

COAL MINING WASTE MANAGEMENT AND ITS IMPACT ON THE GROUNDWATER CHEMICAL STATUS EXEMPLIFIED IN THE UPPER SILESIA COAL BASIN (POLAND)

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Abstract. Sulfidic coal mining wastes constitute one of the the major global bulk waste stream and are extensively reused in engineering constructions. Occurrence in the waste rock of geochemically instable sulfides results in A/NRD (Acid/Neutral Rock Drainage) that makes this material environmentally problematic. The paper is focused on the impact assessment of coal mining waste management practices exemplified in different objects (coal mining waste dumps) and different waste management methods (residual coal extraction, embankment construction, ground leveling and using as a structural fill of a waterlogged quarry) on chemical status of groundwater. Long-term groundwater monitoring data in the vicinity of three dumping sites and the analysis of pore solutions extracted along the vertical profiles of the waste layers of different ARD generation potential shows that high sulfate salinity, which resulted from sulfide oxidation, is the major long-term source of strong groundwater deterioration caused both by ARD and NRD. Abundant constituents released by ARD are Mn and in anoxic groundwater environment also Fe. The results show the need of application of groundwater protection measures both with respect to ARD and NRD generating coal mining waste.

Key words: coal mining waste, groundwater chemical status, waste disposal, waste reuse, coal extraction, engineering constructions, A/NRD generation.

INTRODUCTION

Hard coal is the major fuel worldwide, currently used for generating 41.5% of total world's electricity and 26.5% of global primary energy. Global hard coal production is permanently growing (3489 Mt in 1990, 5442 Mt in 2007, 5845 Mt in 2008) (WCI, 2010). Of top ten hard coal producers, China, USA and India are the leaders with almost 73% of the global output. Poland remains the largest hard coal producer in the EU – 84 Mt in 2008 (GUS, 2009). In comparison with the peak output in eighties (266.5 Mt in 1988), this means nearly 3-fold decline. Despite this, hard coal mining retains high position in the economy of Poland, as well as of the EU, and is still dynamically growing worldwide to meet increasing demand for power generation, the bulk of it coming from non-OECD fast developing countries (WCI, 2010).

Coal mining operations concentrate in a relatively small area of a coal mining basin, where also coal mining wastes are being disposed. In Poland, this is the Upper Silesia coal mining basin. Mechanized coal extraction and a subsequent stage of coal separation processes result in generation of mining waste at the level of 30–50% of coal output, which gives an estimate of global annual coal mining waste generation in the range of 1750–3000 Mt.

In Poland, coal mining waste generation in 2008 reached 30.86 Mt that accounted for 37% of hard coal output and was concentrated in the Upper Silesia administrative district. Of this amount, 89.7% was reused in civil engineering (for construction of flood banks and polders, railway and highway embankments, ground leveling, and reclamation of areas impacted by subsidence or industry), while 600.5 Mt was laying in dumps (GUS, 2009).

Practically all coal mining wastes, both disposed and reused, are being exposed to the atmospheric conditions. Thus,

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the safe waste management requires evaluation of their long-term environmental pollution potential pertain to the content of constituents susceptible to mobilization and leaching, and in particular to occurrence in the waste rock of geochemically instable sulfides (mostly FeS_2 – pyrite, markasite) resulting in Acid Rock Drainage (ARD) generation. This multi-stage process is commonly described by a summary reaction:

$$\operatorname{FeS}_{2(s)} + \frac{15}{4} \operatorname{O}_2 + \frac{7}{2} \operatorname{H}_2 \operatorname{O} \to \operatorname{Fe}(\operatorname{OH})_3 + 2\operatorname{SO}_4^{2-} + 4\operatorname{H}^4$$

