# FLOODING OF THE SCHLEMA-ALBERODA URANIUM MINES: ASPECTS OF THE PROGNOSIS REGARDING THE URANIUM CONCENTRATION IN FLOOD WATERS

# ZATÁPĚNÍ URANOVÝCH DOLŮ SCHLEMA-ALBERODA: PROGNÓZY VÝVOJE KONCENTRACE URANU V ZÁTOPOVÝCH VODÁCH

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### Abstract

The main points of interest of this paper are several uranium mines in Western Saxony which have been flooded or are in a flooded state. Particularly significant are the large Schlema-Alberoda uranium mine in the vicinity of the city of Aue, and the small Pöhla mine on the border with the Czech Republic. Both mines are in the final stage of remediation by WISMUT GmbH. Uranium is the only main pollutant that is emitted by WISMUT GmbH into the Zwickauer Mulde River. The main sources are the Schlema-Alberoda mine and the largest mining waste rock pile at the development site. WISMUT was thus prompted by the authorities to erect and operate a special water treatment facility for the seepage water from the aforementioned largest waste rock pile in order to remove uranium. The maximum level of residual uranium concentration tolerated in flood water and seepage water is currently 0.5 mg/l, for which the relevant authorities are considering an appropriate reduction.

All these facts underline the importance of uranium in the remediation of the Schlema-Alberoda mining area.

## **Abstrakt**

Uranový důl Schlema–Alberoda se nacházel v rozsáhlé těžební oblasti podkrušnohorských pánví v údolí řeky Zwickauer Mulde, SSZ od města Aue a je společně s hlušinovým odvalem hlavním zdrojem uranu ve vypouštěných vodách. Společnost WISMUT GmbH zde provádí od roku 1990 komplexní sanační a likvidační práce uzavřeného dolu. V rámci realizace Evropské směrnice byla společnost WISMUT vyzvána vybudovat čistírnu průsakových vod z odvalu za účelem odstranění uranu v těchto vodách, jehož koncentrace dosahovala až 8 mg/l. V současné době je maximální koncentrace uranu ve vypouštěných vodách z ložiska a průsakových vod z odvalu na hodnotě 0,5 mg/l. Všechny tyto skutečnosti podtrhují význam sanace ložiska Schlema – Alberoda.

**Key words:** mine waters, flooded mines, uranium concentration.

# 1 INTRODUCTION

The main points of interest of this paper are several uranium mines in Western Saxony which have been flooded or are in a flooded state. Particularly significant are the large Schlema-Alberoda uranium mine in the vicinity of the city of Aue, and the small Pöhla mine on the border with the Czech Republic. Both mines are in the final stage of remediation by WISMUT GmbH (see Fig. 1).

In addition to these, there exist several older uranium mines which were closed down as long ago as the mid 1960s, and have been flooded for decades. Of particular interest are the:

- Schneeberg silver and cobalt mines directly neighbouring Schlema-Alberoda;
- Johanngeorgenstadt uranium mine on the Czech border;

• and the Zobes uranium mine in Vogland, not far from the city of Plauen.

The Schlema-Alberoda uranium mine is situated beneath an extensive mining area on both sides of the Zwickauer Mulde and the Bad Schlema locality. WISMUT GmbH has been carrying out complex redevelopment measures in this area since 1990. Many sites, and above all waste rock piles alongside them, were affected by this for days. These redevelopments take into consideration the local interest in the revival of the Oberschlema radon springs, which had been liquidated as a result of the uranium mining. In the new spa area, the redevelopment has already been completed.

The flood water that has risen in the uranium mine is currently at a level of 20 m under the hydraulic point of discharge into the Zwickauer Mulde River. To keep the area clear, this point of discharge was newly created over a special mining development downstream from the area. This development ends at a 50 m deep shaft on the site of the water treatment facility. The flood water is pumped out of this spring for treatment (Figs. 2; 3).

The mine flooding began in 1991 and was initially controlled by the regular stack pump system starting from the operating time of the mine, which pumped out lightly polluted infiltration water. By 1997, this pump system had become inoperative due to excessive flooding. A new system of under water pumps has been in service since 1999, controlling the flooding by pumping out flood water.

