Distribution and environmental impact of coal-mining wastes in Upper Silesia, Poland

J. Szczepanska · I. Twardowska

Abstract About 50 million tonnes/year of waste rock from coal-mining is generated in the limited area of the thickly populated Upper Silesian Coal Basin (USCB) in Poland. There are 380 coal-mining waste dumps, including 76 active dump sites covering over 2,000 ha. About 15-16 million tonnes/year of waste rock is being reused for civil engineering purposes in the same area. This brings about a problem of ground water deterioration by constituents leached from waste rock exposed to atmospheric conditions. The major factors determining the ground water contamination potential from waste rock are chloride salinity, sulfur content and acid generation potential. The concept behind the presented studies was to provide data for correct evaluation and prediction of contaminant release from the waste rock, based on the characterization of coal-mining waste properties, as well as on longterm laboratory, lysimetric and field studies. The results show that coal-mining waste dumps can be a long-term source of ground water contamination, lasting for decades and increasing with time. Ground water down-gradient from the disused 15–30-years-old part of the studied dump displays high and increasing acidification, high TDS, SO_4 , and the highest, still increasing concentrations of Mn, Fe and Zn. Cost-effective and efficient pollution control measures, similar to the presented design and construction elements of the dump site, can mitigate the negative environmental impacts.

Key words Coal mining waste · Pore solutions · Leachate · Ground-water pollution

Received: 3 July 1997 · Accepted: 9 September 1998

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Introduction

Waste rock from hard coal mines is the predominant group of solid waste lying in dumps and generated annually in Poland. In 1995, 53.7 million tonnes of these wastes were produced while 712.5 million tonnes were already stored in dumps. With respect to the total amount of waste generated and disposed of in Poland it accounts for 43.7% and 36.2%, respectively (Chief Statistical Office 1996). Almost all production of hard coal-mining waste (50.2 million tonnes/year) is concentrated in the Upper Silesian Coal Basin (USCB) in southern Poland, an area that comprises 1% of Poland (5 600 km²) and includes 7.9% (3.1 million) of its population (Fig.1). The annual amount of waste rock generated reflects general trends in the coal market and has been stabilized at the present level of about 50 million tonnes since 1985. Of this amount, 36–40% (about 20 million tonnes/year), has been reused for various purposes since 1990, including backfilling of mine workings (4–5 million tonnes/year) and in civil engineering uses (15–16 million tonnes/year) such as leveling/reclamation of areas impacted by subsidence, and fill for construction of railway beds, river and pond embankments etc. In most cases, the mine waste material used for civil engineering purposes is being exposed to atmospheric precipitation and air to a similar or even greater extent than at mine waste dumps. The distribution of coal-mining dumping sites in the USCB reflects the location of mines and mine-fields and is particularly dense in the southwest area of the Rybnik mining district and in the central part of the USCB (Fig. 2). There are 380 coal-mining waste dumps, including 76 active dumps covering 2166 ha in the Katowice district (State Inspectorate of the Environment Protection 1995). Up to the late seventies, the self-ignition of coal-mining waste dumps was a primary concern, and potential pollution of ground water was not considered important. As a result, ground water protection was rarely taken into consideration when waste rock dumps were sited. Currently, the pollution potential with respect to ground waters is of growing concern. This concern is greater where waste rock reuse for civil engineering purposes causes an increase in the exposed area of waste rock and a resulting intensification of weathering processes and generation of increased pollutant loads. The amount and nature of the released contaminants depends on the characteristics of the extracted and disposed waste rock material.

Fig. 2

Distribution of coal-mining waste dumps in the Katowice district (USCB area; State Inspectorate of the Environmental Protection 1995)

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Characteristics of coal-mining wastes

Waste material is discharged from coal mines in the form of run-of-mine rock (1–500 mm), usually dry, and from coal preparation plants as discard from heavy liquid separators (30–250 mm), jigs (0.5–30 mm) and tailings from flotation $(< 1$ mm). The environmental impact of these coal-mining wastes is determined by the characteristics of the extracted rock material, which can vary significantly, both horizontally and vertically through the coal-bearing Carboniferous strata.