The resultant ARD generation potential depends on the neutralizing potential of waste rock materials due to presence of sufficient amounts of carbonates that is often expressed as the Neutralization Potential Ratio NPR = NP/AP, where NP is the Neutralization Potential and AP is the Acid Potential. The relevant aspects of these processes occurring in extractive wastes have been presented elsewhere and are briefly summarized by Appelo and Postma, (2005) and BREF (2009). Leachate from mining waste that is close to neutral or alkaline Neutral Rock Drainage (NRD) may also have high pollution potential e.g. due to the elevated salinity. European extractive industries, including Polish mining, are now legally obliged to prevent or minimize any adverse effects on the environment and health risks resulting from the waste management. The management practice must comply with the provisions of the EU legal documents in force implemented to the Polish national legislation, including Directive 2006/21/EC on the management of waste from extractive industries, Water Framework Directive 2000/60/EC and Groundwater Directive 2006/118/EC. The Polish national correspondent legislative documents include Law of 10 July 2008, on the extractive waste (2008), Decree of Ministry of the Environment of 20 August 2008 on the qualification of the surface water status (DME, 2008a) and the Decree of Ministry of the Environment of 23 July 2008 on the criteria and methods of assessments on groundwater status (DME, 2008b).

In 2004 a Reference Document on Best Available Techniques (BAT) for Management of Tailing and Waste-Rock in Mining Activities was developed and was adopted by the EC in 2009 (BREF, 2009). In 2005 a Technical Committee CEN/TC 292/WG8 at the European Committee for Standardization (CEN) was also established for the development of standards and guidelines for the characterization and environmentally safe management of waste from extractive industries.

This paper is focused on the impact of different engineering constructions from coal mining wastes and of widely used waste management methods on chemical status of groundwater. It is aimed to the identification and reversal of upward trends in pollutant concentrations, in accordance to the Mining Waste Directive 2006/21/EC, Groundwater Directive 2006/118/EC and the relevant Polish legislation (Law on the extractive wastes, 2008; DME, 2008b).

WASTE DISPOSAL

Coal mining waste dumps are mostly constructed above the ground level, and sited in the disused sand or gravel quarries with a permeable bedrock. At such constructions, bulk coal extractive waste are exposed to atmospheric conditions, and take part in a natural water circulation. Soluble constituents are being leached from the rock by infiltrating water and transported through the vadose zone to groundwater and to draining it surface water. Contaminant loads occurring in freshly disposed coal mining wastes and generated during their physical and chemical weathering transformations constitute a long-term source of the aquatic environment deterioration, lasting for decades. The characteristics of the coal-bearing Carboniferous strata and of waste rock that influence the environmental impact of coal mining wastes have been presented in detail elsewhere (Szczepańska, Twardowska, 1999, 2004). In brief, the major factors pertain to the pollution potential from coal mining wastes are: (a) contents and reactivity of iron sulfides (pyrite, marcasite), i.e. kinetics of their oxidation and generation of sulfate and acidity determining the AP, (b) contents and dispersion of buffering constituents, mostly carbonate minerals - calcite, dolomite, determining the NP, (c) occurrence of Potentially Toxic Elements (PTE), i.e. metals mobilized by ARD; (d) occurrence of other soluble salts e.g. chlorides that may contribute to high Total Dissolved Solids (TDS) in A/NRD, adversely affecting groundwater chemical status downgradient of a dumping site.

Here, the character of adverse environmental impact of coal mining dumps is exemplified by data from the groundwater monitoring conducted prior, during and after the waste reuse activity in the vicinity of three dumping sites: (1) the Buków dump of the Anna coal mine; (2) the Dębieńsko dump of the closed Dębieńsko mine; (3) the Smolnica dump of the Szczygłowice mine (Fig. 1).

At the beginning of the ground- and surface water monitoring at these sites in 1997, 1999 and 1988, respectively, at the active, 20–30 years old dumps of the area from 45 (Buków) to 140 hectares (Dębieńsko), waste rock from coal separation was being disposed. It originated from the lower marine-brackish shale-sandstone Namurian A seams 600 and 700 (Buków), the upper continental limnic shale Westphalian A, B seams 300 (Dębieńsko) and from the shale-sandstone and sandstone Namurian A, B, C to the shale Westphalian A, B seams from 850 to 300 (Smolnica). The waste rock showed different potential for ARD generation related to NPR (BREF, 2009), from not potentially generating (Buków dump, NPR = 2.37), through possibly generating (Dębieńsko dump, NPR = 1.86) to likely ARD generating (Smolnica dump, NPR = 0.70).



