Utilization of Olsi-Dranonin uranium deposit after mine closure

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Abstract

In the course of the development and exploitation of particular uranium deposits, the quality of mine waters changes depending upon the extent of recharge area, total volume of worked-out mineral, its mineralogical composition and the achieved depth of mining. In the course of flooding the underground mine after closure many important changes occur in the content of dissolved substances in waters including significant increases in uranium, radium, iron, and others. Mine waters of flooded former uranium mines thus represent, with reference to their considerable volumes, a significant source of uranium. The paper presents the partial results of a running research project dealing with possibilities of the intensive utilisation of mine waters from flooded uranium mines as a source of uranium with an accompanying effect of shortening the time necessary for the purification of mine waters discharged into watercourses.

Keywords: uranium, mine closure, groundwater modelling

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 partial results of a project dealing with the possibilities of the intensive utilisation 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 utilising 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 utilised instead. If the present intention to utilise 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 tonnes of uranium). The subsequent reduction in the time necessary for mine water purification, which is financed by the Czech government, would also be significant.

An overview of the exploitation of individual deposits

Altogether about 109000 t of uranium has been mined in the Czech Republic since 1945. Six main mining areas have participated in this production; a small amount has also been extracted as part of the geological exploration in other regions. Periods of mining in individual areas and their proportion to overall production are given in Table 1. Uranium deposits in the Czech Republic (Figure 1) occur in a

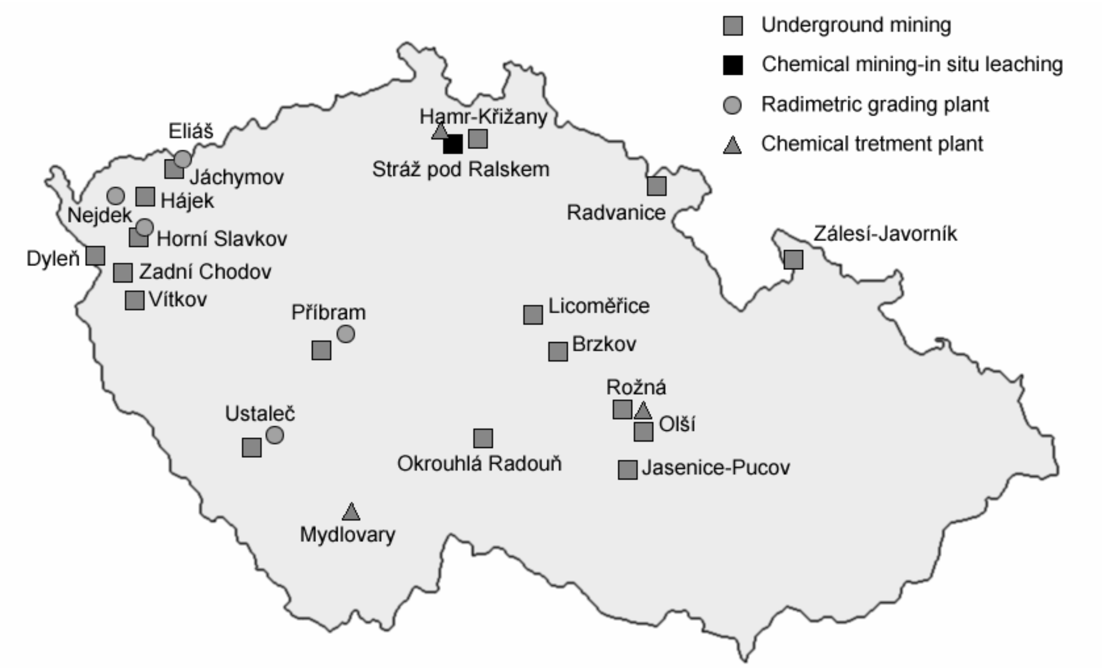
Variscan geological unit that is larger than the Czech Republic itself, namely in the Bohemian Massif which represents one of the greatest uranium-bearing provinces in Europe. Uranium mineralisation is represented here by endogenous and exogenous deposits: the endogenous deposits being confined predominantly to basement series and granitoid masses, the younger, exogenous deposits to Permian-Carboniferous, Cretaceous and Tertiary platform formations.

