Extended Abstract

In the course of open pit lignite mining large quantities of overburden sediments are shifted. Due to the thereby proceeded aeration and the temporary exposure of the dump to atmosphere until the deposition of covering layers as much as by the groundwater lowering the oxidation of irondisulphides is initiated. The release of acidity leads to the formation of acid mine drainage with low pH-values and high contents of iron and sulphates as well as the mobilisation of heavy metals. Caused by the decrease of lignite mining in Central Germany a number of abandoned mines is flooded. A joint project of the Institute of General and Applied Geology of the Ludwig-Maximilians-University Munich and the Environmental Research Center Leipzig-Halle Ltd. deals with the investigation of hydrochemical and hydraulic processes in connection with the flooding of the abandoned lignite mine Cospuden south of Leipzig.

The geochemical characteristics of the dump material, i.e. the acidification potential caused by the oxidation of the irondisulphides is determinative for the hydrochemical setting, in particular for the pH-value of the mine-groundwater. With pyrite concentrations up to 8 wgt.-% on the average the material of the overburden dump contains no increased tracemetal concentrations in the solid, however, the mobility and thus the concentration of the analysed tracemetals (Cr, Co, Ni, Cu, Zn, As, Cd, Pb) in the dump water correlates closely with the pH-value of the percolating water.

With the registration of hydrochemical main-parameters (pH, E_h, κ) predictions about the extent of the loads of the eluates can be done. Because of the buffering of the pH-values within the range of ≥ 6 these concentrations are limited below the ecotoxic limiting values. In this pH-range carbonate-buffering is effective. The mode of operation of different carbonatic systems shows different results. If hydrogencarbonate-bearing water runs through acid dump material, a rise of the pH-value from pH 2.2 to approx. 4.5 and thus a reduction of initial high solution loads of sulphate, iron and traceelements can be noticed. If acid mine water with high solution loads flows into carbonatic dump sediments, as a consequence of neutral pH-values there is an overall smaller discharge of products of pyriteoxidation. For a complete buffering of the acid resulting from the pyriteoxidation by calciumcarbonate, however 3.3 wgt. % $CaCO_3$ per 1.0 wgt.-% FeS_2, would be required.

The discharge of Fe_{tot} and $SO_4^{2-}{}_{tot}$ with pH-values < 3 corresponds with a stoichiometric ratio of 1:2 released by the oxidation of irondisulphides. This ratio is shifted by the precipitation of ironhydroxides with rising pH-values towards to superproportional sulphate discharge. In the pH-range below 2.9 the seepage water is buffered due to precipitation and solution reactions of iron(III)hydroxides. In acid eluates (pH < 4.5) the permitted limit values for traceelement concentrations are exceeded by a multiple.

The almost pyrite-free area, a result of secondary oxidation due to oxygen diffusion into the dump achieves after one year of exposure a thickness between 1 m and 2 m. In these investigations a decrease of irondisulphides contents within one year was determined with originally 3.0-3.4 wgt-% to 0.6 wgt.-%.

An obvious effect on the discharge-rate of the dissolved elements shows the way of percolating the laboratory columns. Thus an intermittent flow shows in relation to an even flow approx. 12 % higher discharge rates. By comparing concentrations of the solid before beginning the experiment and after finishing it the specific proportional solution of the originally contents could be determined. Here a decreasing sequence of the relative solubility is given with Cd > Ca > Co > Mn > As > Ni > Mg > Zn > Fe > Pb > Al > Cu > Cr > K > Na.

By investigating the bonding-types of the traceelements by means of the method of sequential extraction it could be shown that out of the area of the temporary superficially exposed oxidation zone in contrast to the internal area of the dump higher contents of Fe, Cr, Co, Ni and Pb were eluated. On the other hand from the dump area zinc and cadmium were solved in larger quantities. A shifting of the bonding strength of the traceelements from larger proportions of strongly fixed and moderately mobile parts in the internal dump area to larger proportions of weakly mobile up to the residually fixed parts within the area of the oxidation zone could be determined. This is a hint on the fact that from that decomposition zone the easily mobilizable proportions were already removed.

By comparison of the traceelement concentrations in the eluating waters and the element concentrations in the sediments a prediction about the mobilization can be made. A decreasing relative solution in the following order was determined: Ni = Zn = Co > As > Cu > Cr > Pb > Cd. For cobalt, nickel and zinc

clear correlations to iron were shown. This leads to the conclusion that these elements are mobilized from iron(II)sulphides by oxidation contrary to the not correlating elements copper, cadmium and lead.

