SELECTED HAZARDS TO QUALITY OF GROUNDWATER RESULTING FROM THE RECLAMATION OF AN OPEN PIT WITH WASTE ROCKS - A CASE STUDY OF A BACKFILL SAND MINE

WYBRANE ZAGROŻENIA DLA JAKOŚCI WÓD PODZIEMNYCH ZWIĄZANE Z REKULTYWACJĄ WYROBISKA ODKRYWKOWEGO ZA POMOCĄ ODPADÓW POGÓRNICZYCH – STUDIUM PRZYPADKU KOPALNI PIASKU PODSADZKOWEGO

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The use of rock wastes after mining and processing of hard coal for reclamation of opencast workings is a common form of restoring utility functions to these areas. The origin of these wastes, their properties, petrographic and mineral composition as well as the method and time of depositing in the excavation directly affect the emissions to the soil and water environment of pollutants, which may affect the quality of groundwater in the area of the open pit and significantly affect the possibility of their economic use. On the example of CTL Maczki-Bór S.A. backfill sand mine an analysis of the leachability of selected components from post-mining wastes, differentiated in terms of their deposition time and exposure to external factors, was performed. The article presents the results of analyzes of the content of components (elements and/or ions) in the form of their concentrations in the aqueous solution and per unit of dry mass of the tested wastes. The correlation was assessed between the concentration values of selected components, obtained in laboratory tests in pore extracts, and those observed in-situ in the drainage well made in the waste dump.

Keywords: *open pit, reclamation, post-mining wastes, leachability, pollutants, groundwater*

Stosowanie do rekultywacji wyrobisk odkrywkowych materiału skalnego stanowiącego odpad po wydobyciu i przeróbce węgla kamiennego jest powszechną formą przywracania tym terenom funkcji użytkowych. Pochodzenie tych odpadów, ich właściwości, skład petrograficzny i mineralny oraz sposób i czas deponowania w wyrobisku wpływają bezpośrednio na zakres emisji zanieczyszczeń do środowiska gruntowo-wodnego. Zanieczyszczenia te w różnym stopniu mogą kształtować jakość wód podziemnych w rejonie odkrywki i istotnie wpływać na możliwość gospodarczego ich wykorzystania. Na przykładzie kopalni piasku podsadzkowego CTL Maczki-Bór S.A. dokonano analizy wymywalności wybranych składników z odpadów pogórniczych, zróżnicowanych pod kątem czasu ich deponowania i ekspozycji na czynniki zewnętrzne. Analizie poddano zawartość składników (pierwiastków i/lub jonów) w formie ich stężeń w roztworze oraz w przeliczeniu na jednostkę suchej masy badanych odpadów. Dokonano oceny korelacji pomiędzy wartościami stężeń wybranych składników uzyskanymi w badaniach laboratoryjnych w wyciągach porowych, a obserwowanymi in-situ w studzience odciekowej wykonanej na zwałowisku skał płonnych.

Słowa kluczowe: *wyrobisko odkrywkowe, rekultywacja, odpady pogórnicze, wymywalność, zanieczyszczenia, wody podziemne*

Introduction

The binding formal and legal conditions require entrepreneurs to reduce the risk of environmental pollution (including the soil and water environment) in connection with their business activities (Environmental Protection Law of April 27, 2001. [Dz. U. 2001 no 62 pos. 627 with subsequent changes]). Hence, the reclamation of opencast workings by backfilling can be carried out only with the use of inert materials, i.e. environmentally neutral. One of the groups of wastes approved by the legislator for use in backfilling open-pit workings are waste rocks, which are material from exploitation and processing, among others hard coal deposits. These wastes are marked with 01 01 02 code and due to their properties they are not treated as hazardous waste (Regulation of the Minister of Climate of 2 January 2020 on the waste catalog [Dz.U. 2020, pos. 10]). Under natural conditions, i.e. in the redox environment, where Carboniferous sandstones, siltstones and claystones are water-saturated, rock-forming minerals (mainly sulphide minerals such as pyrite or marcasite) do not pose a threat to the quality of groundwater. However, after drainage of the rock mass, after the exploitation of waste rocks accompanying the coal deposits, and then after depositing them in heaps or dumps (including central dumps in former opencast excavations), intensive weathering processes are initiated, causing the transformation of some rock-forming minerals and oxidation of sulphide minerals and emission of sulphate ions to the soil and water environment (Twardowska et al. 1988, 2004, Twardowska, Szczepańska 1995, Younger, Wolkerdorsfer 2004, McCloskey, Bless 2005, Banks et al.