Fig. 1. Location of the Buków, Dębieńsko and Smolnica coal mining waste dumps

Table 1

Chemical constituents exceeding MCL in pore solutions in undisturbed and re-disposed waste layers after residual coal extraction (Dębieńsko dump)

Constituent	Dębieńsko ¹⁾ pH = 4.54	$Dębieńsko^{1)}$ $pH = 5.50$	Dębieńsko ¹⁾ pH = 7.13	Dębieńsko ²⁾ pH = 6.51	MCL for groundwater of a good chemical status ³		
	mg/l						
Na ⁺	3.04	4.84	24.92	223.8	200		
K ⁺	20.38	27.24	67.50	31.80	15		
Ca ²⁺	558.2	773.6	386.8	837.4	200		
Mg ²⁺	266.9	310.4	318.4	729.9	100		
Mn ²⁺	28.52	18.98	0.080	22.03	1		
Zn ²⁺	3.506	1.398	0.022	0.464	1		
Ni ²⁺	0.708	0.467	0.007	0.442	0.02		
Co ²⁺	0.27400	0.00880	0.002	0.16000	0.2		
Hg ²⁺	0.0014	0.0017	0.0006	0.00018	0.001		
Cd ²⁺	0.0065	0.0035	0.0001	0.0058	0.005		
Al ³⁺	6.260	0.189	0.005	0.004	0.2		
SO_4^{2-}	2450	2916	2208	5568	250		

¹⁾ Undisturbed waste layer, ²⁾ Re-disposed waste layer after residual coal extraction; ³⁾ According to the Decree of Ministry of the Environment of 23 July 2008 on the criteria and methods of assessments of groundwater status (DME, 2008a)





Fig. 2. Hydrogeochemical profiles of pore solutions in the undisturbed waste layers at the Buków and Smolnica coal mining waste dumps

The long-term monitoring data showed that pore solutions and groundwater downgradient of the Buków dump remained close to neutral (pH 6.38–8.09), while at the Smolnica site acidic leachate actually occurred 8 years after the beginning of waste disposal, and pH of waters downgradient of the dump was permanently acidic (pH 2.49–7.58). The Dębieńsko dump also appeared to generate ARD (pH 4.38–7.42). The chemical composition of pore solution in the vertical profiles of the undisturbed waste layer of the dumps shows characteristic increased downward redistribution of macrocomponents (Fig. 2). High salinity, mostly sulfate, of both neutral and acidic leachate makes groundwater downgradient of the dump unfit for any use, and from this standpoint the pollution potential of both NRD and ARD is similarly high (Fig. 3). Leachate and receiving groundwater are usually enriched in Mn up to 3.05 mg/l (Buków), 15.98 mg/l (Dębieńsko) and 17.54 mg/l (Smolnica) and under anoxic conditions also in Fe, up to 35.5 mg/l (Buków), 165 mg/l (Dębieńsko) and 310 mg/l (Smolnica).





upgradient

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Fig. 3. Cumulative frequency diagrams

Chemical analyses of real pore solutions at the lowest and mean pH values (Tab. 1) indicate that in acid solutions, besides Mn and Fe (under anoxic conditions), also Ni may be released in amounts resulting in relatively high concentrations. An occurrence of elevated contents of Cd, Cu and Zn can be also anticipated, although in concentrations close to Maximum Permissible Concentration Level (MCL), which can influence the receiving groundwater status only rarely.