Table 1 *Mining areas with uranium deposits and an overview of their exploitation*

Mining area	Uranium deposit	Exploitation duration	Production
			% of total CR production
Příbram	Příbram	1949 - 1992	36.5
West Moravia	Rožná, Olší-Drahonín, Zálesí-Javorník, Chotěboř, Slavkovice-Petrovice, Radvance	1957 - to present (Rožná), exploitation ceases after 2010	19.2
North Bohemia	Hamr – Křižany	1974 - 1993	10.3
	Stráž pod Ralskem (leaching in situ)	1969 - 1996, since 1996 uranium acquired by chemical leaching as part of remediation	15.1
West Bohemia	Zadní Chodov, Vítkov, Dyleň, Okrouhlá Radouň, Hájek, Ruprechtov	1954 - 1992	9.0
Jáchymov	Jáchymov	1947 - 1962	6.2
Horní Slavkov	Horní Slavkov	1949 - 1963	2.2
Geological exploration	Jasenice-Pucov, Brzkov, Licoměřice, Ustaleč	1955 - 1990	1.5

The endogenous deposits are situated in the areas of Příbram, West Moravia, West Bohemia, Jáchymov and Horní Slavkov and are formed by highly dipping ore bodies of zone, vein and metasomatic types situated in compact rocks. The prevailing thickness of ore mineralisation ranges from 1.5 to 2.0 m, and less frequently up to 10.0 m. The depth range of mining was usually from the surface to a depth of 600-700 m. The deposits of Zadní Chodov (1250 m) and above all Příbram (1550 m) were mined at great depth and the deposit of Rožná (1200 m) is still being exploited.
Exogenous deposits are found in the Cretaceous sediments of North Bohemia. They are represented by occurrences of uranium mineralisation in sandstones and siltstones. Here we find subhorizontally laid ore bodies of polygenous type (infiltration and hydrothermal) and great thickness at a depth of about 250 metres. Stráž pod Ralskem and Hamr-Křižany belong to such deposits that were exploited in the Czech Republic. The deposit of Hamr-Křižany was exploited by underground mining with backfilling the worked-out stopes with consolidated material and the deposit of Stráž pod Ralskem was exploited by the acidic leaching of uranium from the ore directly in ore bodies (in situ leaching).

Figure 1 Main mining and treatment capacities of uranium mining in the Czech Republic



Present state of selected mining sites after their closure

When 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 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.

The aforementioned research task, "Non-traditional utilisation of uranium deposits after underground mining completion" concerns the possibilities for intensive utilisation of the mine waters of flooded uranium mines in endogenous type deposits.

For the detailed researches into the mine waters of flooded uranium mines, 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 is available for project use. Basic research is therefore being carried out in this locality.

Hydrogeological conditions of the Olší - Drahonín deposit

For solving the research task, the Olší-Drahonín deposit was selected as a pilot locality. In the locality, the hydrogeochemical regime of mine waters, the progress of mining operations and mine drainage, the geological structure of the deposit, the hydrology and hydrogeology of the area and mine are all documented very well. The Olší-Drahonín deposit was exploited from 1959–1989. At the time exploitation of the deposit ended, mining operations were carried out at depths below the 10th level (+18 m above sea level, i.e. 467 m below ground level) and the deposit was developed by blind shafts down to the 18th level (–374 m below sea level, i.e. 859 m below ground level).

Basic data on the Olší - Drahonín deposit

- Spontaneous flooding of the underground mine in the Olší-Drahonín deposit began in April of 1989 after exploitation ended. The volume of accumulated mine waters underground is about $2.3 \cdot 10^6$ m^3.
- For the controlled use of mine waters (discharge of excess waters from the flooded mine) a drainage adit was driven and at its mouth, a mine water purification plant was built.
- In January 1996, mine waters had risen to the level of 451.3 m above sea level (floor of the drainage adit).
- Since 1997, the mine water level has been maintained by pumping at 1.5 – 7.0 m below the drainage base (floor of the drainage adit), i.e. at a level of 449.8 to 444.3 m above sea level, and this ensures that contaminated mine waters will not outflow spontaneously and uncontrolled into the surrounding environment. The overall amount of water purified in 2006 was 245073 m^3, i.e. the average rate of discharge from the mine was about 7.7 $l \cdot s^{-1}$.
- In the course of mining operations, the maximum pumping rate was recorded in the years 1981 and 1986 and amounted to about 17 $l \cdot s^{-1}$.
- The chemistry of mine waters changed during the exploitation of the deposit so that in the course of mining development (increase in the volume of stoped-out workings), the initial concentrations of substances in water decreased. This effect is caused by the creation of preferential pathways between surface water and underground mine workings due to lowering the mine water level in the centre of depression, and thus increasing the rate of groundwater flow to the point of pumping. Erosion of the weathered zone of deposit does not occur during the creation of preferential pathways..
- From the moment when the pumping of mine waters stopped, backfilled and mining-affected parts of the deposit were flooded. Solutions were enriched with readily soluble compounds which had been earlier exposed to oxidation in the centre of the deposit.
- The purification of mine waters discharged by the drainage adit from the deposit has been carried out since 1996 (Figure 3).

