Petrological and Environmental Geochemical Studies on the Abandoned Maghara Coal Mine

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ABSTRACT: Samples from outcrops of Jurassic coal seam in Maghara coal mine have been analyzed for their proximates, ultimates, ash constituents and trace elements. Microscopic observations have revealed that the coal is mainly composed of vitrinite, liptinite and inertinite macerals. Vitrain plays an important role in the outbreaks of spontaneous combustion because it absorbs oxygen more than other coal components. Calorific values were 4,763 cal/g and 7,422 cal/g for weathered and fresh coal samples, respectively, which indicate extensive weathering processes in the mine. The abandoned coal mine may be caused several environmental impacts such as high concentrations of Fe and SO₄²⁻, liberated from oxidation of pyrite, high salinity contents and some toxic trace elements in waters. The mechanisms of the mass balance of some heavy metals indicates that the highest accumulations were 15.36, 7.74, 6.79, 1.69, 1.39 and 0.122 kg⁻¹ha⁻¹yr⁻¹ due to Ni, Cr, Pb, Fe, Co and Cd, respectively. The most dangerous process of environmental impacts in the studied area is that groundwater in Maghara area is unsuitable for drinking, domestic and irrigation purposes.

KEY WORDS: Maghara coal mine, pyrite oxidation, heavy metals and mass balance.

Introduction

The abandoned coal mine may cause the release of harmful trace element to the environment (Swaine 2000). The concentration and distribution of trace elements in coals have been documented in numerous articles, especially those by Bouska (1981), Swaine (1990) and Liu et al. (2001).

Coal weathering is the process of natural oxidation of coal as soon as coal comes into contact with O_2 in air. Many authors (Jones and Townend 1949, Schmal 1989, Berkowitz 1979, Clemens and Matheson 1996) agreed that a minimum amount of water (about 1% of coal mass) is necessary for the interaction between coal and O_2 . This may be due to the dual role of inherent water which acts as a catalyst and reactant in the oxidation reactions (Jones and Townend 1949, Berkowitz 1979, Petit 1991).

Petrographical components of coal play an important role in the oxidation process, where vitrain absorbs oxygen more than other coal components (Ferrari 1938, Kukharenko et al. 1959). Thus, oxidation of sulphide minerals may result in the discharge of large quantities of SO_4^{2-} together with metal ions into adjacent drainage systems (Blowes et al. 1991, Fuge et al. 1993) that can constitute a serious threat to the aquatic organisms (e.g., Andre's et al. 2000, Baudrimont et al. 2003).

Results of input-output budgets in small hydrological catchments yield useful data on the metabolism of ecosystems (Pačes 1985). Melegy (2005) reported that inputs and outputs link the ecosystem with its surrounding and the sum of all the fluxes composes a budget of mass balance in the system.

The major target of this paper is to provide some petrologic data on coal macerals types and influence of pyrite oxidation in Maghara coal mine on geochemical mass balance of trace elements.

The study area

Maghara region is located nearly in the center of North Sinai between longitude 33°10' and 33°40' E, and latitude 30°35' and

30°50′ N, covering an area of about 1,300 km² (Fig. 1). The region forms about 54 km long and 30 km wide asymmetric anticline with a curved axis trending about 55° NE and thus it is parallel to the Syrian Arcs.

Many authors studied geology of the region (Said 1962, Jenkins 1990, Moustafa and Khalil 1990). Al-Far (1966) described region in detail and divided the Jurassic section into six formations (Fig. 2), from the base to the top: Mashabba Formation, Rajabiah Formation, Shusha Formation, Bir Maghara Formation, Safa Formation and Masajid Formation.

Safa Formation is the more important continental unit in the section because it contains the economic coal bed. Safa Formation contains more than 11 coal seams but only two of them are minable – the Main Coal Seam (MCS) and Upper Coal Seam (UCS). The thickness of the MCS in the Maghara area ranges from 130 to 190 cm (Fig. 3) and the geologic coal reserve is estimated to be 52Mt based on the drilling data of more than 50 exploration wells (Mostafa and Younes 2001). These coals originated in lakes or lagoons adjacent to the coastline (Jenkins 1990).

Zodrow (2005) mentioned that as a result of ventilation process of mine breakdown, where air conditions are allowed to stagnate, remarkably narrow range of relative humidity and temperature exited. Natural oxidation of pyrite in coal took its natural course, forming hydrated sulfate minerals. Figure 4 shows the fall down of coal weathering products on mine floor. Accordingly, this may lead to the deterioration of groundwater quality.