The coal-bearing Carboniferous sequence in the USCB incorporates the complete Upper Carboniferous section, including Namurian (A,B,C), Westphalian (A,B,C,D) and Stephanian ages (Stopa 1967). In a vertical succession, the lower marine-brackish shale-sandstone rocks (Namurian A seams 600-900) grade upward into the upper continental limnic strata. Overlaying Namurian B,C seams correspond to the Upper Silesian sandstone formation (seams 503-510 and 408-500). Upward Westphalian A,B (seams 328-407/4 and 302-327) belong to the Upper Silesian shale formation. The upper portion of the mined Westphalian C,D (seams 208-302, 118-207 and 100-117) represents Cracow sandstone formation.

Freshly wrought waste material consists of argillaceous and arenaceous rocks in the form of siltstone and mudstone with a variable amount of sandstone, depending on the mined coal seam. Predominant mineralogical components of wastes are therefore silicates and aluminosilicates, but clay minerals, mainly non-swelling (kaolinite, illite, chlorite and mixed-book structures of illite/chlorite type) are also important because of their ion-exchange capacities. The constituents of concern with respect to the leaching properties and contamination potential, occur in the waste rock in the accessory amounts and include chloride, iron sulfide (pyrite, marcasite FeS₂), buffering minerals including calcite $(CaCO₃)$, dolomite [CaMg(CO₃)₂] and ankerite [Ca(Mg,Fe)(CO₃)₂]. A number of heavy metals are present in trace concentrations, mainly in the form of sulfides, such as sphalerite (ZnS), galena (PbS), millerite (NiS), covellite (CuS) and chalcopyrite and can be released in the process of sulfide oxidation.

Chloride salinity in Carboniferous rocks has been found to be associated predominantly with pore water and therefore chloride ions are highly mobile. Chloride salinity varies in a wide range, from 10^{-3} to $10^{-1}\%$. It depends upon chloride concentrations in ambient ground water and rock porosity, being a consequence of filling the pore spaces with underground saline waters (Herzig and others 1986). In the USCB, these parameters show distinct spatial and vertical zonality, which facilitates predicting the chloride salinity of freshly wrought coal-mining waste rock. Spatially, two hydrogeological regions of a different mode of ground water recharge are developed, depending on the distribution of a sealing horizon formed by Tertiary clay sediments that insulate the Carboniferous strata

Fig. 3

Spatial zonality of the total dissolved solids (TDS) in mine waters 500 m below sea level in the Upper Silesian Coal Basin (USCB; Różkowski 1995). *1* extension of the USCB; 2 coalfields; *3* hydrogeological regions; *4* sealing horizons of Tertiary clay sediments; 5 Tertiary salt deposits; 6 isopleths of TDS (g/dm³)

from the percolation of atmospheric precipitation (Fig. 3). In both hydrogeological regions, three vertical hydrochemical zones can be distinguished including (1) infiltration, (2) mixed, and (3) connate waters. This regionality and zonality determines the distribution of underground waters of different chloride salinity and total dissolved solids (TDS) ranging from 0.1 to 230 g/dm³ (Pluta and Zuber 1995; Różkowski 1995). A simple method of estimating chloride concentration in rocks based on the data on chloride salinity of mine water and rock porosity has been presented elsewhere (Herzig and others 1986). Iron disulfide is the main component that gives rise to the permanent environmental problems related to the coal-mining waste disposal. The major sulfur-bearing material in the Carboniferous strata is coal. Sulfur contents in the USCB coals are relatively low. Mean total sulfur concentration (S_t) is slightly above 1%, but in two-thirds of the mined and penetrated seams, the mean S_t concentration is $\langle 1\% \rangle$. In general, mean sulfur concentrations in coal seams of the USCB increase spatially from SW to NE, and are the highest in the Westphalian D seams of the eastern coalfields of the USCB (Fig. 4). Sulfur in coal seams of the USCB occurs predominantly in the inorganic form (mainly pyrite $FeS₂$, which contributes about $85-95\%$ S_t). Sulfur content in the waste material reflects the relevant concentrations in the mined coal seams, and also depends upon coal contents in rock and on the coal preparation process (lesser contents occur in coarse material, higher in fine tailings). The maximum mean S_t concentrations range from 0.7% in run-of-mine waste, to 3.0% in washery discards, and 5.6% in tailings, the average mean value is generally around 1% S_t.