Mine waters of the Olší - Drahonín deposit

In the period of mining the Olší-Drahonín deposit, mine waters were mixed waters from the following sources:

- surface waters infiltrating from atmospheric precipitation, to a limited degree also from surface watercourses, infiltrating primarily along priority paths into mine workings at the uppermost levels of the mine,
- waters from natural sources – mostly waters with limited water exchange with the earth surface, passing through fault-joint systems into mine workings as a result of anthropogenic actions (artificial lowering of drainage base by mine working drainage),
- waters for industrial use – are proportionally minimal in the mine waters of the Olší-Drahonín deposit – usually non-drinkable fresh water,
- other sources of mine water (technological liquids, and others).

Monitoring the quality of mine waters at the Olší-Drahonín deposit was prescribed by mining regulations and during mining operation, sampling points were situated in the shafts Olší (O–1) and Drahonín (O–4). The waters could be described as mixed waters of Ca (Mg) - SO_4 - HCO_3 type with a temperature of about 8 to 10°C (depending upon the season, distance from the shaft and depth below the surface). At present, mine water is monitored before entry into the purification plant and after purification, i.e. water discharged into the watercourse. The changes in the chemical composition of mine waters of the deposit after flooding are documented in Tables 2 and 3. However, these changes concern the upper part of the aquifer , not the whole deposit.

Table 2 *Chemical composition of mine waters in the period of exploitation of the Olší – Drahonín deposit (by 1989)*

		Shaft Olší (O-1)	Shaft Drahonín (O-4)
pH		6.50 – 6.90	6.50 – 6.80
Residue on evaporation 105 °C	$mg \cdot l^{-1}$	750.00 – 960.00	740.00 – 1050.00
Ca^{2+}	$mg \cdot l^{-1}$	29.50 – 169.60	22.40 – 226.40
Mg^{2+}	$mg \cdot l^{-1}$	18.40 – 78.20	26.50 – 96.60
Fe^{2+}	$mg \cdot l^{-1}$	2.20 – 4.80	0.20 – 3.00
Cl^{-}	$mg \cdot l^{-1}$	18.70 – 46.30	15.90 – 36.50
SO_4^{2-}	$mg \cdot l^{-1}$	338.20 – 1152.00	126.70 – 1388.40
NO_3^{-}	$mg \cdot l^{-1}$	1.960 – 92.50	1.96 – 105.30
CO_3^{2-}	$mg \cdot l^{-1}$	38.60 – 114.00	49.10 – 110.20
HCO_3^{-}	$mg \cdot l^{-1}$	115.90 – 170.90	128.10 – 347.80
O_2	$mg \cdot l^{-1}$	5.30 – 11.80	5.20 – 12.20
U	$mg \cdot l^{-1}$	0 – 4.46	0 – 2.45
Ra^{226}	$Bq \cdot m^{-3}$	0 – 1150.00	0 – 860.00

Table 3 *Chemical composition of mine waters from the flooded Olší - Drahonín deposit entering the purification plant between 1996 – 2006 (*results from merely one sample)*

		Entry to purification plant		Exit from purification plant
		Min – max	Average	Average
pH		6.10 – 7.52	6.92	7.64
Ca^{2+}	$mg \cdot l^{-1}$	350.00*	-	not observed
Mn^{2+}	$mg \cdot l^{-1}$	1.43 – 8.71	4.55	1.00
Fe^{2+}	$mg \cdot l^{-1}$	2.33 – 18.85	9.79	0.20
Cl^{-}	$mg \cdot l^{-1}$	27.00*	-	not observed
SO_4^{2-}	$mg \cdot l^{-1}$	871.10 – 1843.90	1390.64	1100.00
NO_3^{-}	$mg \cdot l^{-1}$	1.00 – 12.00	4.37	not observed
HCO_3^{-}	$mg \cdot l^{-1}$	560.00*	-	not observed
U	$mg \cdot l^{-1}$	5.26 – 13.10	9.20	0.05
Ra^{226}	$Bq \cdot m^{-3}$	110.00 – 2700.00	1225.10	80.00