WASTE RE-USE

Table 2

In the last decade, a considerable progress in the waste reuse occurred. In 1990-2006, the amounts of generated industrial waste decreased by 15% at the simultaneous increase of production potential. In 1990, 35% of mining waste was landfilled. In 2008 this percentage was reduced to 10.4%, while the rest of waste was re-used (GUS, 2009). Mining Waste Directive 2006/21/EC and implementing it Polish Law on the extractive wastes (2008) provide the principles of the environmentally safe extractive waste management. Waste reuse from existing dumps for residual coal extraction, and in the last decade also for construction aggregate production for engineering constructions have resulted in the disturbance of primary dump layers and re-exposure of admixed material to the atmospheric conditions. This activity requires thus a reliable long-term Environmental Impact Assessment (EIA) and evaluation of risk to the environment of the specific reuse options, e.g. for embankments and flood polders construction. Assessing transformations occurring in coal mining wastes during residual coal extraction, transport and re-deposition of the granular material at the re-use destination (e.g. railroad embankment, road base, ground leveling etc.) is of a particular importance in such cases (Stefaniak, Twardowska, 2005a, b).

RESIDUAL COAL EXTRACTION

Residual coal extraction by physical methods comprise re--mining of a disposed material, its transport, multiple sieving, separation in heavy liquids and re-disposal. An essential role in these processes plays water used for transportation, washing and separation. The water- to-waste ratio in this process accounts for 1:1, and the total exchange of circulating water occurs every 10 days (Stefaniak, Twardowska, 2005a, b). Re-disposed wastes have higher water content, exposed specific surface and sulfide content than the primary rock material. This results in significant changes in generation and migration of contaminants. In general, in comparison to the primary undisturbed waste layers, discharge of leachate from the re-mined dump and leached contaminant loads increase (Tab. 2). Sulfate and chloride concentrations in the groundwater stream downgradient of the dump show an upward trend (Fig. 3), while the pattern of hydrogeochechemical profiles of pore

Dump, profile			Mean concentration	Water discharge	Mean load
			[mg/l]	[m ³ /year]	[t/year]
Undisturbed wastes		pН	7.69-8.41		_
	Buków (1997)	TDS	4596	160 120	777
		sulfates	2616	109 120	442
		chlorides	107.2		18
	Smolnica (2001)	pН	6.44–7.46		_
		TDS	10840	129.250	1391
		sulfates	7212	128 350	926
		chlorides	502.8		65
	Dębieńsko (2004)	pН	4.54-7.50		_
		TDS	2921	225.020	689
		sulfates	2061	235 930	486
		chlorides	11.0		3
	Smolnica (2004)	pН	5.69-7.25		-
		TDS	1743		224
		sulfates	1222	128 350	157
		chlorides	11.9		2
Re-disposed wastes	Buków (2001)	pН	6.99–7.84		_
		TDS	6932	225.000	1629
		sulfates	4447	235 000	1045
		chlorides	823.5		194
	Smolnica (2001)	pН	6.24–7.52		_
		TDS	5892	150.250	1051
		sulfates	4400	1/8 350	785
		chlorides	392.9		70
	Dębieńsko (2004)	pН	6.51-7.48		_
		TDS	5286	227.050	1733
		sulfates	3672	327 850	1204
		chlorides	70.7		23
	Smolnica (2004)	pН	6.94–7.38		_
		TDS	3089		551
		sulfates	2122	178 350	379
		chlorides	12.5		2

Contaminant loads leached from undisturbed and re-disposed waste rock at the coal ming waste dumps



Fig. 4. Hydrogeochemical profiles of pore solutions in the re-disposed waste layers at the Buków and Smolnica coal mining waste dumps

solutions in a re-disposed waste layer appears to be similar to a freshly disposed material and shows the highest concentrations at the top of a profile (Fig. 4). This has been defined as a "waste re-activation", when the leaching process begins anew.

