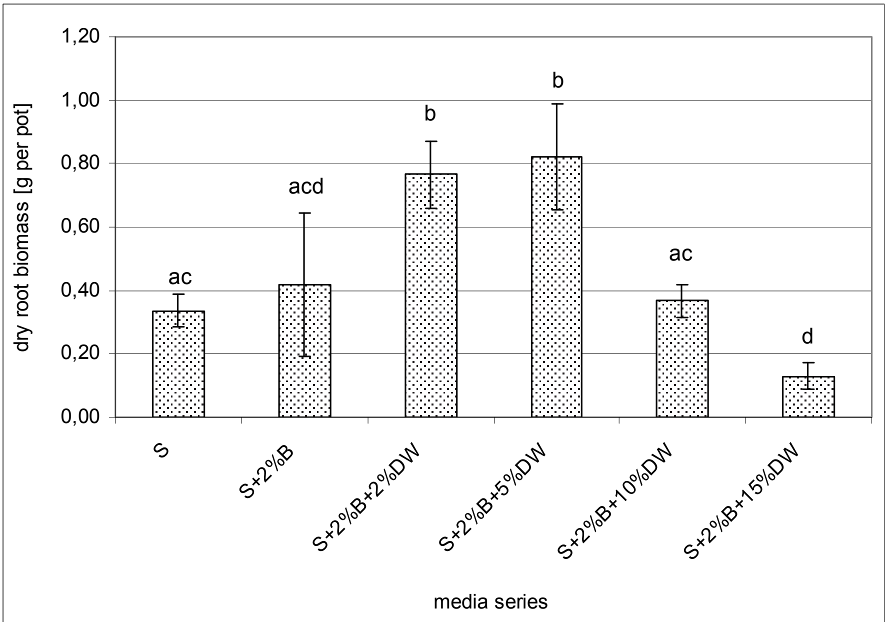

Fig. 3. Mean dry root biomass of orchard grass in media series. S – soil; B – Biona-312; DW – drilling waste; different letters above bars indicate significant differences between mean values; I – standard deviation
Rys. 3. Średnia sucha biomasa korzeni kupkówki pospolitej w seriach doświadczenia wazonowego. S – gleba; B – Biona-312; DW – odpad wiertniczy; różne litery nad słupkami wskazują istotne różnice między wartościami średnimi; I – odchylenie standardowe

The tendencies of increase in vegetative parameter values observed for media series enriched with 2% Biona addition and with 2, 5 or 10% waste dose as compared to soil alone were similar to those as compared to soil plus 2% Biona addition. Specifically, wet stem biomass of plants in series with 2, 5 or 10% waste addition was 2-3.5 times higher than that obtained in the control series (Fig. 1). The dry stem biomass obtained in series S+2%B+2%DW, S+2%B+5%DW and S+2%B+

10%DW was 2-3 times grater than the one for plants growing on soil plus 2% Biona addition (Fig. 2). The total biomass of orchard grass on soil enriched with 2, 5 or 10% waste addition exceeded that found for the soil only supplemented with Biona substrate by 35-147% (Fig. 4). The dry root biomass obtained in series S+2%B+2%DW or S+2%B+5%DW was almost two times higher than the one of plants growing in series S+2%B (Fig. 3). Conversely, the root biomass of plants growing on soil with 10% waste addition was lower than the one obtained in the control series; however, the difference in value of this parameter was not statistically significant between the compared media series (Fig. 3).

Contrary to waste doses in the range of 2-10%, 15% addition introduced into 2% Biona-soil mixture negatively influenced a vegetation of orchard grass, significantly reducing most of values of growth variables. Wet stem biomass of plants in series S+2%B+15%DW was lower than that obtained in series S and S+2%B by 28% and 41%, respectively (Fig. 1). Dry stem biomass of orchard grass on soil supplemented with 15% waste addition was about 2 times lower than the one on soil alone and soil only enriched with Biona substrate (Fig. 2). Dry root biomass found in series S+2%B+15%DW was reduced as compared to that obtained in series S and S+2%B by 62% and 69%, respectively (Fig. 3). Total dry biomass of plants growing on soil with added 15% waste was over 2 times lower than the one found in series S and S+2%B (Fig. 4). The drop in total plant yield at 15% waste dose observed in our studies, was higher than that observed for *Festuca rubra* L. by Czekaj et al. (2006). In that case, the total dry biomass of *F. rubra* growing on soil supplemented with 10-25% waste addition was 29% lower as compared to the one on soil alone.

