MATHEMATICAL MODELLING APPLICATION FOR OPTIMISATION OF UTILISATION OF MINE WATERS WITH HIGH URANIUM CONCENTRATIONS FROM FLOODED MINES

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ABSTRACT

The paper summarizes the results of the research project Non-traditional Utilization of Uranium Deposits after Underground Mining Completion" which has dealt with problems of the utilization of mine waters of abandoned and flooded uranium mines as a secondary source of uranium. After flooding the deposit, the intensive dissolution of uranium minerals takes place and uranium concentration in the waters increases markedly. Simultaneously, the vertical stratification of mine waters occurs; in deeper horizons of the former mine, uranium concentrations are conspicuously higher that in the subsurface parts of the mine. Potential utilization of these mine waters as a secondary uranium source is, to a certain extent, analogous to the "in situ leaching" method of mining. Nevertheless, the process of transferring uranium into solution is not intensified by introducing acids into the rock environment, but instead the natural processes of dissolving uranium minerals in mine waters after their preceding oxidation in the stage of deposit exploitation are used.

The project has dealt with possibilities of intensive utilization of mine waters with high uranium concentrations as secondary source of the raw material with the accompanying effect of shortening the time necessary for the treatment of mine waters drained. By means of the field studies, theoretical calculations and mathematical modelling (geochemical models and groundwater flow and uranium reactive transport models in the environment with dual porosity- FEFLOW) the solution has been optimized and recently tested by pilot field study.

1. INTRODUCTION

From the year 1945 to the middle of the 90's of last century, uranium mining belonged to the important branches of industry in the Czech Republic, and as far as the production of uranium concentrate is concerned, the Czech Republic held a foremost position in the world. At present, the mining of uranium is performed merely in one underground mine in the deposit of Rožná with the planned completion of mining operations in the year 2010.

However, because the demand for uranium as an energy raw material is significantly growing, matched by market price increases, substitutes for depleted resources are intensively sought after. Likewise the possibilities of using non-traditional methods for obtaining this raw material from the rock mass are being thoroughly examined. This paper is based on the results of a project dealing with the possibilities of the intensive utilization of mine waters from flooded uranium mines as a source of uranium with the additional effect of shortening the time period necessary for the purification of mine waters discharged into surface watercourses. These possibilities arise because the mine waters from flooded former uranium mines represent a significant secondary source of this raw material due to their considerable volumes and the high concentration of dissolved uranium. The idea of utilizing mine waters from closed and flooded uranium mines, is based on the spontaneous natural process of uranium minerals dissolving in mine waters after their previous oxidation during the exploitation stage. To a certain extent, this is analogous to the mining method "in situ leaching". In contrast to this method however, the process of uranium transfer to the solution is not intensified by adding acids into the rock environment, rather the natural processes for dissolving uranium minerals in mine water are utilized instead. If the present intention to utilize mine waters from flooded uranium mines as a secondary source of raw material is supported by theoretical calculations and modelling, then the subsequent implementation of the scheme will enable a non-negligible amount of uranium to be acquired (for instance, in the waters of the flooded uranium deposit at Příbram, the estimate is hundreds of tons of uranium). The subsequent reduction in the time necessary for mine water purification, which is financed by the Czech government, would also be significant.

2. PRESENT STATE AND METHODS OF SOLUTION

When uranium ore exploitation was finished in the mines and the pumping systems were shut down, the process of spontaneous mine flooding started. Depending on the amount excavated, the depression cone area and the hydrogeological conditions of the deposit, this process took several years.

During this time, conditions for proper mine water management in the "collection – controlled draining from underground spaces – purification – discharge" mode had to be created in advance. This ensures that shallow underground and surface water is not threatened by uncontrolled leakage of contaminated water from the flooded mine in future.

The shut-down of uranium mines on the vein, zone and metasomatic deposit types consisted primarily of filling the main mine outlets at ground level, the so-called main shafts. Raises and mining areas coming up to ground level were filled as well. Under the geological and hydrogeological situation in these locations, mining methods used, and in many cases, the considerable underground mining depth, it was not necessary to backfill other mine workings or other open underground spaces in connection with their liquidation. Unconsolidated backfill was used to fill the shafts, raises and near-surface stopes; untreated material from mine dumps created during excavation was used as a backfill material.

