

Tracing the Water – Rock Interaction in the Ibbenbüren Mine - Towards a Reactive Transport Model for Coal Mine Drainage

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Abstract

Traces of water-rock interaction in two drill cores from the West field of the former Ibbenbüren anthracite coal were related to both diagenesis and relatively recent weathering processes along open fractures. The coupled appearance of kaolinite-dickite-illite minerals in weathered and unweathered rock sections was clearly connected to the burial history of the Carboniferous sequence. In contrast, the formation of iron (oxide-) hydroxides together with the presence of oxidized pyrite in weathering profiles along both sides of the fractures was positively related to the geochemical footprint of the coal mine drainage.

Keywords: Coal Mine Drainage; Pyrite Oxidation; Water-Rock Interaction

Introduction

Coal mine drainage may often be characterized by poor water quality (Kessler *et al.* 2020). High salinity may become an environmental issue (Timpano *et al.* 2015), as it may cause toxicity to freshwater communities (Elphick *et al.* 2011). Sulfide mineral weathering can lead to the formation of acid mine drainage solutions (Galán *et al.* 2003; Neal *et al.* 2005; Nieto *et al.* 2007) in which case low pH (Simate and Ndlovu 2014) and relatively high sulfate concentrations (Wang *et al.* 2016) may have adverse toxicological effects on the receiving communities. In particular, clogging of the riverbed by the formation of hydrous ferric oxides has a negative effect on the flora and fauna of the affected rivers (McKnight and Feder 1984). In addition coal mine drainage may also carry relevant levels of toxic metals, often related to pyrite oxidation (Cravotta and Brady 2015; Gombert *et al.* 2018).

In the former anthracite coal mine in Ibbenbüren highly saline brines are present in the former East field (Rinder *et al.* 2020), whereas iron and sulfate rich solution are dominant in the former West field (see table 1).

Geological framework

The coalfield, known as the “Ibbenbürener Karbonscholle”, is a horst structure of Carboniferous origin, uplifted to the surface in the Cretaceous. As a result, the coalfield is separated from the surrounding area with an offset of up to 2000 m. It consists of two hills (Schafberg and Dickenberg) divided by a NNE-SSW striking graben structure known as the Bockradener Graben (see Figure 1). The hill chain is surrounded by Triassic, Jurassic, and, to a lesser extent, Permian rock formations in the direct vicinity of the fault structure, surrounding the block. The chain reaches a maximum height of 176 m above

Table 1 Chemical composition of mine drainage from the West- and Eastfield.

	Temp [°C]	pH	conductivity [μS/cm]	Na ⁺	Ca ²⁺	K ⁺	Mg ²⁺	Cl ⁻ [mg/l]	SO ₄ ²⁻	HCO ₃ ⁻	Fe	Ni
West field	23	3.6	3230	232	312	11	131	171	1688	37	99	0.21
East field	43	6.0	225000	77315	1193	445	464	109805	2150	100	34	< 0.05