Besides the total concentration, the reactivity of pyrite, that is the kinetics of oxidation/hydrolysis reaction in the material exposed to atmospheric air and humidity, with formation of Fe(OH)₂, sulfate SO₄⁻ and acidity (H₃O⁺) ions), is the controlling parameter which determines the pollution potential of waste. Complex biochemical reactions describing the process are presented in a more or less simplified form in numerous publications (Caruccio 1978; Kleinmann and others 1981; Hutchison and Ellison 1992; Shirav and Robol 1993; Jarvis and Braithwaite 1994). Pyritic sulfide oxidation is generally considered to be a multi-staged process comprising four basic reactions proceeding either abiotically or by direct bacterial oxidation by *T*.*ferrooxidans*. The reaction stages are controlled by chemical conditions (pH range, sulfate and iron concentrations, acidity, Fe^{3+}/Fe^{2+} ratio; Kleinmann and others 1981). In coal-mining waste dumps of the USCB, the process of bacterial pyritic sulfide oxidation has been found to be highly controlled by the buffering capacity of the material. In the buffered waste, sulfide oxidation proceeds mainly abiotically as a one-stage process. In the low-buffered material, the role of the direct bacterial oxidation increases significantly as pH falls and process proceeds generally as a two-stage one (Twardowska 1986, 1987).

Pyrite reactivity is defined as a half-period of decomposition $t_{1/2}$ (days), while the process of pyrite decomposition as a function of time can be described as a first-order reaction:

$$
(G_s)_t = (G_s)_0 \cdot e^{-kt} \tag{1}
$$

where:

$$
k = \frac{\ln 2}{t_{1/2}}
$$

 (G_s) ⁰-initial concentration of pyritic sulfur S_p in the fresh-wrought material,

 $(G_s)_t$ -concentration of pyritic sulfur S_p in a time period *t*. It was first shown by Caruccio (1972), that the type of iron disulfide determining its reactivity, and not the amount was a critical factor in the production of sulfate and acidity. This conclusion was also reached by the studies on the Polish coal seams, though no direct correlation between pyrite structure, texture, mineralogical mode and the process kinetics was found (Twardowska and others 1988; Witczak and Postawa 1993). The pyrite half-period in the USCB coal seams varies from 29 to 10502 days, with a mean $t_{1/2}$ =588 days and shows a distinct relation to the seam location in the Carboniferous strata; the shortest half-period for the material from seams 100–200 (Westphalian C,D) and 700–800 (Namurian A,B), the longest for material from seams 300–400 (Namurian D; Westphalian A,B) (Twardowska and others 1988). In general, pyrite decomposition is the key process determining the capacity of waste rock to generate acidity, while the buffering properties are attributed to Ca and Mg-carbonates (calcite, dolomite) and the cation exchange capacity of clay minerals. Occurrence of carbonates along the stratigraphic column of the USCB and their forms show considerable diversity. Mean content of carbonates in run-of-mine waste ranges from 0.31 to 1.64%, whereas spoil from coal preparation plants have higher amounts (up to 4.60% in coarser waste 20-200 mm and up to 11.0% in flotation fines). At the upper part of the stratigraphic section (Westphalian C,D), Ca-Mg carbonates are present mainly as a cement in clastic rocks. In Westphalian A,B and Namurian B,C strata these minerals most commonly form concretions, and minor amounts form cement or dispersed grains. This causes substantial variability of buffering capacity of waste rock from these seams. The highest carbonate enrichment is in waste rock from Namurian A seams. Total ion exchange capacity of the Carboniferous rocks of the USCB, typically associated with kaolinite, ranges from 29 to 123 eq/Mg. The vulnerability of the rock material of the USCB to acidification was found to be well defined by the equivalent ratio of buffering constituents ($\mathrm{G}_{\mathrm{buf}}$) to pyritic sulfur concentration (G_{ac}) :

$$
\xi = \frac{G_{\text{buf}}}{G_{\text{ac}}}
$$
\n(2)