For the detailed research, a pilot locality has been selected, namely the Olší-Drahonín uranium deposit, where exploitation was finished in 1989 and since the year 1996, excess waters have been discharged (under control) from the deposit and subsequently purified. In the deposit, hydrological steady-state exists and sufficient data was available for project use. Basic research is therefore being carried out in this locality.

The deposit at Olší – Drahonín was mined between 1959 and 1989. At the time exploitation of deposits ceased, mining operations were at depths below level 10 (+18 m above sea level, i.e. 467 m below ground level) and the deposit was opened by a blind shaft as deep as the 18th level (374 m below sea level, i.e. 859 m below ground level). Ore bodies of the Olší – Drahonín deposit were characterised by their irregular shape, variable thickness and uranium content. Small and medium-sized bodies developed and were predominant on levels two and three. With depth, the size of ore bodies diminished and the coefficient of mineralisation decreased as well. The richest ore bodies were localised on levels 2 to 5 of the deposit. Generally it can be stated that ore bodies had complicated internal structures; ore mineralisation in them was non-continuous and occurred in irregular ore lenses and layers.

In this locality, the progress of mining operations and mine drainage, the geological structure of the deposit, the hydrology and hydrogeology of the area and also of the mine and the hydrogeochemical regime of mine waters are all well documented (Hájek et al., 2006).

Since 1997, the mine water level has been maintained by pumping at levels of 1.5–7.0 m below the overflow level (the floor of the drainage adit), i.e. at a level ranging from 449.8 to 444.3 m above sea level. Therefore contaminated mine waters will not infiltrate spontaneously and without control into the surface water. The total amount of purified water was 218,950 m^3 in 2007, i.e. the average rate of outflow from the mine was about 6.9 $l.s^{-1}$. During mining, the maximum pumped quantity of mine waters was recorded in the years 1981 and 1986, and amounted to about 17.0 $l.s^{-1}$.

The average content of uranium in waters pumped and drained from the flooded deposit through the drainage adit to the water purification plant has gradually decreased from 11.7 mg/L in 1996 to 5.9 mg/L at present (see Figure 1).

A declining trend in the uranium concentration of mine waters drained through the drainage adit from the flooded Olší – Drahonín deposit (Figure 1) is completely logical, because in the upper part of the aquifer, a so-called shallow circulation of mine waters, there is a significantly higher proportion of waters infiltrated from atmospheric precipitation or infiltrated from surface watercourses. Thus this trend is not representative of the uranium content in the aquifer of accumulated waters in abandoned mine workings as a whole. Within the terms of the research we have been conducting, just those waters accumulated in deeper parts of the former mine, in a so-called quasi-stagnant regime, form the environment of interest. The extent of shallow circulation depends on the hydrogeological conditions and the method in which the deposit was developed, as well as the flow of waters induced by controlled drainage of mine waters (either by pumping or by gravity); whereas quasi-stagnant waters are impounded in the mine, almost without movement, and the concentration of dissolved substances, is markedly higher than in the shallow circulation waters. The expected distribution of mine waters in the flooded mine is illustrated in Figure 2.