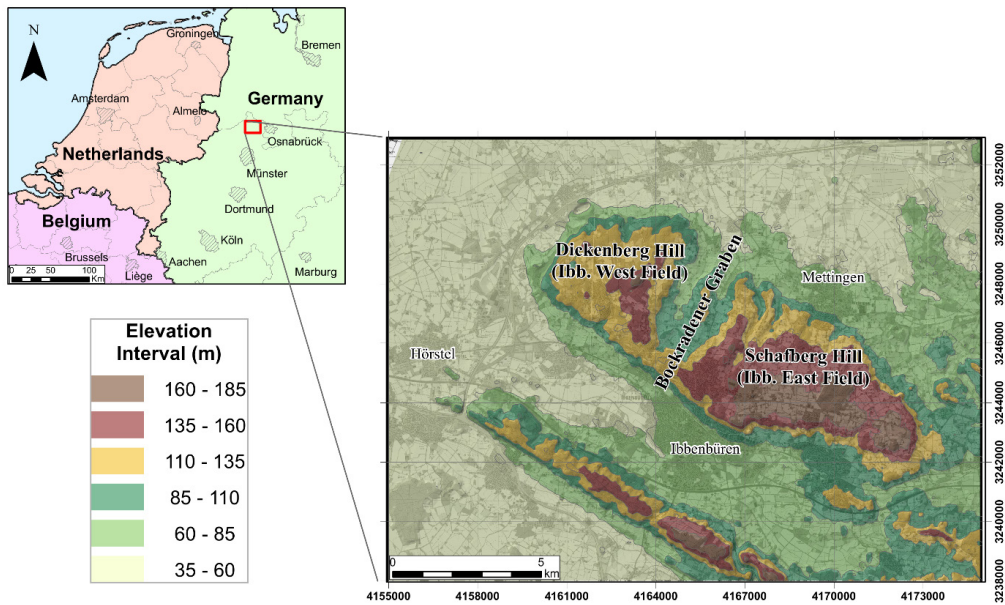


Figure 1 The Ibbenbüren coalfield; modified after (Bedoya-Gonzalez et al. 2021).

sea level, with a length of 14 km and a width of 4 to 5 km (Bässler 1970). During mining operations, the Schafberg and Dickenberg were named “East field” and “West field”, respectively.

Origin of mine drainage

In an earlier study, the salinity in the deep Na-Cl brines of the East field was assigned to halite dissolution through Na/Cl and Br/Cl ratios. Within the context of the local geological situation, the origin of these brines from outside of the coalfield was suggested by $\delta^{34}\text{S}_{\text{SO}_4}$ values typical for the dissolution of Mesozoic evaporites that surround the coalfield (Rinder et al. 2020). In addition ongoing water-rock interaction of those brines with the siliciclastic rocks of the Carboniferous host rock was indicated by radiogenic strontium. Additionally, $^{87}\text{Sr}/^{86}\text{Sr}$, $\delta^2\text{H}_{\text{H}_2\text{O}}$, $\delta^{18}\text{O}_{\text{H}_2\text{O}}$, K^+ and Li^+ signatures of the mine fluids indicated the presence of formation waters within the sandstones, contributing to the water chemistry (see figure 2).

In contrast, in shallow mine waters down to the third level \approx 270 m below surface showed a $\delta^{34}\text{S}_{\text{SO}_4}$ composition typical for the oxidation of sulfides. No influence of halite, gypsum, or anhydrite dissolution was found in those waters. A similar situation is assumed

for the West field. It represents an isolated mountain range, topographically elevated with respect to the foreland. Since flooding of the mine it discharges through the Dickenberg adit, which is elevated above the surrounding Mesozoic sediments. Accordingly, it has been hypothesized that the actual chemical signature of the drainage is influenced by the interaction of percolating rainwater with sulfur-bearing rock layers above the adit (Rudakov, Dmitry V.; Coldewey, Wilhelm G.; Goerke-Mallet 2014). Yet the chemical composition of the mine drainage changes over time, also because sulfide related acidity can be vestigial or juvenile (Younger 1997). Accordingly, the prediction of the long term chemical evolution warrants a closer investigation on possible sulfur sources.

Methodology

We investigated evidences of rock alteration in two drill cores pierced through the rock sequence above the Dickenberg adit. Drill cores were examined through a combined mineralogical and elemental analysis on thin sections using a petrographic microscope, Scanning Electron Microscope (SEM), X-ray diffraction microscope (XRD) and XRF spectroscopy. Special focus was given on signs of water – rock interaction.