1997, Rashidinejad et al. 2008). The scale and intensity of this process depend on the original content of components susceptible to weathering in the lump of post-mining wastes used in excavation reclamation, as well as on the technology of depositing, compacting and sealing the rock debrises, and on the scale of their water saturation. The formation of the content of elements in the mixture of wastes deposited in the excavation in the total and leachable form is significantly influenced by the age of the waste (Klojzy-Kaczmarczyk et al. 2016).

In order to illustrate the influence of the longest possible time of the impact of external factors on post-mining wastes deposited in the open pit, which shape the current qualitative condition of the soil and water environment, the research was carried out on a reclaimed open pit CTL Maczki-Bór S.A. It is a backfilling sand mine in the final period of its operation, located within the Upper Silesian Coal Basin (USCB). Since 1972, it has been filled with Carboniferous waste rocks. This material is a mixture of sandstones, mudstones and claystones with an admixture of hard coal in various proportions depending on their place of origin. Throughout the reclamation period, the wastes were sourced from various suppliers, from almost all hard coal mines in the USCB, and from all productive carbon lithostratigraphy cells. These rocks are characterized by a significant diversification of physical and mechanical parameters and a wide range of strength depending on the state of their saturation with water (Bukowska 2012). Rock rubble at the place of its deposition was and still is thickened in layers, and since 1994 it has also been caulked, e.g. by domestic clay material and, until 2004, also power plant waste in the form of water pulp.

The effects of intensive weathering processes (which taking place in the dump) and washing the lump of waste with infiltration water, are reflected in the chemical composition of groundwater, observed within the boundaries of the excavation. In piezometers drilled in the waste dump, groundwater in the excavation base is monitored, which most often shows a significantly increased level of sulphate, chloride, calcium, sodium, iron and manganese ions. However, the quality of these waters may also be affected by the impacts from the nearby municipal waste landfill (sub-level and above-level) located in the northern part of the closed part of the CTL Maczki-Bór S.A. mine. In the current hydrodynamic condition (drainage of the excavation is carried out), the impact range of post-mining waste is limited to the limits of its occurrence, and the mixed groundwater drained by the mine drainage system, after removing the suspended solids, is discharged to the Biała Przemsza River. The waters discharged into the receiver meet the quality criteria for wastewater in accordance with the Regulation of the Minister of Maritime Economy and Inland Navigation of July 12, 2019 on substances particularly harmful to the aquatic environment and the conditions to be met when discharging sewage into water or soil, as well as when discharging rainwater or snowmelt into waters or into water facilities [Dz.U. 2019, pos. 1311]. After the completion of the reclamation and exploitation of the sand and the cessation of the excavation drainage, the migration of pollutants will follow the direction of groundwater filtration, ie to the south-west of the excavation boundaries.

Research methodology

Rock rubble samples with different time spent on the embankment were selected for laboratory tests. It was a rock material: younger than half a year, 5-year-old, 15-year-old, 15-year-old with earlier coal recovery and older than 35 year--old. According to the division of waste depending on the time of its storage (Skarżyńska 1997, Mirkowski, Badera 2015), this material is waste: fresh, partially weathered, weathered and very weathered. The samples were taken from the top of the dump, after removing the top layer of the weathered material, in places covered with vegetation, from the natural process of succession.