It seems that the influence of waste additions on plant growth and hence, on soil productivity, was connected with media pH. The soil used in the present study was characterized by pH value equal to 4.77, which was below the range of 5.6-7.8 described as the one at which most of crop plants grow (Lityński & Jurkowska 1982). Parallel additions of Biona substrate and drilling cuttings to soil in 2 or 5% doses (accompanied by the highest plant yield) increased pH media to the level 6.5-7.0 – as mentioned above – the optimal one for cultivation of most plants (Lityński & Jurkowska 1982). The application of 10% waste dose increased pH medium to the level of 7.2, which could be a reason for lower increase in plant yield in series S+2%B+10%DW, as compared to the increases obtained in media series with 2 or 5% waste additions. However, despite the afore-mentioned lower increase in plant biomass, it seems that 10% waste dose is acceptable as well for supplementation of acidic soils. At the 15% waste dose, a sharp decrease in biomass of orchard grass was observed and the pH value for soil-2% Biona-15% waste mixture was 7.47, which was above the optimal pH range.

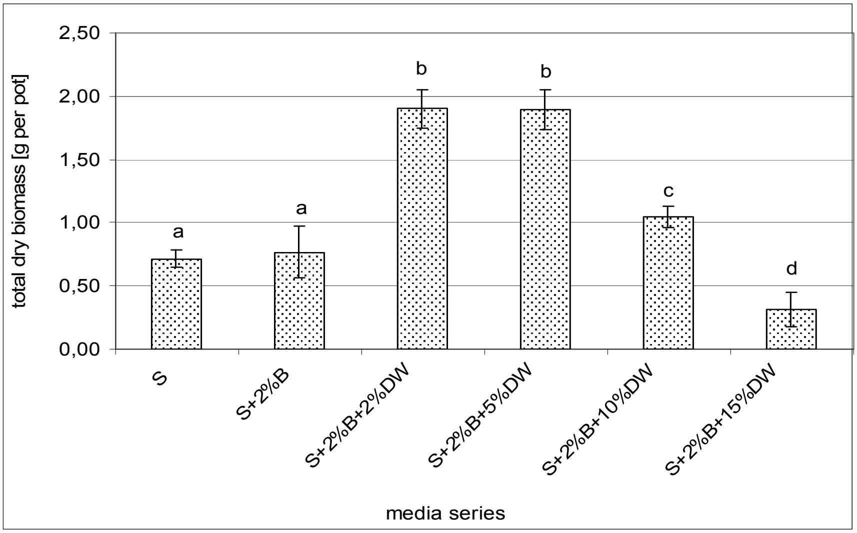

Fig. 4. Mean total dry biomass of orchard grass in media series. S – soil; B – Biona-312; DW – drilling waste; different letters above bars indicate significant differences between mean values; I – standard deviation
Rys. 4. Średnia całkowita sucha biomasa kupkówki pospolitej w seriach doświadczenia wazonowego. S – gleba; B – Biona-312; DW – odpad wiertniczy; różne litery nad słupkami wskazują istotne różnice między wartościami średnimi; I – odchylenie standardowe

It should be noticed that an inhibition of seed germination was observed in the S+2%B+10%DW and S+2%B+15%DW series. The mean number of germinated seeds on soil supplemented with 10% and 15% waste addition was respectively 2 times and 4-5 times lower than that on soil alone, soil-2%Biona mixture, soil-2%Biona-2%waste mixture and soil-2%Biona-5%waste mixture. Kisic et al. (2009) also observed the negative influence of drilling waste addition to soil on the growth of *Trit-*

icum aestivum L. and *Glycine hyspida* L. in the germination/emergence stage resulting in lower number of mature plants. These authors reported that *T. aestivum* and *G. hyspida* density in media series with waste additions was lower by 2-41% and 36-64%, respectively, in comparison to that on the control soil.