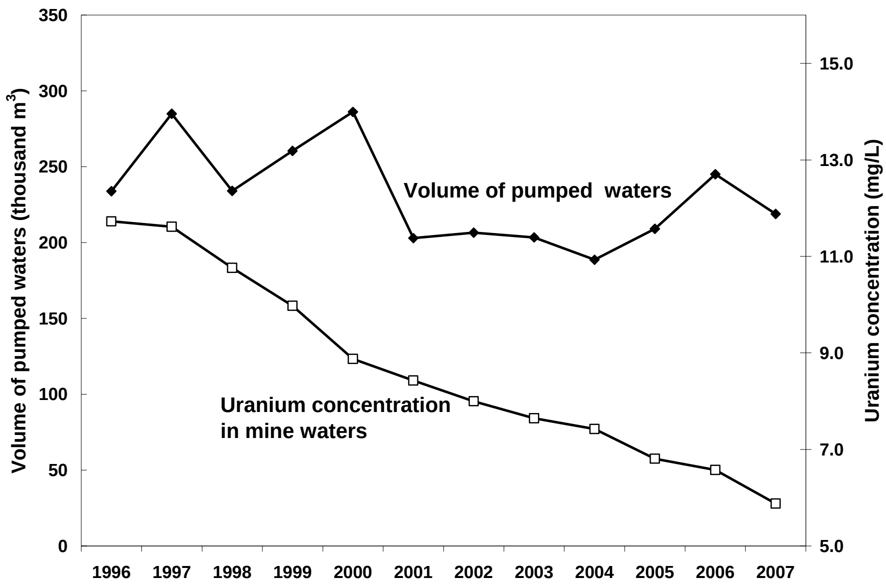

Figure 1. Amount of mine water and uranium concentration in mine waters drained from the flooded Olší – Drahonín deposit to the purification plant

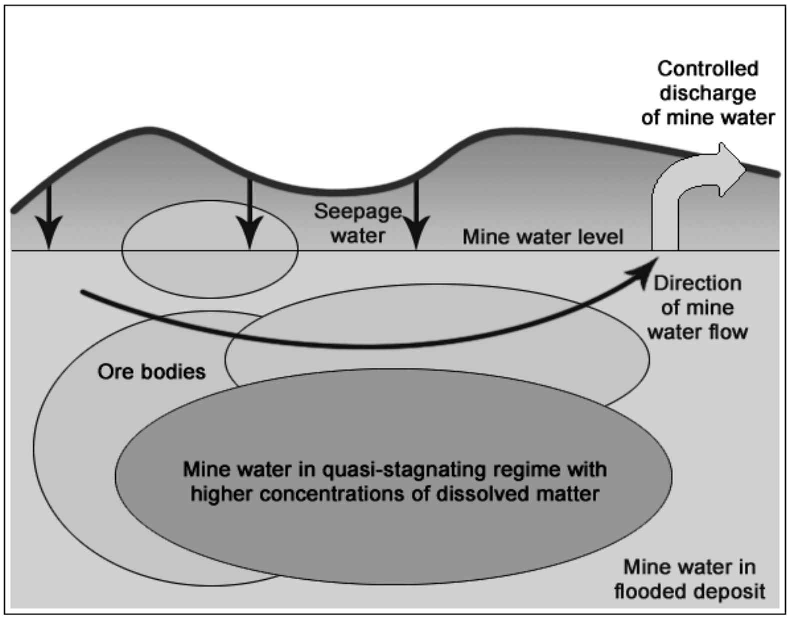

Figure 2. Diagram of expected distribution of mine waters in the flooded mine under closure

The verification of properties (chemical composition) of mine waters accumulated in abandoned mine workings after flooding the underground mine, represents a serious technical problem. Mine waters are impounded in the former mine and the single permanently accessible point to sample and analyze them is the point of controlled drainage of waters from the mine. However, at this point these waters are more or less waters of shallow circulation having other properties and composition in comparison with waters accumulated in deeper parts of the former mine.

One possibility for acquiring data on the composition of mine waters outside their shallow circulation is represented by shafts, provided that they have not been fully backfilled as part of the mine's decommissioning, and providing that they have remained accessible for the sampling of waters at various depths even after the mine has flooded. With regard to former uranium deposits, before now only some shafts in the Příbram deposit in Czech Republic have been accessible and suitable for taking the samples of waters. Here, the properties of mine waters have been observed in this way periodically since 2004 (Kalous et al., 2006).

The only possibility for obtaining a representative sample of mine waters impounded in the deeper parts of the former mine was a borehole drilled from the surface to a mine working in the central part of the deposit and outside the expected area of active drainage of the flooded mine. In the Czech Republic, such a hydrogeological borehole was drilled for the first time as part of the current research project in the Olší – Drahonín deposit (Michálek et al., 2007).