By means of thermodynamic modelling species-calculations were carried out. They show that almost the entire iron discharge result in Fe^{2+} or Fe^{2+}-complexes. Approx. 35-45 % of the total sulphate contents are bonded in sulfocomplexes. Saturation calculations provide evidence that limitation of sulphate contents by precipitation of gypsum is only partly possible, since only small supersaturation exists with concentrations exeeding approx. 2000 mg/l. With simultaneous undersaturation in primary mineral phase calcite at pH-values < 6.5 it comes to the transition of the Ca^{2+} -cation to the secondarily formed mineral phase gypsum.

Near-surface waters flowing to the lake were analysed regarding to their mining-influenced variables (pH, E_h, κ, HCO_3, Cl, Na, Ca, Mn, Ni, Zn). By means of statistical procedures they could be determined according to their origin out of the quarternary or the tertiary aquifer, the dump, as well as from the nearby situated open pit minings Zwenkau and Profen. A potential impairment of the lakewater quality due to high solution loads (up to 10 g/l) and an exceeding of the tracemetal limits (above all nickel, cobalt and zinc) in the dump water is determined.

According to this low pH-values within the dump area and the southern area of the tertiary aquifer control the mobility of heavy metals whereas above all nickel and cobalt occur in very high concentrations. A classification of the waters in Ca-SO_4 resp. Fe-SO_4-types displays a moderate to strong load by sulphide oxidation products. Because of the small discharge (± l/min.) of the individual wells of the quarternary and the tertiary aquifer and the waters withdrawing from the dump embankment an impairment of the quality of the lakewater is not to be expected. However on a long-term basis the acidifying potential after finishing the flooding of the dumps becomes obvious.

On the basis of preceding field investigations in the overburden dumps large unsaturated areas, hanging bodies of water and systems of preferential flows were determined. In the course of a simulation of a dump-section on laboratory scale and for parameter definition for hydrodynamic computer modelling hydraulic sedimentparameters were determined such as the distribution of grain- and poresize and the unsaturated hydraulic conductivity. According to this the rate of micropores in the unweathered area is increased about three times compared with the surface. The saturated hydraulic conductivity calculated from the particle size distribution comes within the range of $1.4*10^{-7}$ ms^{-1} (unweathered) to $2.1*10^{-6}$ ms^{-1} (weathered). A small-scale simulation of a dump-section on laboratory scale and the investigation of the saturation process verify the inhomogenous saturation due to the applied method of deposition along formerly oxidized layers.

The unsaturated hydraulic conductivities - determined by the experimentally created water retention curves- comes, in respect of their moisture, in the range of $4.5*10^{-8}$ ms^{-1} (unweathered) to $1.5*10^{-7}$ ms^{-1} (weathered) and therefore approximately an order of magnitude below the values for the saturated status. From this distribution of saturated and unsaturated hydraulic conductivities determined here the inhomogenous pattern of saturation bound to the dump structure is well explainable.

For the determination of the extent of load of the dump groundwater due to mining industry-induced hydrogeochemical processes the sulphate concentration can be used in a first estimation, since by the solution of the irondisulphides it correlates with the Fe_{tot}- concentrations and beyond this with some traceelement concentrations and the pH-value, determining the mobility of the heavy metals.

Considering the required period to reduce the sulfate concentrations in the groundwater below the geogene background value of approx. 1900 mg/l by dilution and removal - however not by $CaCO_3$-buffering and fixing -, time intervals of 400 to 4000 years arise corresponding to a pore water exchange of approx. 10 until 20 times. From this on a long-term basis the enormous load potential of the irondisulphide oxidation products stored in the dumps becomes clear.

The actual load of the dump-water depends on the extent of solution resp. re-fixing of ecotoxic traceelements in secondary minerals and thus on the development of the pH-value of the dump groundwater. The kinetics of oxidation processes and the propagation of the oxidation zone into the dump are closely connected with the availability of oxygen and thus with the development of the state of saturation bound to the dump structure resp. the covering of the dump. Small Fe^{3+}-concentrations permit anaerobic oxidation of the irondisulphides only in subordinated extent.

The estimation of the development of the load of the waters is exclusively based on inorganic reactions. The effects of the organic contents as much as the microbiology on the complex reactions are here unconsidered and must be subject of further investigations.