If ratio $\xi \ge 2.4$, system is well buffered, at $\xi \le 0.6$ acidification will occur, at $2.4 \ge \xi > 0.6$, the system is unstable (Twardowska 1989)

Permeability of the material to air and water is another critical factor with respect to the pollution potential to ground water. The permeability to water of the freshly generated material in the compacted state is generally high and ranges from 10^{-4} to 10^{-5} m/s, while the permeability of the compacted weathered material is of 1-2 orders of magnitude lower (from 10^{-5} to 10^{-7} m/s). Coarse,

essentially granular material from the deeper coal seams (300-800) is resistant to compaction and weathering. It remains permeable or semi-permeable to water and air even after a long period of time and a heavy compaction, while the waste from the upper parts of the Carboniferous strata (coal seams 100-200) shows good compaction and self-compaction properties.

Ground water pollution potential

Long-term laboratory, lysimetric and field studies define several general factors controlling the degree of potential hazard to the ground water under the vadose zone conditions, where percolation of precipitation water through the dump occurs (Twardowska and Szczepanska 1990). These factors include: (1) the vertical redistribution of chloride and sulfate loads in solutions infiltrating through the dump body (vadose zone), particularly important where equilibria limitations are minimal (e.g. chlorides and sulfate ions equilibrated by Na⁺ or H₃O⁺), (2) dynamics and kinetics of sulfate and acidity genera-

Table 1

Chemical composition of leachate from the upper low-buffered waste layers. Long-term lysimetric studies: lysimeter SM-1: buffered 8-year-old waste; lysimeter SM-2: non-buffered

tion (FeS₂ content, its reactivity, and the availability of air and water), (3) buffering capacity of the rock material (the amount of Ca- and Mg-carbonates and their proximity to the centers of the acidity generation, and (4) the pore-water exchange rate in the waste layer. The ground water pollution potentioal of low-buffered waste is illustrated by the chemical composition of leachate from a waste layer 1.8-m thick, 8 and 18 years old (Table 1, Fig. 5). Freshly generated coal-mining waste is buffered, and alkaline or close to neutral material. Major ions released from the waste matrix to infiltrating water from atmospheric precipitation are chlorides and sulfates balanced predominantly by Na⁺ cations. After fast decrease of mobile species (usually, after a single pore-water exchange in the waste layer), the chemical composition of leachate stabilizes at a level controlled by the dynamics of sulfide decomposition and the carbonate and gypsum equilibria in the system Ca-Mg-C-O-H-S. Compounds that may cause deterioration of ground water quality are total dissolved solids (TDS), mainly sulfate ions balanced by Ca^{2+} and Mg^{2+} , while trace metals are bound in the rock matrix within the pH-Eh stability field (lysimeter SM-1). Gradual depletion of Ca-Mg-carbonates

18-year-old waste; waste layer 1.5 m thick; waste from the Smolnica coal-mining waste dump, Rybnik region of the USCB; ~below the detection limit DL

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Fig. 5

pH, SO_4^{2-} , Zn, and Al concentrations in leachate from the lysimeter SM1 (buffered waste, 7–12-years-old) and SM2 (non-buffered waste, 15–20-years-old). Smolnica coal-mining waste dump, Rybnik region of the USCB, long-term lysimetric studies

results in acidification of low-buffered waste material and mobilization of the trace species from the matrices of wastes and other materials in contact with the acidic leachate. Major compounds released from 18-year-old waste in amounts many times exceeding maximum permissible concentration level (MCL) are TDS, SO_4^{2+} , Al, Zn, Mn and Fe ions which precipitate abundantly as $Fe(OH)_{3}$ in oxic conditions. As a result of H_3O^+ ions reaction with aluminosilicates in rock matrix, Ca_{2+} and SiO_2 ions are released into solution and alumino-rich clay minerals are produced. Other trace elements also occur in elevated concentrations compared to the buffered system (lysimeter SM-2). This results in a substantial increase of the pollution potential of low-buffered waste with time (Table 1, Fig. 5).