After a dynamic phase of flooding, only a slow increase in the flooding level has been instigated since 2000, in order to guarantee the mining stabilization of the inhabited area and to rule out severe geomechanical flooding effects (subsidence/uplift; seismic reactions) (Fig. 4).

#### 2 HYDROCHEMICAL EFFECTS OF FLOODING

The mine flooding led first of all to a clear mobilization of substances from the mine stock. Environmentally relevant components are uranium; radium ( $^{226}$ Ra); arsenic; iron and manganese. Until 1999, a notable increase in the concentration of uranium of up to 8 mg/l could be recorded. The commissioning of the treatment of the flood water brought about an initial steady decline in the concentration of uranium to the current level of 2 mg/l, at which the rate of decline is gradually decreasing. Observations of the balance of materials show a dilution of the flood water with less polluted infiltration water to be the significant cause of the decrease in concentration (Fig. 5).

The treatment of the flood water with regard to uranium is expensive. The long-term average amount of flood water in the mine is 7 million  $m^3/a$  with an annual fluctuation margin from approx. 6 million  $m^3/a$  to 9 million  $m^3/a$ . The current annual costs amount to  $\in$  6 million, of which the removal of uranium makes up about a half.

Uranium is the only main pollutant that is emitted by WISMUT GmbH into the Zwickauer Mulde River. The main sources are the Schlema-Alberoda mine and the largest mining waste rock pile at the development site (Fig. 6). Another significant pollutant is arsenic, which for the most part originates from the Schneeberg ore mine not run by WISMUT GmbH. The relevant authorities are showing an increasing interest in both parameters in connection with the implementation of the European Union Water Framework Directive at the Zwickauer Mulde River.

WISMUT was thus prompted by the authorities to erect and operate a special water treatment facility for the seepage water from the aforementioned largest waste rock pile in order to remove uranium. Other rock piles could follow if necessary.

The maximum level of residual uranium concentration tolerated in flood water and seepage water is currently 0.5 mg/l, for which the relevant authorities are considering an appropriate reduction.

All these facts underline the importance of uranium in the remediation of the Schlema-Alberoda mining area, as well as the interests of WISMUT in a reliable prognosis of perspective uranium emissions.

A special hydrochemical aspect here is the possibility of the in situ immobilization of uranium in flooded mines by way of chemically reductive processes.

## 3 CHEMICALLY REDUCTIVE URANIUM REMOVAL

For the purposes of evaluation, a comparison will firstly be drawn of the two WISMUT mine floodings in Schlema-Alberoda and Pöhla.

The physical parameters in situ have up until now shown the following environmental conditions (Fig. 7):

- neutral pH levels;
- warm, mineralized flood water;
- low oxygen content;
- intermediary/decreasing redox milieu.

With regards to the hydrochemical parameters of the flood water in each mine, each showed partly corresponding, partly contrary developments.

In both flooded mines, an intensive convective mixing of the flood waters was observed. A significant cause of this is the thermal influence on the water as a result of the geothermal temperature gradients in the rock mass (3 - 3.5 K/100m) together with a multitude of mine workings which it can flow through.

At the Pöhla mine, a rapid decrease in the concentration of uranium, sulphate and manganese to a very low residual level was already established during the flooding (Figs. 8-10). The levels of uranium subsided from 4.5 mg/l to <0.1 mg/l. As a result, the removal of uranium within the complex water treatment process could be completed ahead of schedule. In contrast to this, a long-term increase in concentration in both parameters initially took place in the Schlema-Alberoda mine during the flooding, which, after a phase of stagnation, has since given way to a lengthy period of decrease. (See above).

The developments in the levels of arsenic and radium (<sup>226</sup>Ra) are also conflicting (Figs. 11; 12). In Pöhla, the levels of concentration increased during the flooding, and have remained static at this level since the end of the flooding (1995). In Schlema, the increase during the further flooding was followed by a very clear (As) and moderate (<sup>226</sup>Ra) decrease according to exponential time functions.

For iron, both mines showed differing strong increases in concentration in the initial stage of the flooding, followed by a moderate decline. Since then, both mines have shown identical, largely static concentration levels (Fig. 13).