The leachability tests were performed on the basis of the methodology in accordance with PN-EN12457-4:2006 norm, on air-dry samples. The elution of components was performed using the leachability test in the ratio 1:10 (solid/liquid phase), ie 0,1 kg of wastes/1 dm^3 of demineralized water. In addition, the obtained values were converted into the amounts of pollutants released from the waste per dry weight unit of the sample (expressed in mg/kg) in accordance with the following formula (Klojzy-Karczmarczyk et al. 2016).

$$A = C \cdot \left(\frac{L}{M_D}\right)$$

where:

A – the amount of contaminating component released, i.e. the amount of washout, for a solid to liquid ratio 1:10 [mg/kg],

C – component concentration in the eluate (pore extracts) [mg/dm^3],

L – the volume of water used in the tests [dm^3],

M_D –dry mass of the analytical sample [kg].

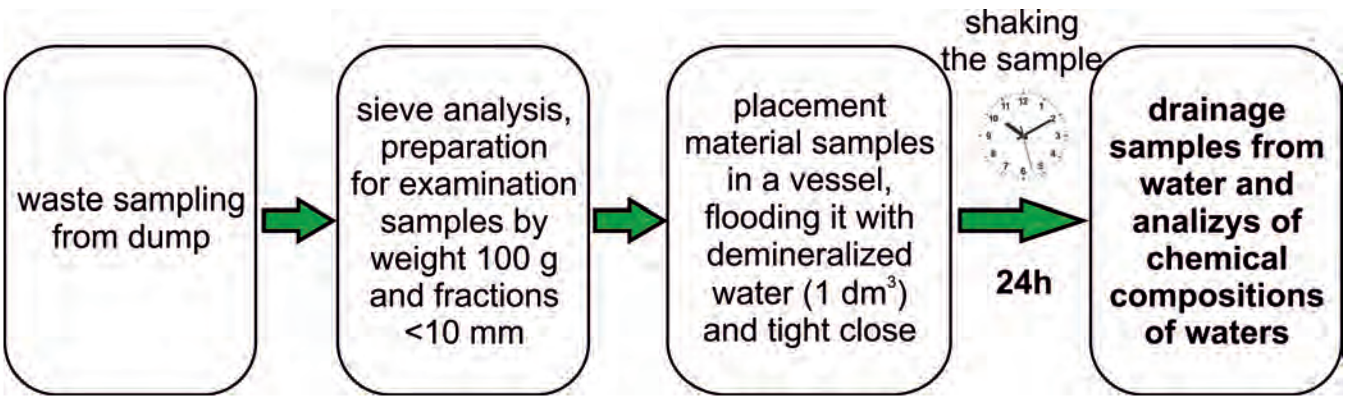

Fig. 1. Scheme of the study of the leachability of pollutants from post-mining wastes
Rys. 1. Schemat przebiegu badania wymywalności zanieczyszczeń z odpadów pogórniczych

In accordance with the requirements of the standard, 0,1 kg of rock material with a fraction below 10 mm was collected for laboratory tests. The markings were carried out in accordance with the diagram shown in Figure 1.

Characteristics of leaching of pollutants from wastes used in the reclamation of opencast excavations

Each material provided by the producer of waste for recovery in the form of raw material used in the reclamation of opencast workings, leveling the land surface as well as in road, railway and hydrotechnical construction is the subject of prior analyzes of its physico-chemical condition. However, the analysis covers only the so-called fresh wastes, which in the case of the CTL Maczki-Bór S.A. dump are only deposited locally. Pollution indicators in water extracts are tested by entrepreneurs (suppliers of material for reclamation) for wastes, e.g. from coal enrichment and for energy wastes. Archival analyzes of fresh wastes from selected hard coal mines (suppliers of waste rock rubble) indicate that the average concentrations of chloride ions in water extracts from waste range from 0,5 to 133,6 mg/dm^3, and for sulphates from 7,0 to 135,0 mg/dm^3. For energy wastes used in caulking the dump (i.e. fluidized sands, slag-ash mixture, fly ashes and slags), these concentrations for chloride ions are in the range of values from 0,5 to 313,0 mg/dm^3, and for sulphate ions from 42,0 to 1868,0 mg/dm^3.