Changes in mine water chemistry

The conceptual model of the project is based on the fact that the chemistry of mine waters changes markedly in the course of the development, exploitation and subsequent abandonment of the deposit after mine decommissioning. This is a result of the following several factors:

- a change in the water sources for the equation of mine water balance (Grmela and Rapantová, 2006):

    - deposit development:

      $$Q_{pump.} = \boldsymbol{Q_{operation}} + \boldsymbol{Q_{natural\ sources}} - Q_{vent.} \pm Q_{loss} \tag{1}$$

    - deposit exploitation:

      $$Q_{pump.} = \boldsymbol{Q_{operation}} + \boldsymbol{Q_{natural\ sources}} - \boldsymbol{Q_{vent.}} - Q_{exploitation} \pm Q_{loss} \pm Q_{accumulation} \tag{2}$$

    - abandoned deposit:

      $$Q_{\ outflow\ (pump.)} = \boldsymbol{Q_{natural\ sources}} \pm Q_{loss} \tag{3}$$

The significant water balance components are given in bold. Flux from the natural sources is variable and is a function of hydraulic gradient to the level of the drainage base in the given period. A non-negligible proportion of anthropogenic substances affecting mainly the qualitative parameters of mine waters is not presented.

- a change in the physical-geochemical and hydrogeochemical regime over the course of mine development (Grmela and Rapantová, 2006):

    - changes induced by the gradual changes of water fluxes from natural sources as the mine develops in the mining claim area and also as the mine is deepened,
    - changes caused by the opening of various rock environments (with various mineral compositions and various reactivities of minerals to emerging oxidation processes), changes caused by reactions of hydrogeochemical solutions in the mine, effects of natural and industrial waters on the rock environment, and others,
    - the influence of mine atmosphere on various kinds of rocks and mine waters.

- changes in the physical-chemical regime after mine decommissioning:

    - changes induced by the inaccessibility of air and changes in the composition of residual mine atmosphere,
    - changes in the composition of mine waters in connection with the abandonment of the source of so-called service waters and the decreasing of proportion (even disappearance) of some natural sources,
    - lowering the oxygen balance over time and with depth,
    - changes connected with the minimization of previously induced and maintained flow of mine waters through mine workings,
    - the transfer of substances (evaporites) from "dry" mine workings into solution after flooding the mine workings.

The problems being solved are significantly connected with and simultaneously utilise the last mentioned factor. After decommissioning the mine, the concentration of uranium (but not only of uranium) significantly increases in waters as the mine is flooded. The development of a marked vertical stratification of mine waters follows. This is especially conditioned by the minimization of mine water flow after the disappearance of the hydraulic gradient originally induced by the active dewatering and the following flooding of the mine. These regularities are common to almost all underground ore and coal mines (Kalous et al., 2004, Rapantová and Grmela, 2004). As an example of the situation in the locality, two graphs (Figure 2 and 3) are presented (Michálek et al., 2006).

Figure 2 *Comparison of uranium contents in mine waters of the flooded Olší - Drahonín deposit and the deposit of Rožná in operation*