The research hydrogeological borehole passes through the complex of overlying rocks consisting of amphibolites and biotitic gneisses, and below level 3 of the former mine (about 160 meters below ground level) reaching earlier extracted vein structures. It passes outside mine workings that serve the active drainage of the former mine. Its mouth is located in a mine working on level 5, i.e. at a depth of 245 meters below ground level.

After completing the borehole, the pumping test was performed in two stages. The purpose of the pumping test was partly to verify the stability of inflows but mainly to determine the chemical composition of mine waters at specific levels, where inflows of mine waters into the borehole had been captured. The hydrogeological borehole, or more specifically the pumping test, has confirmed the assumption that an accumulation of mine waters with uranium content suitable for utilization as a secondary source of this raw material exists in deeper parts of the former mine.

After stabilization of the water regime, a uranium concentration of 17.5 mg/L was however determined in quasi-stagnant waters outside the zone of active draining.

The following aspects needed to be addressed in the next stage of research:

- What is the volume of waters in quasi-stagnant regime in the flooded mine?
- What is the “uranium potential” of quasi-stagnant waters, i.e. what is the amount of uranium that can be obtained from these waters?
- How will the pumping of quasi-stagnant waters influence both the chemical composition of shallow circulation waters drawn to the purification plant, and the hydrological regime of the deposit?

For resolving these questions, mathematical modelling and practical verification in the form of pilot scale test are used.

3. CONCEPTS AND RESULTS OF MATHEMATICAL MODELLING STUDY

Mathematical modelling should provide answers to two basic questions. First, whether there is a stagnating, non-flowing mine water area in the flooded mine or not and what is the amount and spatial layout of this water, and second, what is the theoretical uranium concentration in this mine water.

The task to be solved by this project is demanding. This is due to a high level of uncertainty following a minimal amount of calibration data, whether of levels and/or flow rates measured within the simulated structure. That is why it is necessary to base the model solution on a reliably calculated water balance of the deposit.

From the point of view of model inputs, we suppose that the deposit is merely fed by recharge. Therefore, it is essential to determine correctly the water balance of the partial river catchment area, i.e. the division of components in the hydrological balance – the direct runoff (surface runoff and overburden runoff), evapotranspiration and effective infiltration into the modelled structure.

The undisturbed rock mass usually has the interstitial or dual (interstitial-fissure) type of porosity. Commonly used models of groundwater flow do not enable the hydraulically correct simulation of dual porosity. As soon as the rock mass is developed underground, a system of mine workings creates free spaces with a karst type porosity and the flow subsequently has its own specifics following from the marked, secondarily produced, hydraulic inhomogeneity and anisotropy of the environment. The main problem in the simplification of this type of environment (anthropogenic pseudokarst) is to describe changes in the hydraulic properties of preferential flow paths (Rapantova et al., 2007).

The project at the former, already flooded, uranium mine at Olší – Drahonín required the application of a modelling code that could simulate double porosity flow as well as preferential flow along mine workings.

The FEFLOW code (Diersch, 2006) was selected as the best available software since the flexibility of finite elements mesh design to enable the geometrization of the uranium ore deposit to an acceptable level of simplification. In addition to 3D elements, it is possible to work with a combination of planar and linear elements applicable for simulation of fractures as well as vertical and horizontal mine workings. Within these elements, there is a choice of hydraulic calculations based on Darcy’s law for porous media, the Hagen-Poiseuille law for fracture flow, or the Manning-Strickler law for channel flow. The problem in conceptualisation and modelling of the mining environment consists of correctly describing and quantifying the hydraulic properties of preferential pathways. Depending on the site, one can decide to use either the Darcy or Manning-Strickler equations for mine workings. A three-dimensional model of mine workings was built for the Olší – Drahonín Mine using geographic information systems (GIS) – see figure 3.

A geological model of the area of interest was built as was a conceptual model of groundwater flow. The model domain was discretized into 10 numerical layers corresponding to mine levels. The planar and linear elements were used to simulate flow in fractures and mine workings based on real geometry of mine structure.