The content of several dozen ingredients was determined in laboratory tests. The results for selected components are presented in the diagram (Fig. 2) and summarized in Table 1 in the form of their concentrations in the pore extracts, as well as converted into the amount of leaching of these elements per kilogram of waste mass. The obtained values were compared with the limit values established for wastewater discharged into water or into the ground (Regulation of the Minister of Maritime Economy and Inland Navigation of July 12, 2019 on substances particularly harmful to the aquatic environment and the conditions to be met when discharging sewage into water or soil, as well as when discharging rainwater or snowmelt into waters or into water facilities [Dz.U. 2019, pos. 1311]), as well as with the criteria to be met by waste authorized for storage in landfills (Waste Act of December 14, 2012 [Dz. U. 2012 pos. 21 with subsequent changes]), although, according to the law, the reclaimed excavation is not a landfill, but a disposal facility. None of the analyzed components in the tested samples exceeded the permissible concentrations contained in the above-mentioned legal acts.

Reaction of tested water extracts (pH) from post-mining wastes deposited in the excavation of CTL Maczki-Bór S.A. depending on the time of their storage on the dump, ranged from 6,30 to 7,15 (slightly acidic to slightly alkaline reaction). In general, the lower the pH, the faster the leaching of heavy metals from the waste takes place (Alloway 1995). This is reflected in the test results, where in the case of lead, higher concentrations were observed in six-month and 15-year samples, for which the pH values ranged from 6,30 to 6,59. Much lower concentrations of lead were found in 5-year waste, for which the pH of water from pore extracts was 7,15. Generally, in the tested water extracts, apart from the presence of lead and zinc, no other heavy metals were found in concentrations higher than the limits of their quantification in the used analytical method. This may indicate the presence of other heavy metals in the waste in forms sparingly soluble in water and/or a significant washing out of these components in the long-term exposure of post-mining waste to external factors.

Tab. 1. The concentration of selected components in water extracts and the amount of their leaching from post-mining waste used in the reclamation of the open pit
Tab. 1. Zestawienie koncentracji wybranych składników w wyciągach wodnych oraz wielkości ich wymywania z odpadów pogórniczych stosowanych w rekultywacji odkrywki

Sample	The concentration of ingredients in pore extracts (mg/dm^3)				pH
	Cl^-	SO_4^{2-}	Zn	Pb	
< 0,5 year	11,32	116,0	0,0068	0,0097	6,59
5 years	32,01	95,0	0,0212	0,0011	7,15
15 years	2,08	197,0	0,0210	0,0089	6,55
15 years*	4,17	95,0	0,0322	0,0108	6,30
>35 years	4,62	94,0	0,0028	0,0045	6,90
The amount of elution of components (mg/kg dry mass)					
< 0,5 year	113,2	1160	0,068	0,097	
5 years	320,1	950	0,212	0,011	
15 years	20,8	1970	0,210	0,089	
15 years*	41,7	950	0,322	0,108	
>35 lat	46,2	940	0,028	0,045	