Infiltrating water exchange rate in the dump is of particular importance to assessing the dynamics of dissolved solids removal. In the vadose zone conditions, the liquid to solid (L/S) ratio is low and a single exchange of pore water in the waste layer 20-m thick will take at least 5 years. This determines a long-term impact on the ground water for even mobile constituents such as chloride. In the surface layers, chloride is almost depleted, but it is increased in the leachate at the bottom of the dump. The similar pattern of a vertical redistribution is displayed by

sulfates in the pore water percolating through the lowbuffered waste layer (Fig. 6). In the pore water of buffered material, sulfate concentrations are limited by the equlibria constraints with CaSO₄. Besides macrocomponents such as chloride, sulfates, total dissolved solids (TDS), also elevated amounts of trace elements may be leached out from unbuffered wastes. Trace elements may be released from acidic waste, but also from the vadose zone matrix underlying the dump, or from other materials such as concrete or metal constructions contacting with the acidic leachate (Table 1, Fig. 5). Data from 3-year local ground water monitoring in the vicinity of the Smolnica coal-mining waste dump, active as a dump site since 1965, show significant ground water quality changes down-gradient from various parts of the dump (Table 2). These changes are attributed to variation of waste material properties resulting from weathering and the addition of fresh-wrought waste through time. The results show that coal-mining waste dumps can be a

long-term source of the ground-water contamination, lasting for decades. For example, after 30 years of operation of the Smolnica dump, increases of TDS in ground water by 1-2 orders of magnitude compared to background concentrations are observed. Disposal of fresh wrought waste causes high contents of \overline{Cl} , SO_4 , Na, Mg, Fe and COD, while ground water down-gradient from the disused 15–30-year-old part of the dump displays high and increasing acidification, high TDS, SO_4 , and the highest, still increasing values of Mn, Fe and Zn. The deep deterioration of ground water quality, both with respect to MCL and background concentrations of ground water up-gradient of the dump, disqualifies these waters for any

Table 2

Groundwater quality up-gradient (from 30 to 650 m) and down-gradient (from 30 to 300 m) of the Smolnica coal mining GQM network: 1994–1997

waste dump. Dump location: Rybnik region of the USCB. Data from the local groundwater monitoring sampling of the local

^a MCL maximum concentration limits for drinking water and for water of I quality class (in brackets)

use. Similar adverse changes of the ground water resources in the area impacted by leachate from dumping sites are being observed throughout the USCB area. As a result of renewable ground water resources deterioration, in many communities local wells have been abandoned and tap water is supplied from the remote sources.

Ground water pollution control measures

Currently, environmental protection requirements and liabilities with respect to dump siting and management are regulated in Poland by several consecutively actualized legislative acts. These acts and directives place strict liabilities on the responsible parties associated with the contamination due to active or inactive dump sites and provide responsible agencies with authority to execute compliance with regulations. The institutions responsible for legislative acts enforcement are State Inspectorate of

the Environmental Protection and its District Inspectorates, as well as Ecological Departments of the Regional and District Administrative Boards. Their inspection and evaluation activities are supported by the certified laboratories for the environmental monitoring, research and control, as well as by the licensed corporate and individual experts of the Ministry of the Environmental Protection, Natural Resources and Forestry.

Due to the "polluter pays" principle enacted in the legislative framework for existing waste dumps, an obligation to provide an Environmental Impact Assessment for the newly designed dumping sites, and the growing environmental awareness of the local communities, an increasing effort is being applied to develop environmentally safe coal-mining dump construction, as well as to mitigate the adverse impact of existing dumps on ground water. Considering size and the long period of the coal-mining dump use, the protective measures need to be efficient, cost-effective and appropriate for both operational stage and the post-closure periods. For this purpose, the technical measures for dumping site construction, based on the coal-mining waste environmental behavior, have been

Fig. 6 Hydrogeochemical profile of Smolnica coal-mining waste dump. Vadose zone, waste layer 10–15-years-old

elaborated (Twardowska and others 1988). The major concept for the abatement of released contaminants consists in the reduction of, (1) infiltration of precipitation water into the waste dump, (2) amount of leached waste, (3) generation of contaminants in the waste layer and (4)

rates of contaminant release from the leached layer. These aims are being attained by the horizontal and vertical air-tight and water-tight dump designing and limitation of its exposed surface by means of an adequate construction and shaping.