In contrast to the fundamentally similar concentrations of pollutants of both flooded mines, Schlema-Alberoda shows significantly higher concentrations of the main cations and anions (for instance HCO<sub>3</sub>; Fig. 14). The content of these in the flood water reached maximum levels more or less ahead of schedule, and have since been in a continual decline, according to exponential time functions.

From these brief characterizations of individual parameters, the present development conditions of both mines can be subdivided as follows:

- increases in concentration as a result of the mobilization of long-established weathering processes in the air-filled mines;
- occasional stagnation in concentration as an effect of chemical buffer substances (SO<sub>4</sub>; HCO<sub>3</sub>);
- gradual decrease in concentration through a mixing of the contaminated flood water with substantially less polluted infiltration waters;
- for Pöhla: rapid decrease in concentration through specific processes of reductive immobilization (U; SO<sub>4</sub>; Mn), and from follow-up processes (<sup>226</sup>Ra).

As such, intensive reductive immobilization processes have up to now only been able to be identified in the Pöhla mine. The parameters affected by this are  $O_2$ ;  $NO_3$ ;  $Fe^{3+}$ ;  $Mn^{3+}$ ;  $SO_4$ ; U; TOC. In the Schlema-Alberoda mine, so far only a weak chemical reduction seems to have taken place, which has affected the parameters  $O_2$ ;  $NO_3$  and  $Fe^{3+}$ . This significant difference exists despite the fact that the physical flood water milieu in both mines is nearly similar.

The question arises as to the causes of this difference and to the prognostic development in the Schlema-Alberoda mine, in particular concerning uranium.

For Schlema-Alberoda, one must take into account that in this mine, in the upper non-floodable part of the mine, a spacious mine ventilation with an exhaust shaft is operated, which is not the case in Pöhla. Furthermore, in contrast to Pöhla, the Schlema-Alberoda mine is overlaid by an expansive waste rock pile complex, whose leachate is contaminated (up to 5 mg/l U and 5g/l SO<sub>4</sub>), and could have an effect on the flooded area of the mine.

In addition, there is the assumption that the relatively high concentration of sulphate mobilized in the flood water acts hydrochemically as a redox buffer, which could block the following reduction step with regards to uranium. Such a dependency could be empirically deduced from the flooding process in Pöhla. There, the reductive immobilization of sulphate was established earlier than that of uranium. The immobilization of uranium was seemingly only established at a residual concentration level of  $SO_4$  of approx. 250 mg/l (Fig. 15).

The question is whether this happened as a coincidence or if it corresponds to a fundamental regularity.

Additional information from the flooding of other uranium mines is useful in helping to answer this question, although their particular characteristics are to be taken into consideration. The examination of the aforementioned uranium mines in Western Saxony which have been flooded for decades provided some useful information (Fig. 16).

The uranium mine in Zobes bears clear similarities to the Pöhla mine. The flood water shows a reducing milieu, uranium and sulphate are immobilized, while radium continues to be emitted in high concentrations. Apart from the current measurements, however, no time functions of flood water parameters are available for an evaluation of the hydrochemical development during and after the flooding.

This equally applies to the Johanngeorgenstadt uranium mine. Here, from medium to decreasing redox ratios, low concentrations of uranium (0.01 mg/l) and moderate concentrations of sulphate (50 mg/l) are currently recorded, at which there are elevated levels of radium (0.3 Bq/l).

The further metrologically accessible old Schneeberg ore mine manifests completely different conditions. Here, oxidising conditions prevail, which result from an intense flow of large annual quantities of fresh infiltration water through a small flooding volume. The similarly low concentrations of uranium (0.01 mg/l) result from this, as well as from the already low uranium stocks of the deposit.

The number of well known "cases" of evaluable mine floodings and corresponding measurements is still rather low to be able to make empirical generalizations. Therefore, an extension of the "case count" of relevant mine floodings in the region of the Czech Republic should be tested, as many uranium mines were operated here.

Particularly important are current and possible historical measurements of the composition of the mine water during and after the flooding. This with the background of information about the mining and hydrogeological conditions of the mine. The Pribram uranium mine should be included in this, as a direct comparison of this with the Schlema-Alberoda mine would seem to be appropriate.