WASTE RE-USE FOR ENGINEERING CONSTRUCTIONS

Coal extraction from coal mining waste provides coal and the construction aggregate for engineering constructions. Chemical composition of groundwater in the vicinity of an embankment of the Buków flood polder constructed from about 120,000 m³ of secondary waste obtained after coal extraction, showed almost instant strong adverse environmental impact of this embankment and a change of groundwater quality from a primarily good into poor chemical status. In particular, sulfide oxidation and sulfate generation, along with "re-activation" of chloride leaching resulted in concentrations many times exceeding MCL (Fig. 5). As monitoring data show (Stefaniak, Twardowska, 2005a), also K, Na, Fe and Mn concentrations in groundwater exceeded MCL (DME, 2008a) at pH values within the MCL range.

The reuse of "raw" coal mining waste for ground leveling (Fig. 6) also contributes to high sulfate salinity of pore solutions. A gradual acidification of the rock material, along with elevated concentrations of SO_4 (580–2766 mg/l),



Fig. 5. Temporal alterations of pH, sulfate and chloride concentrations in groundwater in the vicinity of the polder embankment constructed of re-used coal mining wastes

TDS (925–4132 mg/l), K (22–79 mg/l), Ca (145–416 mg/l), Mg (77–240 mg/l) and in acid solution also Ni (up to 0.08 mg/l) were observed.

The reuse of coal mining waste as a structural fill for liquidation of waterlogged quarries is considered one of the recommended environmentally safe methods of sulfidic waste management due to insulation of sulfide from contact with oxygen, provided the sulfidic material is permanently covered with water (BREF, 2009). In such case, a temporary decrease of the groundwater quality can though be anticipated at the time of filling a quarry with waste, resulted from a release of soluble salts occurring in the material and their further migration in groundwater. This can be illustrated by the temporal adverse transformations of groundwater quality in the waterlogged gravel quarry ZB-33 in the Buków area filled with the re-used non-ARD generating wastes and in another quarry ZB-32 hydraulically connected with ZB-33 and located downgradient of ZB-33 (Fig. 7).

In this case, groundwater deterioration lasted for 4 years, until a thorough exchange of water in the filled quarry occurred, which was followed with a gradual improvement of water quality in ZB-33 and with a similar time-delayed process in ZB-32. The decline of water level in ZB-33 in 2009 below the waste surface caused visible deterioration of water quality in both quarries. This example shows that in the case of underwater disposal of sulfidic waste, a phase of contaminants release already occurs in the material and the need of water level control should be always taken into account.





Fig. 6. Hydrogeochemical profiles of pore solutions in the coal mining waste used for ground leveling



Fig. 7. Temporal changes of SO_4 and Cl concentrations in waterlogged gravel quarries ZB 33 – quarry filled with the re-used coal mining wastes; ZB 32 – disused quarry located downgradient of the groundwater flow from ZB 33

CONCLUSIONS

1. In coal mining waste management practice, the major source of a groundwater deterioration appears to be the high sulfate salinity accompanied with high Mn and (under anoxic conditions) also high Fe release, independently on the susceptibility of a material to acidification. Therefore, groundwater protection measures should be undertaken both with respect to ARD and NRD generating waste, in case of their disposal and bulk reuse in engineering constructions.

2. Bulk coal mining waste re-use for residual coal extraction and as a common fill in engineering constructions results in the disturbance of primary formed hydrogeochemical conditions in re-deposited wastes. Due to the increase of the exposed surface, and application of the additional portions of water in the residual coal extraction process, an intensification of contaminant generation and leaching both: at waste re-disposal after coal extraction, and at its re-use for engineering constructions, has been observed.

3. An underwater disposal of coal mining wastes, although considered environmentally safe, causes temporary adverse impact on the groundwater chemical status pertain to release of contaminant loads occurring in the material at the moment of waterlogging. The water level decline below the waste surface also should be strictly controlled to avoid groundwater deterioration.

4. The presented examples confirm the need for efficient "cradle-to-grave" protection measures in any case of sulfidic coal mining waste disposal and re-use.

Acknowledgements. This work was partly supported by statutory funds of the AGH University of Science and Technology (project no 11.11.140.139).

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