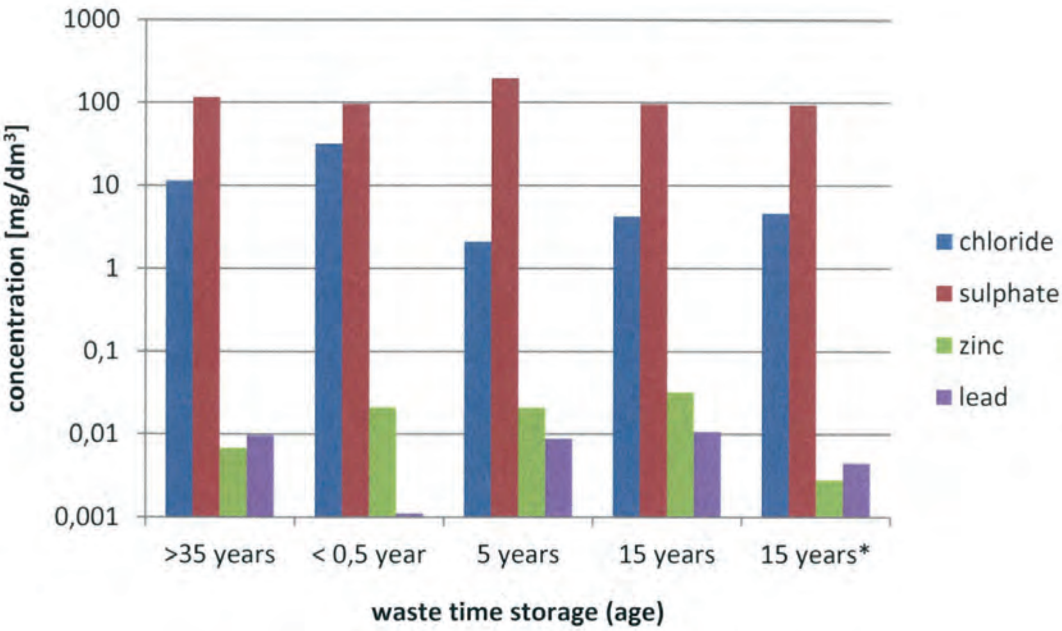

Fig. 2. Concentrations of selected components in water extracts from post-mining waste of different ages
Rys. 2. Stężenia wybranych składników w wyciągach wodnych z różnowiekowych odpadów pogórniczych

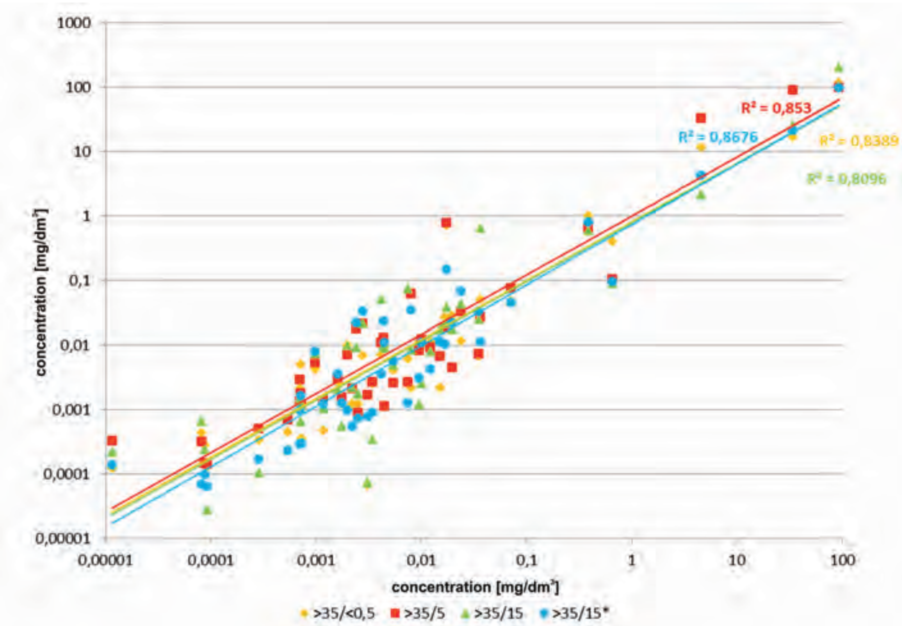

Fig. 3. Correlation graphs of concentrations of all determined components between the oldest sample and other samples (>35/0.5 - between the 35-year-old and the six-month-old sample; >35/5 - between the 35-year and the 5-year sample; >35/15 - between the 35-year-old and the 15-year-old sample; >35/15 * - between the 35-year-old and the 15-year sample after coal recovery).