## RESUMÉ

Uranový důl Schlema–Alberoda se nacházel v rozsáhlé těžební oblasti podkrušnohorských pánví v údolí řeky Zwickauer Mulde, SSZ od města Aue. Společnost WISMUT GmbH zde provádí od roku 1990 komplexní likvidační a sanační práce uzavřeného dolu. Fáze dynamického přirozeného zatápění dolu trvala od roku 1991 do roku 2000. Následovalo řízené pozvolné zatápění při současném odčerpávání důlních vod s cílem stabilizace těžbou postižené obydlené oblasti, vyloučení závažných geomechanických účinků zatápění (poklesy, zdvihy terénu, seismické jevy) a minimalizace zatížení hydrosféry oblasti. Zaplavení dolu způsobilo mobilizaci environmentálně závažných polutantů (U, <sup>226</sup> Ra, arsen, železo a mangan). Postupně do roku 1999 byl v důlních (záplavových) vodách registrován nárůst koncentrace uranu, a to až na hodnotu 8 mg/l. Po zavedení čištění důlních vod byla snížena koncentrace uranu ve vypouštěných vodách na úroveň 2 mg/l. Průměrné roční množství záplavových vod je cca 7 mil. m³ a roční náklady na jejich čištění jsou 6 mil. € (z toho cca polovinu tvoří náklady na separaci uranu).

Hlavním zdrojem uranu ve vypouštěných vodách je důl Schlema–Alberoda a jeho hlušinový odval. V rámci realizace Evropské směrnice byla společnost WISMUT GmbH vyzvána vybudovat čistírnu průsakových vod z odvalu za účelem odstranění uranu v těchto vodách. V současné době je maximální koncentrace uranu ve vypouštěných vodách z ložiska a průsakových vod z odvalu na hodnotě 0,5 mg/l.

V dole Schlema–Alberoda a Pöhla jsou specifické hydrochemické podmínky, které umožňují imobilizaci uranu v záplavových vodách také díky redukčním podmínkám. V obou dolech dochází k intenzivnímu míšení záplavových vod, které je způsobeno geotermálním gradientem skalního masívu (3 až 3,5K/100m) a rozsahem důlních děl, kterými může voda protékat. Na dole Pöhla byl prokázán během zatápění rychlý pokles koncentrace uranu (ze 4,5 mg/l na <0,1 mg/l), síranů a manganu, což umožnilo ukončení čištění důlních vod ještě před

plánovaným termínem. Naopak v dole Schlema–Alberoda docházelo během zaplavování k nárůstu koncentrace těchto polutantů, což prodlouží dobu jejího očekávaného poklesu. Vývoj koncentrace arsenu a radia ( $^{226}$  Ra) je u dolu Pöhla a Schlema–Alberoda odlišný. Na dole Pöhla došlo ke zvýšení koncentrace během zatápění na úroveň, která zůstává od ukončení zatápění (1995) konstantní. Na dole Schlema–Alberoda je znatelný nárůst během zatápění a později velmi výrazný (u As) a mírný (u  $^{226}$ Ra) pokles. V obou dolech bylo při zatápění prokázáno silné počáteční zvýšení koncentrace železa s následným pozvolným poklesem na úroveň, která však již nyní zůstává stabilní.

Výrazné redukční prostředí bylo prokázáno jen v dole Pöhla přesto, že fyzikální podmínky záplavových vod v obou dolech jsou velmi podobné. Možnou příčinou je, že oproti dolu Pöhla se v dole Schlema–Alberoda nacházejí nezaplavené části s funkčním ventilačním systémem. Na povrchu je důlní pole překryto rozsáhlými odvaly, ze kterých mohou průsakové vody infiltrovat do důlních děl (dotace až 5 mg/l U, 5g/l  $SO_4^{2-}$ ).

Pro obecné empirické závěry je stále ještě málo potřebných údajů a proto byla využita možnost výzkumné spolupráce s odborníky v České republice, kteří mají k dispozici aktuální i historická měření složení důlních vod, informace o těžbě a hydrogeologických podmínkách uranových dolů na svém území. Předmětem zájmu je zejména ložisko Příbram, které je vhodné pro srovnání s dolem Schlema–Alberoda.

**APPENDIX**: Presentation "Flooding of The Schlema-Alberoda Uranium Mine / Prognosis of Uranium Concentration" \*) - October 2009 in Königstein, Germany

<sup>\*)</sup> Appendix of individual articles has not language correction