Experimental techniques

Petrographic analyses were determined by the staff of Institute of Rock Structure and Mechanics of the Academy of Sciences of the Czech Republic. The reflectance and maceral composition of coal samples were determined from particulate polished blocks using NIKON microscope with Image analysis system





Fig. 1. Location and geological map of the Maghara coal mine in Egypt. Modified after Al Far (1966).





- Fig. 3. 150 cm thick coal seam above the base of sandstone in the Maghara coal mine (Bathonian age).
- Fig. 2. (a) Stratigraphic column of formations exposed at Maghara (Said 1962, Al-Far 1966, Jenkins 1990). (b) The sequence of strata in relation to the Main coal seam and Upper coal Seam (Well log C8/6).



Fig. 4. Natural weathering product of coal (grey colour) which falls down on mine floor.

LUCIA for random reflectance measurement (immersion objective with 20× magnification) and UMSP 30 Petro Opton-Zeiss microscope for reflected monochromatic light (λ = 546 nm), immersion objective with 25× and 40× magnification and oil immersion with refractive index of n = 1.518. Measurements of reflectance employed an optical standard (sapphire) with the reflectance of R_r = 0.585 %.

The mineralogical composition of ashes was determined by X-ray diffraction (XRD) techniques. XRD was carried out using Phillips X-ray diffractometer (Amsterdam Netherlands) with $CuK\alpha$ radiation and nickel filter.

Air-dried coal samples were oven-dried at 40 °C for 48 hours, then crushed and sieved with a 150-mesh screen. After that, powder were placed in a clean porcelain crucible and heated in a muffle furnace at 800 °C for 5 hours. About 0.5 g of the ashed sample was digested with 5 ml concentrated nitric acid, 10 ml hydrofluoric acid and 1 ml perchloric acid in the teflon beaker. The prepared samples were analyzed using inductivity coupled plasma (ICP).

Results

Results of this paper include some geochemical information on fresh coal (FC) and natural weathered coal (WC) in abandoned Maghara coal mine such as proximate and ultimate analyses, calorific values, forms of sulfur, major element concentrations, mineral matter contents and the relative proportions of coal macerals.

Proximate and ultimate analyses, calorific values and forms of sulfur are summarized in the Table 1. Total moisture contents are 2.65 wt. % for FC and 4.45 wt. % for WC. Volatile matter contents are 50.6 wt. % in FC and 39.36 wt. % in WC. Fixed carbon contents are 40 wt. % in FC and 29 wt. % in WC. Proximate results indicate that the coal samples have a low average ash content of 4.12 wt. % in FC and a high average ash content 22.48 wt. % in WC.

Calorific values were 4763 cal/g and 7422 cal/g for WC and FC samples respectively, placing most of the fresh coal samples in the high volatile bituminous coal. Coal loses 35.8 % of its calorific value (Table 1) as a result of intensive weathering process.

Demonsterne	Fresh coal sample	Weathered coal	
Parameters	(FC)	sample (WC)	
Proximate analysis			
Ash	4.12	22.48	
Moisture	2.65	4.45	
Volatile	50.6	39.36	
Fixed carbon	40.00	29.00	
Ultimate analysis			
Carbon	71.44	48.5	
Hydrogen	6.62	5.18	
Oxygen (by diff)	9.1	11.32	
Nitrogen	2.27	1.56	
Sulphur	3.8	6.15	
Forms of sulphur			
S (total)	5.77	8.82	
S (sulphate)	1.14	2.30	
S (pyrite)	3.80	6.4	
S (organic)	0.83	0.12	
Calorific value (cal/g)	7422	4763	
Petrographic analysis			
Vitrinite	91.6	41.4	
Liptinite	7.5 11.1		
Intenite	0.0	1.00	
Minerals	0.9	46.5	

Tab. 1. Proximate, ultimate and petrographic analyses of fresh and weathered coal in Maghara mine.

Maghara coals, in general, have high total sulfur values, ranging from 5.77 wt. % in FC to 8.82 wt. % in WC. Total sulfur is dominated by pyritic form. As discussed below, petrographic and X-ray diffraction analyses confirm high content of pyrite in coal.