Rys. 3. Wykresy korelacji stężeń wszystkich oznaczonych składników między próbką najstarszą, a pozostałymi próbkami (>35/0,5 – między próbką 35-letnią a półroczną; >35/5 – między próbką 35-letnią a 5-letnią; >35/15 – między próbką 35-letnią a 15-letnią; >35/15* – między próbką 35-letnią a 15-letnią po odzysku węgla)

The main ions in the composition of water extracts from the tested samples are chloride and sulphate ions. The results of the conducted tests indicate that they are significantly different for all samples with various time of exposure to weather conditions. In the case of chlorides, trends in concentration changes are generally consistent with those observed in post-mining waste dumps in the USCB (Szczepańska, Twardowska 1986). In the oldest wastes (15-year and 35-year old), as a result of many years of weathering processes, a significant part of chloride ions leached out, which together with rainwater infiltrated through the aeration zone into the groundwater, and then into the mine drainage system, from where they were discharged to Biała Przemsza. Hence, probably the concentrations of chloride ions in these samples are significantly lower than in the younger material. On the other hand, sulphate ions, as substances with lower leaching kinetics than chlorides, show much higher concentrations in all waste samples. The concentration of these components (and all others) in water extracts is also determined by their initial concentration in fresh waste transferred by hard coal mines for excavation reclamation.

In all samples of waste, the presence of determined components in similar proportions was found. This is evidenced by the correlation diagrams (Figs. 3, 4, 5) and the coefficients of determination defined mutually between the concentrations of these components for all samples.

The coefficient of determination is a descriptive measure of the fit of a regression model to data variability. According

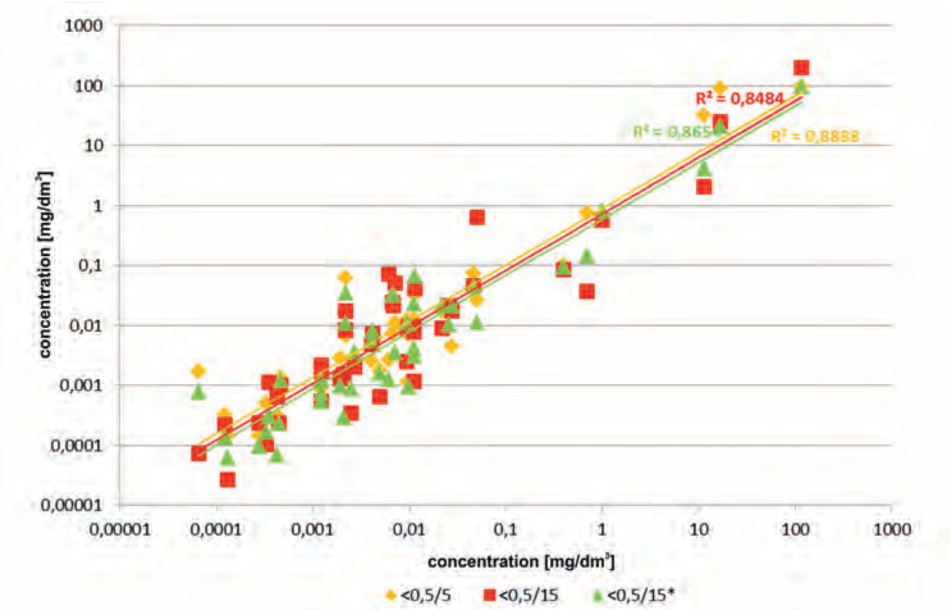

Fig. 4. Correlation graphs of concentrations of all determined components between the youngest sample and between the 5-year-old sample and the 15-year-old samples (<0.5/5 - between the six-month-old and the 5-year-old sample; <0.5/15 - between the six-month-old sample and the 15 years-old samples; <0.5/15 * - between the six-month-old sample and the 15 years-sample after coal recovery)

Rys. 4. Wykresy korelacji stężeń wszystkich oznaczonych składników między próbką najmłodszą, a próbką 5-letnią i próbkami 15-letnimi (<0,5/5 – między próbką półroczną a 5-letnią; <0,5/15 – między próbką półroczną a 15-letnią; <0,5/15* – między próbką półroczną a 15-letnią po odzysku węgla)