As stated earlier, we assumed that the deposit is recharged only from precipitation. Much of this water remains in shallow circulation, and drains into local streams, while only a part of the groundwater reaches the deeper parts of the deposit to flow along preferential pathways, mine workings and some fractures. The groundwater level in the deposit is kept at a specified level by pumping. In order to validate those two components of groundwater circulation, the hydrological balance of the partial watershed has to be assessed carefully.

The rainfall run-off model HEC-HMS was used for this purpose.

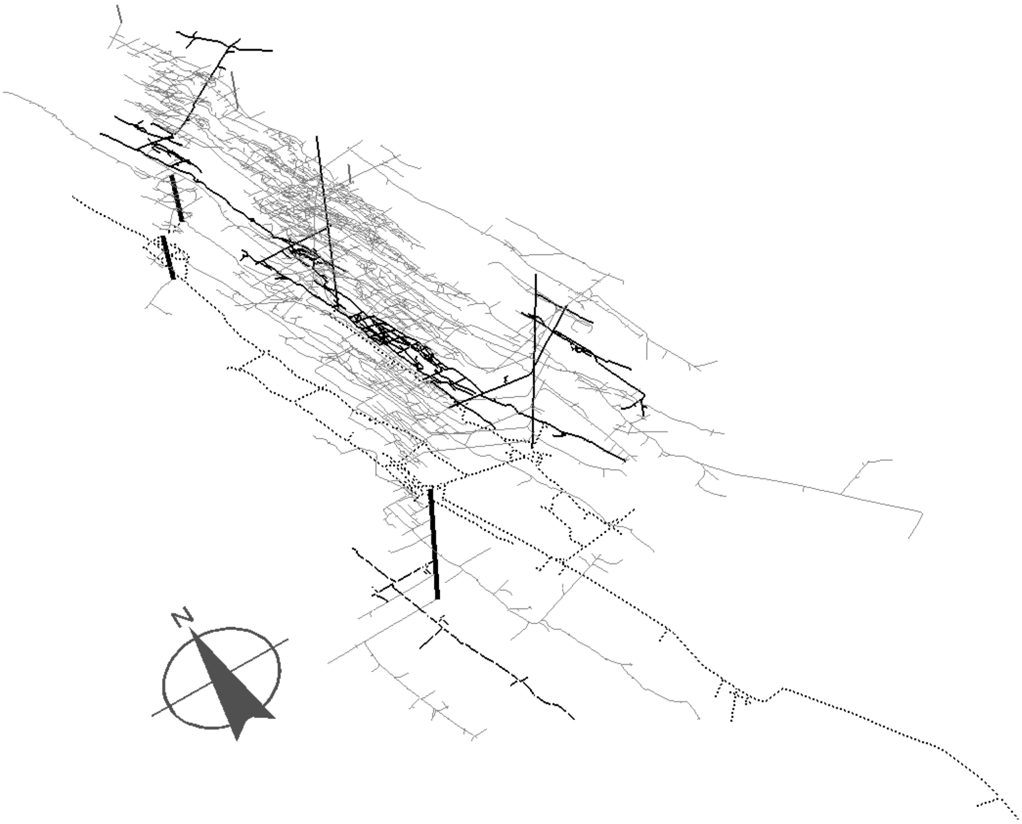

Figure 3. Mine workings scheme in ArcGIS – Olsi – Drahonin Mine

At this stage the reactive transport model of uranium dissolved in groundwater is under development. Thermohaline transport is taken into account due to temperature gradient observed in the mine. In order to assess the uranium mobility under present field conditions geochemical modeling was applied.

Complex modelling of geochemical evolution of mine water and thermodynamic modeling of particular component stability in groundwater and in water discharged from the mine was performed with the aid of Geochemist´s Workbench 6.0 software package (Bethke, 2005). Concentration of dissolved uranium in mine water is determined by redox potential and depending acidity (see figure 4). The mine water is saturated with respect to the uraninite UO_2 containing four valent uranium and other solid phases as U_3O_9 and U_3O_8. Natural uraninite is usually mixture of oxides binding four and more valent uranium as for example $U_3O_9 = 2\ U^{IV}O_2{\times}U^{V}{}_2O_5$ a $U_3O_8 = U^{IV}O_2{\times}U^{VI}{}_2O_6$. According to results of geochemical modelling the equilibrium between dissolved uranium and solid phases can be described by chemical equations