Results of petrographic analyses (Table 2) illustrate the diverse character of coal. Concentrations of the vitrinite are predominant in Maghara beds. Vitrinite macerals range from 86.7 vol.% to 91.6 vol.% in FC and from 70.4 vol.% to 70.5 vol.% in WC. Concentrations of the liptinite macerals group range from 7.5 vol.% to 10.7 vol.% in FC and from 15.3 vol.% to 20.8 vol.% in WC. Inertinite macerals typically make up 0.6 vol.% in FC to a maximum of 2.8 vol.% in WC.

The mineralogy of most of the Maghara coal samples is dominated by quartz, sulfides and clay minerals as shown in the Table 2. Concentrations of trace elements in coal samples are summarized in the Table 3.

Discussion

Coal petrography and mineralogy

There are three basic groups of macerals (vitrinite, liptinite and inertinite groups) in the Maghara coal samples. The vitrinite group is the most abundant in the bituminous Maghara coals. Vitrinite components in all samples mostly consist of either collotelinite or colldetrinite and telinite. Collotelinite occurs as a structureless, homogeneous mass in thin or thick bands (Fig. 5A). Telinite occurs mostly as cell walls of variable sizes and shapes filled with other macerals or minerals (Fig. 5B).

Samples	Fresh coal (FC)		Weathered coal (WC)	
Parameters	1	2	3	4
Reflectance (%)	0.40	0.43	0.46	0.39
VITRINITE (vol. %)	91.6	86.7	70.4	70.5
Telovitrinite	72.9	64.0	55.4	15.8
Detrovitrinite	15.9	20.0	9.3	49.3
Gelovitrinite	1.9	2.7	5.6	5.4
LIPTINITE (vol.%)	7.5	10.7	20.8	15.3
Sporinite	0.9	1.3	3.0	4.0
Alginite	0.8	0.7	2.0	3.3
Cutinite	0.0	0.0	1.6	0.8
Resinite	2.8	3.3	5.3	2.7
Exsudatinite	0.0	0.0	0.4	2.0
Liptodetrinite	1.9	2.7	6.9	1.2
Bituminite	0.9	2.0	1.6	0.8
INERTINITE (vol. %)	0.0	0.6	2.8	1.8
Fusinite	0.0	0.1	0.4	0.5
Semifusinite	0.0	0.0	0.0	0.0
Macrinite	0.0	0.5	1.0	0.4
Funginite	0.0	0.0	0.0	0.0
Secretinite	0.0	0.0	0.7	0.2
Micrinite	0.0	0.0	0.5	0.3
Inertodetrinite	0.0	0.0	0.2	0.4
MINERALS (vol. %)	0.9	2.0	5.9	12.4
Clay minerals	0.1	1.3	0.8	0.7
Quartz	0.4	0.1	3.2	8.7
Sulphides	0.2	0.7	1.7	2.4
Carbonates	0.0	0.0	0.0	0.1
Others	0.2	0.0	0.2	0.5

Tab. 2. The results of microscopic study of Maghara coal samples.

Variables	FC	WC
Ash (525 °C)	4.12	22.48
Ba	79	202
Ni	0.8	0.6
Cu	370	150
Mn	3.91	3.71
Pb	540	1000
Zn	13	28
Cr	BDL	BDL
Cd	BDL	BDL

BDL: Below detection limit.

Tab. 3. Trace elements (dry, ash basis) for Maghara coal samples (All values in ppm).

Of the liptinite group, liptoderinite, sporinite and resinite are present in the greatest amounts, with smaller contents of alginite, cutinite, exsudatinite and bituminite.

Liptoderinite is the most dominant liptinite (1.9 %–6.9 %) (Fig. 6A). Resinite (2.7–5.3 vol. %) occurs as round to oval bodies. Sporinite occurs (0.9–4 vol. %) as elongated thread-like or spindle-shaped bodies (Fig. 6B). Cutinite (0–1.6 vol. %), dark grey to black, occurs as elongated thread like structures with one end serrated. Exsudatinite appears as brown films (Fig. 6B).



• Fig. 5. Photomicrographs of Maghara coal sample. A) Fresh coal sample showing structureless collotelinite (Cl), B) Coal sample showing telinite (Te) and resinite (Rs) macerals and pyrite (Py) minerals. All photomicrographs taken in white light in oil immersion.