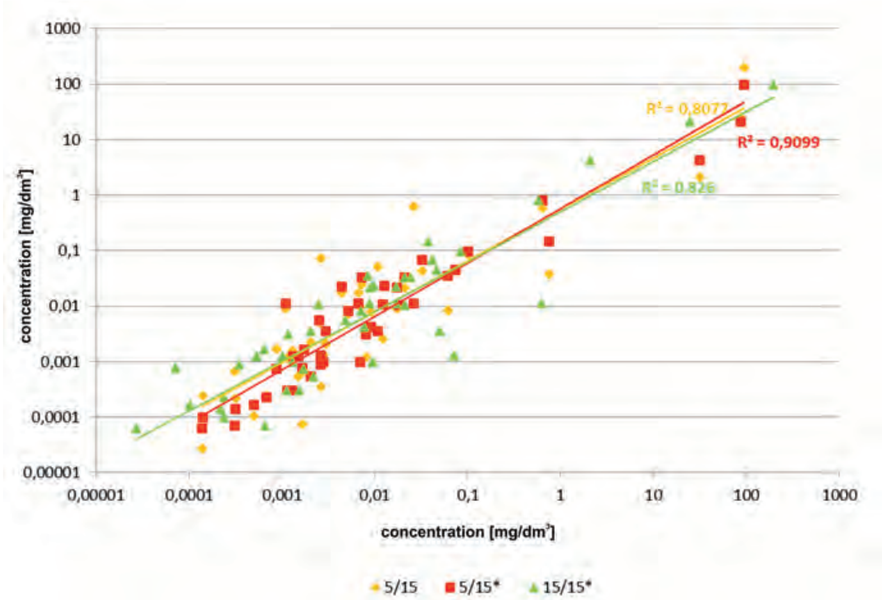

Fig. 5. Correlation graphs of concentrations of all determined components between the 5-year-old sample and the 15-year-old samples, and between the 15-year-old samples before and after coal recovery (5/15 - between the 5-year-old and 15-year-old sample; 5/15 * - between the 5-year-old and 15-year-old sample after coal recovery; 15/15 * - between the 15-year-old and 15-year-old sample after coal recovery)

Rys. 5. Wykresy korelacji stężeń wszystkich oznaczonych składników między próbką 5-letnią, a próbkami 15-letnimi, oraz pomiędzy próbkami 15-letnimi przed i po odzysku węgla (5/15 – między próbką 5-letnią a 15-letnią; 5/15* – między próbką 5-letnią a 15-letnią po odzysku węgla; 15/15* – między próbką 15-letnią a 15-letnią po odzysku węgla)

to the adopted division, $0,9<R^2\leq1$ characterized a very good match, $0,7<R^2\leq0,9$ good match, and $0,7<R^2\leq0,4$ mean match. For all analyzed samples of waste (pore water from waste) the regressions explain from 80,77% to 90,99% of data variability, therefore, according to the adopted division, the match of all models can be described as good and very good, i.e. the determined concentrations of all components show a significant correlation between individual samples. Therefore, no significant increase or decrease in the concentration of selected ions/elements in relation to the concentration of other components was found in any of the pore water samples.

Moreover, the correlation between the physico-chemical composition of water from water extracts and seepage water from a well drilled in a waste dump was also assessed. A sample of 15-year-old waste was selected for analysis, due to its relatively close location to the well, as well as the convergence of the sampling time. From among all the components determined in the pore extracts, six were selected (chlorides, sulphates, bicarbonates, calcium, magnesium, sodium) as reference elements and ions, also determined in the tests of water quality taken from the drainage well. The results are presented graphically in Figure 6. They show a good correlation between the concentrations of the adopted components, thus confirming the correctness of the adopted methodology and the assessment of leachability of pollutants from post-mining waste used for reclamation of the opencast excavation of CTL Maczki-Bór S.A.