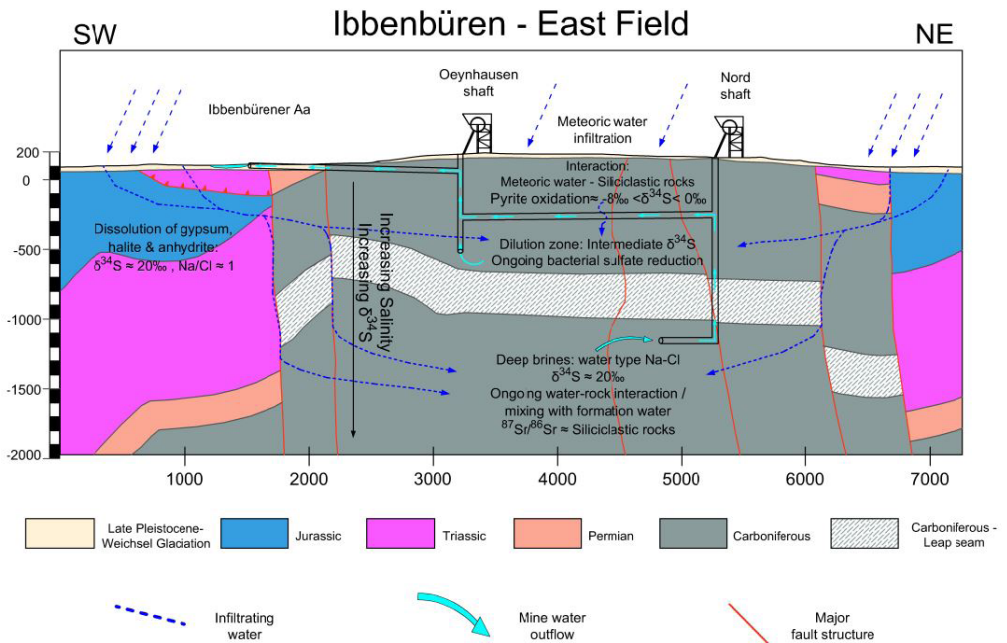


Figure 2 Schematic cross section through the Ibbenbüren Eastfield including a conceptual hydrogeochemical model, depicting the origin and evolution of mine drainage, groundwater, and brines in the mine; taken from (Rinder et al. 2020); cross section modified after (Drozdowski 1985).

Results

The sandstones display high proportions of detrital quartz, with few evidences of overgrowth or sutured contacts. Most of the lithic fragments correspond to chert and ductile mudstones with sericite (Figure 3a). Additionally, several layers contain high proportions of lenses and coal fragments as frame components, some of which exhibit oxidation signs on the surrounding matrix (Figure 3b). Muscovite is detrital and together with the ductile lithics is commonly affected by compaction, being incorporated into the primary pore spaces as pseudomatrix (Figure 3a). Pyrite occurs as clusters of very fine sand and silt crystals (microcrystalline pyrite) usually associated with sedimentary lithic fragments and rarely as frame macrocrystals (Figure 3c). Finally, iron (oxide-) hydroxides were found on lithoclasts surfaces and coating the framework grains of the core intervals crossed by fractures (Figure 3d). SEM and EDX images reveal trace quantities of barite and heavy clay-size particles, enriched in strontium, lead, and Rare Earth Elements

(Lanthanum, Cerium and Neodymium). These are encountered in the primary porosity of the rock, alongside the clayey matrix (Figure 3f).

Traces of water-rock interaction were assigned to two different categories. Illite, kaolinite and dickite are evenly distributed among the matrix of the weathered and unweathered rock zones and are related to the diagenesis history of the area. On the other hand, the presence of iron (oxide-) hydroxides along the fractures is linked to the dissolution of pyrite and some influence of juvenile acidity would be suggested from the geological situation. However, relatively high iron contents in the weathered zone face relatively low pyrite contents in the unweathered rocks. For instance, preliminary batch experiments on the investigated unweathered sandstones did not generate high iron or sulfate contents. The low porosity and permeability of the analyzed samples exclude deep percolation of infiltrated rainwater into the rocks. Thus, open fractures, possibly originated from mining activities, may play a significant role