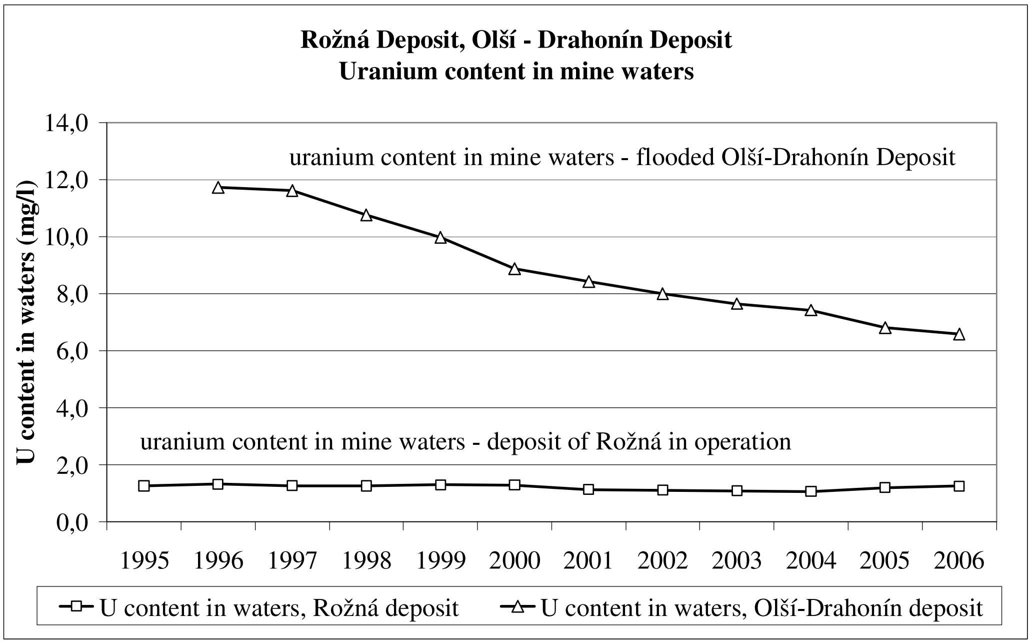


Figure 2 shows the content of uranium in mine waters from the Rožná deposit (in operation) and the Olší-Drahonín deposit (finished mining operations; flooded), from which are evident both a low content of uranium in mine waters in mines with an active drainage regime and an increased content in structures of quasi-stagnating geohydrodynamical systems. In the case of the Olší-Drahonín deposit, the leaching/flushing of the uppermost (subsurface) water-bearing layer of flooded old mine workings due to water pumping at the mine water purification plant can be seen well. This is a case of two adjacent uranium deposits with the same geological structure and mineralisation (Figure 2).
The yield of uranium from mine waters in the Olší-Drahonín deposit in relation to the amount of pumped mine waters and mine water quality (concentration of uranium) can be seen on the Figure 3.

Figure 3 *Course of purification of mine waters discharged from the flooded Olší - Drahonín deposit and U yield at water purification*

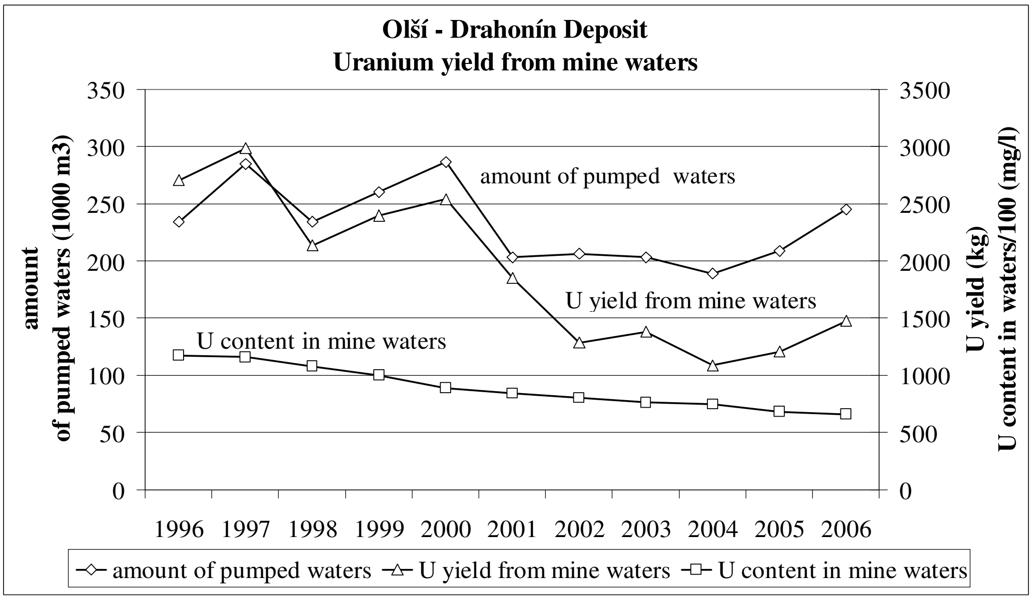
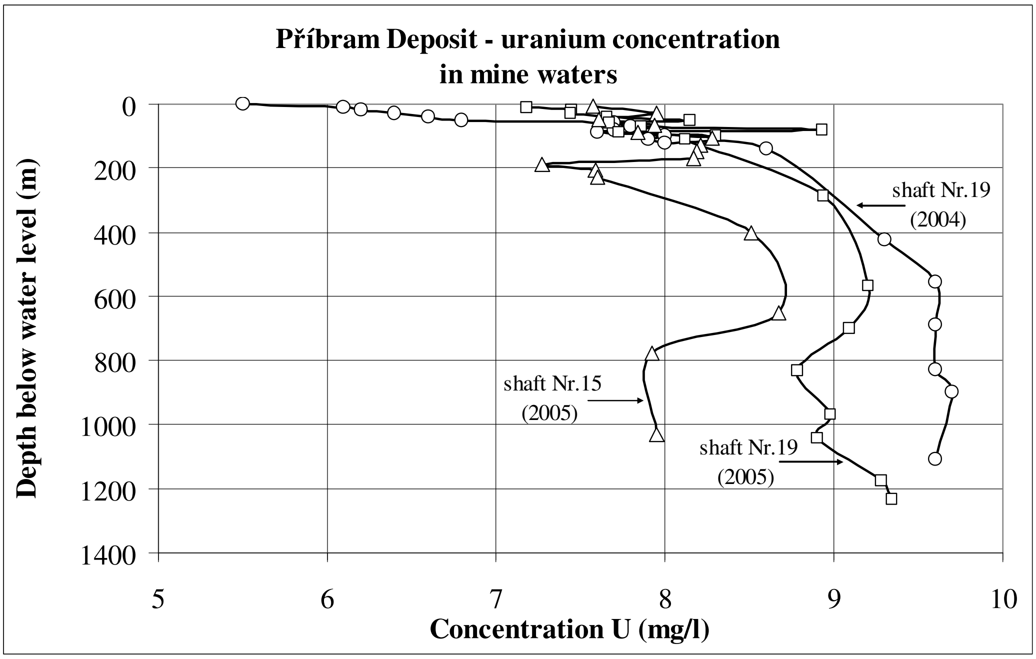