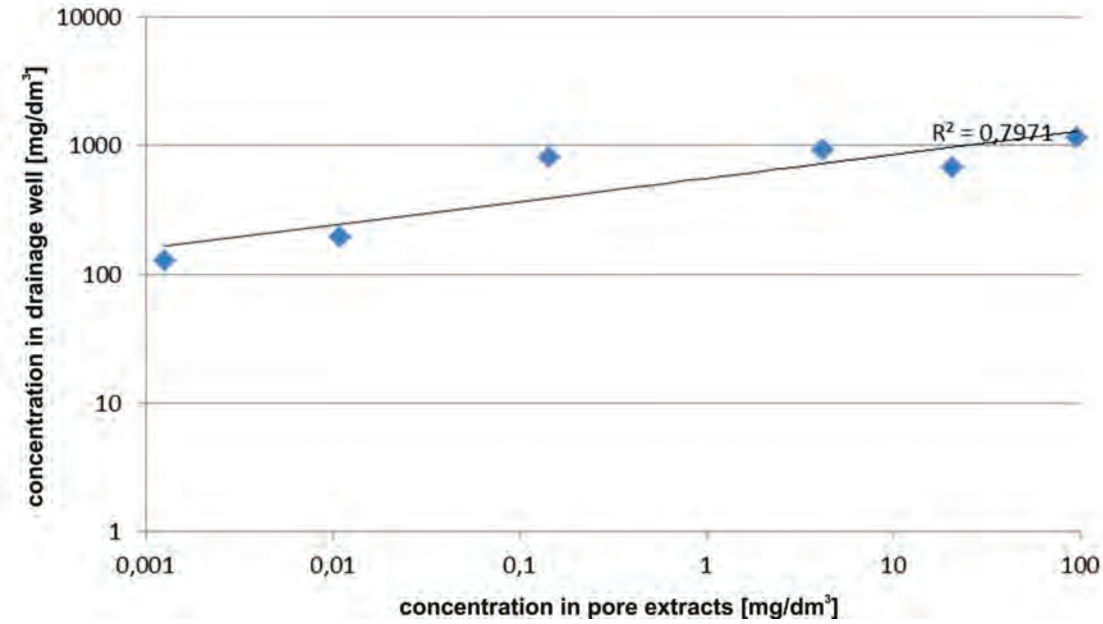

Fig. 6. Correlation diagram of concentrations of selected components in pore extracts in the 15-year-old sample and in leachate waters in a well drilled on a waste dump
Rys. 6. Wykres korelacji stężeń wybranych składników w wyciągach porowych w próbce 15-letniej oraz w wodach odciekowych w studzience odwierconej na zwałowisku skał płonnych

Summary

The scope and extent of the impact of the reclaimed opencast excavation on the quality of groundwater is the result of many natural and technical factors. However, it largely depends on the amount and type of material used to level the post-exploitation void. On the example of an opencast backfill sand excavation, research was carried out on the leaching of pollutants from post-mining waste (Carboniferous waste rocks) with different duration of their storage on the dump (from half a year to over 35 years). The method and place of sampling were to represent the diversity of the collected material as broadly as possible, mainly in terms of its physicochemical and hydrogeological parameters.

The tests, carried out on samples prepared for this purpose in a laboratory scale, indicate a significant differentiation of the concentrations of individual components between all the rock rubble samples. In the case of chloride and sulphate ions, their concentrations and changes over time show the tendencies of changes typical for post-mining waste, which have been observed for many years in the Upper Silesian Coal Basin on waste heaps and dumps. The reactions of these waters found in pore extracts (reaction from slightly acid to weakly alkaline) create conditions that hinder the release of other pollutants, e.g. from the group of heavy metals, which is confirmed by very low concentrations of zinc and lead in pore waters and concentrations of other heavy metals below the limit of their quantification.

The current hydrogeochemical condition around the opencast excavation and within its boundaries is the subject of groundwater and surface water monitoring to the range specified by the legislator. After completion of mine drainage and finishing the reclamation of the opencast excavation, the stream of pollutants will flow, along with the direction of groundwater filtration, to the south-west of the boundaries of the excavation. Concentrations of pollutants in groundwater will, however, depend on their initial concentration in waste and the dynamics of weathering processes that transform rock-forming minerals. The expected scope and extent of this phenomenon can be predicted using mathematical modeling methods, including hydrodynamic modeling, mass transport modelling and modelling the dynamics of chemical processes occurring in the rock/water environment.

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South Pesteana pit lake, Romania