$$USiO_4 + 3\ HCO_3^- \leftrightarrow 3\ H^+ + 2\ e^- + UO_2(CO_3)_3^{4-} + SiO_2(aq)$$

$$U_4O_9(c) + 12\ HCO_3^- \leftrightarrow 10\ H^+ + H_2O + 6\ e^- + 4\ UO_2(CO_3)_3^{4-}$$

$$U_4O_9(c) + 8\ HCO_3^- \leftrightarrow 6\ H^+ + H_2O + 6\ e^- + 4\ UO_2(CO_3)_2^{2-}$$

This equilibrium is strongly dependent on redox conditions in ground water (compare conditions in water from borehole and mine water discharged from the mine).

The longterm evolution of uranium concentration in mine water and kinetic parameters were determined in Microsoft Excel® worksheet using Solver and least square criteria. For details see Zeman et al. ,2009.

a) b)

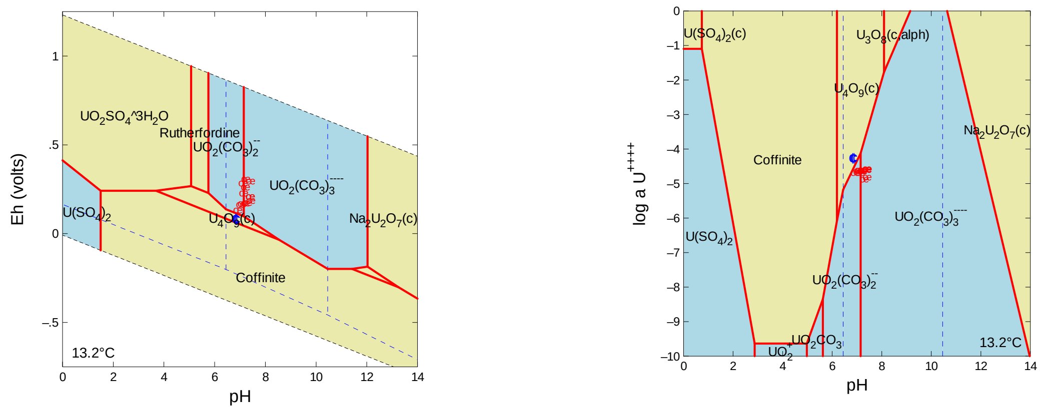

Figure 4. Redox-pH (a) and activity-pH (b) stability diagrams for uranium in mine water at 13.2 °C and component concentrations determined analytically. Blue stars – conditions in mine water from borehole at depth 170 and 210 m below surface, red circles – mine water discharged from mine at dewatering adit.

4. CONCLUSION

It can be assumed that the outputs of model solutions will support, at a given level of reliability, both the proposed engineering solution for the exploitation of uranium from mine waters, and the evaluation of the influence of uranium extraction from mine waters on the process and duration of water purification, especially for

- controlled geochemical influencing of mine waters with the aim of finding the depth level of the maximum (optimum for utilisation) uranium contents in mine waters,
- modifications in the existing engineering solutions for discharging mine waters with high contents of uranium or proposals for new, more effective solutions,
- modifications in mine water purification to achieve maximum yields of uranium whilst satisfying all limits determined to protect the environment (including the assessment of economic effects of project implementation).

To verify whether or not the utilization of quasi-stagnant mine waters as a secondary uranium source is viable, a pilot scale test is being prepared in the Olší – Drahonín deposit. As a site for pumping mine waters with a high content of uranium, the hydrogeological borehole will be used. From the pumped waters, uranium will be separated using the technology of sorption on the ion exchange resin. Because the proposed technology does not remove any other contaminants from mine water, the water will subsequently be injected at another point back into the deposit. The operation of active drainage of the flooded former mine through the purification plant will not be changed in any way. Ongoing changes in the chemistry of mine waters, i.e. both quasi-stagnant waters pumped from the borehole and those in waters of shallow circulation before entering the purification plant, will be continuously observed.

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