***Figure 4** Uranium concentration in mine waters of the flooded Příbram deposit relative to depth*



In the graphs shown, the downward trend in the content of uranium in mine waters of the flooded Olší-Drahonín deposit is completely logical, because it represents a case of draining the deposit merely in the upper part of aquifer (shallow circulation), where a proportion of infiltrated waters from atmospheric precipitation (or recharge from surface watercourses) markedly manifest themselves. Therefore, this is not a representative trend for the content of uranium in the overall accumulated water body in abandoned mine workings where. the representative trend would logically be an upward trend. As part of the problem in question, only deeper water circulation is of interest. In the deeper sections of the former mine, uranium concentrations are significantly higher than in the shallow parts. From the point of view of discharged water purification, a clear requirement for the discharge of shallow waters exists, i.e. waters with minimum contents of dissolved substances. On the other hand, from the point of view of maximization of the yield of uranium from mine waters, it would be desirable to discharge waters with high uranium concentrations, i.e. from the deeper parts of the mine. Furthermore, it is evident from the graphs that it is above all the dynamics of mine water discharge that has, in addition to the sources of mine waters, the most substantial influence on the hydrological regime of the deposit. In a quasi-stagnating regime, an increase in total mineralisation occurs and vertical hydrogeochemical stratification in the flooded mine emerges (changes in pH being indistinct). The stratification of mine waters has been very well recorded in the deposit of Příbram (Kalous et al., 2004, Hájek et al., 2006, Lusk, 2006). The example is shown in the graph given on the Figure 4.

Mathematical modelling of the groundwater flow and chemical changes in mine waters

One of the approaches needed to accomplish the project objectives is the mathematical modelling of both the mine water flow in the flooded Olší-Drahonín deposit and the transport of substances in mine waters. 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 mathematical model is being prepared by collecting the necessary data and by verifying suitable software for this purpose.

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. This is in the preparation stage. 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.
In the Czech Republic, mathematical modelling has already been used for the simulation of the effects of drainage of underground mines in the Ostrava-Karviná Coalfield or applications of modelling for the optimization of drainage of opencast mines. In contrast to existing applications, in order to accomplish the objectives of this project, the application of reactive transport will be required. To determine the time-spatial dynamics of the solution of uranium in mine waters will require the use of software tools, dealing with the hydrochemical processes under various physical-chemical conditions. In this case however, these calculations are also to be used speculatively, because in the majority of cases (unlike laboratory conditions) some necessary data from the natural environment of flooded old mine workings are unknown (above all oxidation potential, pH and Eh).
In spite of the above arguments that point to solution uncertainties, it is necessary to state that the role of mathematical modelling in solving complicated mine-hydrogeological problems is indispensable. The utilisation of mathematical modelling in real scenarios of structure, boundary conditions, recharge and drainage areas is valuable in the creation and verification of conceptual models. Mathematical modelling is also the only means for evaluating the consequences of hydraulic actions and their interference on the level of the reliability of these applications.

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).

Acknowledment

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