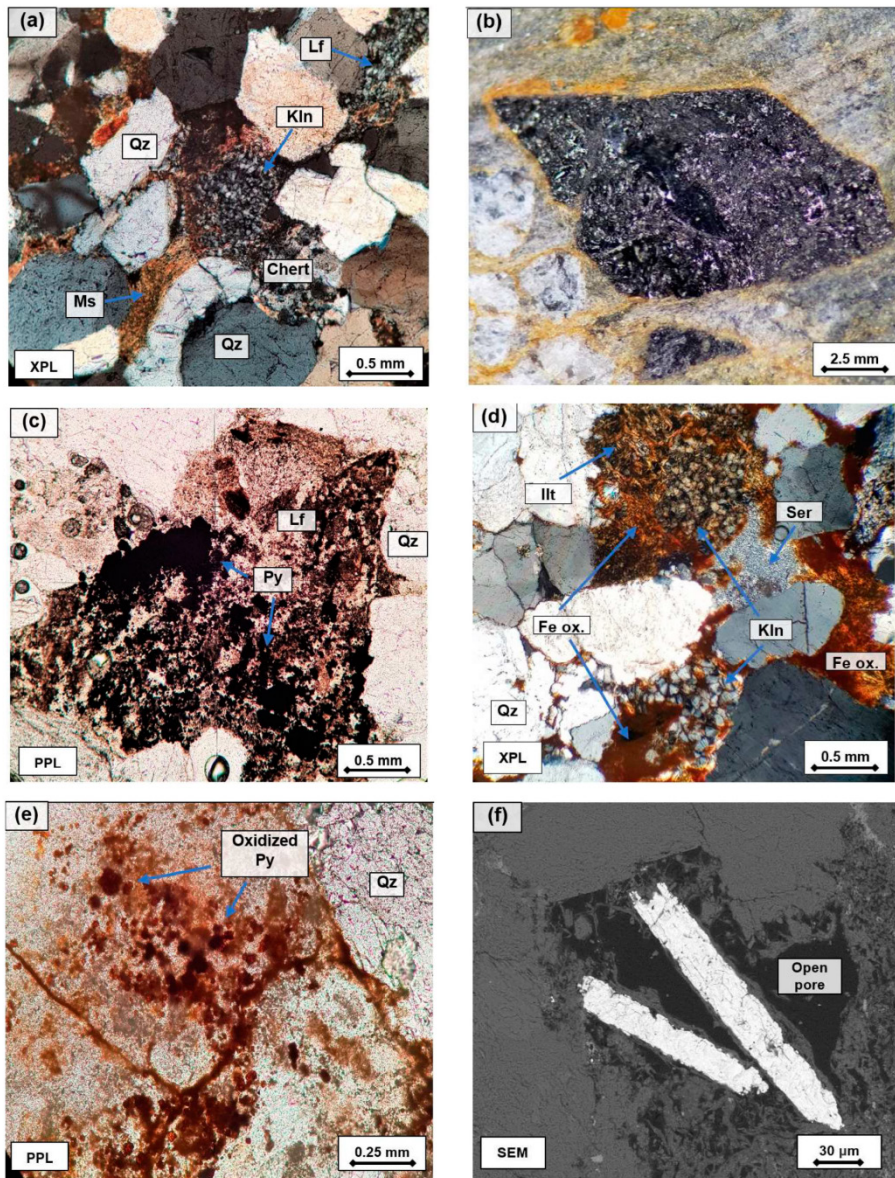


Figure 3 Photomicrographs of the main components found in the rock sequence. Lf = lithic fragment, Ms = muscovite, Kln = kaolinite-group minerals, Qz = quartz, Py = pyrite, Ill = illite, Ser = sericite, Fe ox. = iron (oxide-) hydroxides, PPL = plane polarized light, XPL = cross polarized light, SEM = scanning electron microscope; taken from (Bedoya-Gonzalez et al. 2021).

in the drainage chemistry. The high contents of iron (oxide-) hydroxides could also be the result of outflowing formation water, within a relatively short timespan after the opening of the fractures. Such an explanation however, is at odds with a relatively constant and high sulfate content in the mine drainage during the

last decades. Alternatively, pyrite dissolution from outside of the analyzed core samples is also possible. This points to the heterogeneity, observed within the sedimentary sequence and the challenge of upscaling observations from drill cores to complete rock sequences. The setup of a reactive transport model in the

future will hopefully provide more insights into the conditions necessary to generate the observed sulfate contents.

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