The Maghara coal contains minor quantity of the inertinite macerals, it does not exceed 2.8 vol. % (Table 3). Macrinite (0–1 vol. %) occurs as amorphous (non-granular) and structureless white bodies with reflectance much higher than that of the collotelinite in the same sample (Fig. 7A). Fusinite shows well preserved yellowish white to white cell structure with a very high relief (Fig. 7B).

Vitrinite is characterized by a relatively high oxygen content compared with the macerals of the other groups (ICCP 2001).

The mineral matter content of Maghara coals (samples 1-5) is moderate (0.9–12.4 vol.%) (Table 3). The mineral matter components are predominantly quartz (0.1–8.7 vol.%) and sulphides (0.2–2.4 vol.%) with minor amounts of clay minerals (0.1–1.3 vol.%) (Table 2). The most common occurring sulphides in the Maghara coal is pyrite.

In the mineralogical composition of the Maghara coal ashes, determined by the XRD analysis, prevailed mainly hematite and



Fig. 6. Photomicrographs of Maghara coal sample. A) Sample showing vitrinite (V) and liptodetrinite (Ld) macerals, B) Sample showing exsudatinite (Exd) and sporinite (Sp) macerals and pyrite (Py) and quartz (Qz) minerals. All photomicrographs taken in white light in oil immersion.

magnetite (Fig. 8). The presence of hematite in the ashes of the Maghara coal is due mainly to the complete combustion of pyrite associated within it (Gaigher 1983). The residue after degradation of organic matter by H_2O_2 is mainly gypsum and natrojarosite (Fig. 9).

Geochemical mass balance of some trace elements

Maghara catchments comprise a considerable number of hydrographic basins in which some of these are combined together in one geomorphic unit. However, from the hydrologic point of view, each hydrographic basin is considered an individual basin



■ Fig. 7. Photomicrographs of Maghara coal sample. A) Sample showing macrinite (Ma), vitrinite (V) and inertodetrinite (Id) macerals and quartz (Qz) minerals. B) Sample showing fusinite (Fz) and cutinite (Ct) macerals and pyrite (Py). All photomicrographs taken in white light in oil immersion.

governed by specific drainage pattern, which is bounded by its catchment area. In the studied area many Bedouins people use groundwater for drinking and domestic purposes. Therefore the evaluation of geochemical mass balance of some toxic heavy metals is necessary in the Maghara catchment.

The elementary tool used for interpretation of measured data is calculation of the mass fluxes for individual elements, e.g. determination of the total amount of an element or compound entering or leaving catchment. The input-output budget represents a balance sheet approach to the evaluation of known fluxes. Both fluxes are defined relatively, with respect to the definition of the catchment used. Inputs are entered with a positive sign in the equation, outputs with a negative sign. For each element an equation can be written in the following general form:

Inputs – Outputs = Net (1) Mine discharge is expressed as $1.s^{1}$, it is the only phase of the hydrologic cycle for which reasonably accurate measurements of volume can be made. The mean quantity of input and output minewater discharge (Q) of the Maghara catchment is $1.66 \ 1.s^{1}$ and $0.3 \ 1.s^{1}$, respectively. Ratings of input-output flux were determined from theoretical calibrations (Rantz 1982).

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• Fig. 8. X-ray diffraction pattern of the studied coal ash showing characteristic hematite (H) and magnetite (M) peaks.

Most of acid generated during the oxidation of pyrite was neutralized (Younger 2001). Average value of minewater pH ranges from 7.8 to 8. The results obtained from the analysis of input-output water showed that they have some characteristics

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which differ markedly from those of natural waters. These characteristics are determined by the process of sulfide oxidation in the abandoned coal mine which affects, to a greater or less extent, all of the water analyzed, as is clear from high concentration of SO42- (between 2,425 and 2,549 mg.l1-). As well as liberation of SO_4^{2-} , the high salinity gives rise in the values of EC and TDS as shown in the Table 3. Concentrations of Ca, Mg and Na were especially high, the input values of minewater of these elements being 906, 435.32, 1,766 mg.l1-, respectively, whereas the output values being 902, 413, 1,686 mg.l¹⁻, respectively (Table 3). In general, input-output concentrations of minewater which leaching to the underground in the studied catchment water was much higher than those in natural waters.

The processes of pyrite oxidation in coal mine and mineral hydrolysis in coal mine give rise to the mobilization of some



• Fig. 9. X-ray diffraction pattern of the studied coal ash showing characteristic gypsum (G) and natrojarosite (J) peaks.