4. Conclusions

The analysis of the study results allows for formulating the following conclusions:

1. The media consisting of acidic soil mixed with drill cuttings and ion-exchange substrate were characterized with varied productivity, depending on the amount of drill cuttings introduced,
2. From the point of view of media productivity, 10% v/v drill cuttings addition was the maximum amount which resulted in about 50% productivity increase, in relation to the media without drilling waste addition,
3. The addition of drilling waste to soil in the amount of 5% (v/v) influences the growth of the test species advantageously, significantly increasing the values of all vegetative parameters,
4. The performed studies pointed that drill cuttings, coupled with the addition of ion-exchange substrates, could be applied for the reclamation of acidic soils,
5. The practical application of drilling waste should be preceded by a comprehensive evaluation of their impact on the changes of reclaimed soils properties.

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Zastosowanie odpadów wiertniczych do rekultywacji gleb kwaśnych

Streszczenie

Praca dotyczy nowej metody zagospodarowanie odpadów wiertniczych powstających w procesach poszukiwania gazu z formacji łupkowych. Stosowane metody zagospodarowania tego rodzaju odpadów ograniczają się w praktyce do ich deponowania na składowiskach odpadów niebezpiecznych. Z punktu widzenia ochrony środowiska rozwiązanie takie jest niepożądane i uzasadnia potrzebę poszukiwania nowych metod utylizacji. Odpady wiertnicze to głównie zwierciny, czyli urobek skalny wydostający się na powierzchnię wraz z płuczką wiertniczą. Właściwości fizyczne i chemiczne odpadów wskazują, że mogą być one wykorzystane przyrodniczo do rekultywacji gleb. Biorąc pod uwagę alkaliczny odczyn zwiercin ich dodatek powinien przyczynić się do poprawy właściwości gleb kwaśnych.

Celem prezentowanych badań było określenie wielkości maksymalnej dawki zwiercin z uwagi na zmiany produktywności rekultywowanej gleby. W przeprowadzonych badaniach użyto gleby średnio kwaśnej, a dopuszczalną ilość dodawanych zwiercin ustalono na poziomie 15% obj. w oparciu o pomiary pH komponowanych podłoży. Wyniki analiz zasobności gleby wskazały na potrzebę wzbogacenia badanych podłoży w makroskładniki, co osiągnięto przez zastosowanie 2% dodatku substratu jonitowego.

Przeprowadzony eksperyment wegetacyjny wykazał, że podłoża przygotowane na bazie zakwaszonej gleby z dodatkami zwierciny i substratu jonitowego charakteryzowały się różną produktywnością zależną od ilości wprowadzonych odpadów wiertniczych. Najkorzystniejszym dla rozwoju rośliny testowej (kupkówka pospolita, *Dactylis glomerata* L.) okazał się dodatek zwierciny w ilości około 5% obj. Zaobserwowano wówczas ponad 2,5-krotny wzrost produktywności mierzony całkowitą suchą biomasą roślin. Maksymalnym dodatkiem zwierciny z punktu widzenia produktywności podłoża był dodatek wynoszący około 10% obj., przy którym obserwowany wzrost produktywności w odniesieniu do podłoży bez dodatku zwiercin wynosił około 50%. Przeprowadzone badania potwierdziły możliwość wykorzystania odpadów wiertniczych w rekultywacji gleb kwaśnych w połączeniu z dodatkiem substratów przygotowanych na bazie żywic jonowymiennych. Rozwiązanie takie stwarza dodatkowo przesłanki do kompleksowego zagospodarowania zużytych lub poeksploatacyjnych wymieniaczy jonowych.

Słowa kluczowe:
odpady wiertnicze, gleby kwaśne, substraty jonitowe, rekultywacja gleb

Keywords:
drilling waste, acidic soils, ion exchange substrate, soil reclamation