The main design and construction elements comprise: (1) outgoing construction of the external banks in heavily compacted thin layers preceding the gradual filling and compaction of the internal dump body, (2) maximum limitation of the exposed area to volume ratio, (3) application as a sealing material of appropriate fine-grained wastes of a low permeability and a high water retention capacity (tailings, low-ratio fly-ash/water mixtures etc.), (4) application of the surface drainage constructed on the low-permeable base, and a double system of the toe ditches, to collect and discharge separately leachate and clean run-off waters from the external area (Fig. 7). These solutions, focused on the control of air and water permeability control, also effectively prevent self-ignition. The present design was used successfully for the first in the one of two largest central coal-mining dump sites Koscielniok in Rybnik. There are currently several active dump sites using this design and all new ones are designed this way.

Another regulation requirement for each dump sited in areas of ground water reservoirs or of usable ground water horizons is the monitoring of ground and surface wa-

Fig. 7

General scheme of the coal-mining dump drainage system. *1* acid-resistant drains; *2* collectors; *3* drainage base layer of low permeability; *4* coal-mining waste layer; 5 pipeline; *6* internal ditch; *7* retention/dosage reservoir; *8* pump station; *9* external ditch; *10* recipient of run-off from the external area; *11* monitoring wells

ter quality as an integral part of the dump reclamation. Currently, not many active waste dumps are provided with properly designed, operated and maintained monitoring networks, the Smolnica waste dump in the Rybnik district (W) of the USCB is one that is. There is still substantial inconsistency between the enacted legislative regulations and the general understanding of the environmental behavior of the disposed waste. From this pespective, legislation requiring restoration of the initial landscape and use mode of the disposal site seems particularly unfit. Optimization of land use and attenuation of environmental impact in the most cases dictate fundamental changes of the landscape and the utilization of the site. Also traditionally practiced post-closure biological reclamation by planting trees at close spacing in order to obtain rapid vegetative cover commonly causes severe losses, high costs and incompatibility with the ground-water protection measures due to the deep penetration of a root system. Landscape architecture with a wider use of the combination of herbaceous cover and higher vegetation in clusters as decorative elements can be much more cost-effective, efficient and will not adversely affect the dump surface drainage system. Evapotranspiration processes in properly designed, fast-growing carpet-like vegetation cover can also play a significant supporting role in the control of water infiltration through the dump.

Due to the long term of dump use, special ground water protective measures should be undertaken during the operational period. In particular, dump construction in consecutive sections and their immediate post-closure capping should be considered in large central dumping sites. These few examples show that there is still much is to be done for optimization of the ground water protection measures along with other aspects of restoration and conservation in the complex system that characterizing waste rock dump sites.

Conclusions

- 1. Coal mining waste dumps should be considered as a serious long-term source of ground water deterioration. Pollution potential of low-buffered waste increases with time, as depletion of buffering capacity and acidification of waste occurs as a result of iron disulfide decomposition. Occurrence of a regional zonality of Carboniferous rock properties in the USCB allows prediction of the environmental behavior of the waste material deposited at dumps with a considerable accuracy.
- 2. Coal mining dump construction can be treated as a complex system directed to mitigate adverse environmental impacts.
- 3. Despite substantial advance in designing, construction and operation of coal-mining waste dumps with regard of ground water protection in the last decade, the present situation in Poland is still far from being satis-

factory. The lack of a comprehensive approach to the environmental protection priorities in the design and construction of the coal-mining waste dumps, their incomplete reclamation and then inappropriate use is still too common.

4. Cost-effective and efficient dumping site construction needs to be based on the waste rock long-term environmental behavior in operational stage and in the post-closure period. The environmentally oriented optimization of a dump site design, which includes technical, biological, and landscape planning details, and considers appropriate prospective use of the site, can successfully mitigate the adverse environmental impacts and prevent degradation of recoverable ground water resources.

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