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Ela	Inputs		Outputs		5
Ele-	Concentration	Flux	Concentration	Flux	Li Levelle enlevent
ment	mgl ⁻¹	kg-1ha-1yr-1	mgl ⁻¹	kg-1ha-1yr-1	kg na yr
Fe	0.045	2.4	0.075	0.71	1.69
Cr	0.181	9.5	0.186	1.76	7.74
Ni	0.36	18.8	0.364	3.44	15.36
Cd	0.003	0.16	0.004	0.038	0.122
Со	0.031	1.6	0.022	0.21	1.39
Pb	0.16	8.4	0.17	1.61	6.79

Tab. 4. Geochemical mass balance of some heavy metals in Maghara catchment (during March 2007).

toxic heavy metals, causing abnormally high concentration of these elements in minewater. This was especially proved for Ni, Cr, Pb, Fe, Co and Cd, respectively. Therefore, it was important to calculate geochemical mass balance of these elements for preventing harmful pollutions leakage in groundwater of the Maghara area.

Mass balance in the Maghara area is given by the sum of inputs of toxic heavy metals mentioned above from its surrounding minus the sum of their outputs to the surrounding. A preliminary equation of mass balance (Pačes 1985 and Melegy 2005) of a toxic metal "i" in a small catchment is given bellow:

Input_i – Output_i = \sum_{i} (2) The assumption of a steady state of individual compartments is important for simplifying the mass equation to a frequently used equation 2. The terms in the equation are fluxes in g.ha⁻¹.yr⁻¹ related to a surface area of the studied catchment. \sum_{i} is the net accumulation or depletion of the toxic metal "i" in a small catchment (Fig. 10). The mechanism of the mass balance as estimat-



ed in the studied catchment (\sum_i) is given herein by the sum of inputs of trace elements minus the sum of their outputs as described by the mass balance equation.

In the studied catchment, the most important factor is the input of minewater to the Maghara catchment and the output of runoff water from the Maghara catchment where leaching to groundwater. Therefore, it is assumed that the atmospheric and mechanical erosion inputs and uptake output, are absent, and hence, their input-output of annual accumulation or depletion of toxic elements are negligible. Mass balance equation can be written in the following general form:

$$F_{mi} - F_{ri} = \sum_{i}$$
(3)
here F_{mi} is the flux of minewater discharging from abandoned

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coal mine, F_{ri} is the flux of runoff from the Maghara catchment. The difference of fluxes is positive, where the element accumulates in the catchment and it is negative where the element is de-

pleted. Whereas \sum_i is zero when the element is in a steady state. Input-output fluxes of mine water from abandoned coal mine in the Maghara catchment are given in the Table 4. Input fluxes of mine water for Ni, Cr, Pb, Fe, Co and Cd were 18.8, 9.5, 8.4, 2.4, 1.6 and 0.16 kg⁻¹ha⁻¹yr⁻¹, respectively. As well as, the output fluxes of minewater from Maghara catchment were 3.44, 1.76, 1.61, 0.71, 0.21 and 0.038 kg⁻¹ha⁻¹yr⁻¹, respectively. The mechanisms of the mass balance indicate that the highest accumulations of groundwater were 15.36, 7.74, 6.79, 1.69, 1.39 and 0.122 kg⁻¹ha⁻¹yr⁻¹ due to the content of Ni, Cr, Pb, Fe, Co and Cd, respectively. $\sum i$ (in the Table 4) indicated that the net accumulation of these toxic metals was much higher than depletion rates. Therefore, these toxic metals were leached and accumulated in groundwater.

Conclusions

Petrological, geochemical and mass balance studies in the Maghara coal mine were revealed that:

- the cessation of the Maghara mine leads to increase coal weathering and pyrite oxidation processes,
- the mechanisms of mass balance indicate that the highest accumulations of Ni, Cr, Pb, Fe, Co and Cd in groundwater were much higher than those in natural waters, and,
- the most dangerous in the Maghara area is that water, due to high contents of salinity and trace elements, is unsuitable for drinking, domestic and irrigation purposes for Bedouin people.

Acknowledgements

We thank to Dr. I. Sýkorová. (Institute of Rock Structure and Mechanics, Academy of Sciences of the Czech Republic) for helping with petrographic analyses. Also, I would like to express special thanks to Prof. J. Čurlík from the Comenius University for critically reading the manuscript and helpful suggestions.

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