The Use and Treatment of Mine Waters from Closed and Flooded Uranium Mines

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ABSTRACT

After the decommissioning of underground uranium mines has finished, it is necessary to ensure the safe use of mine waters accumulated in flooded mines, because they contain uranium, radium and other contaminants in high concentrations and may thus, in the case of uncontrollable release from flooded mines, endanger their surrounding environment. On the other hand, with regard to their considerable volume, these mine waters represent a secondary source of uranium. In this paper the authors give an overview of uranium mining in the Czech Republic and the present-day state of underground uranium mine decommissioning. They report on the use of mine waters after mining operations in particular localities have finished and on methods of mine water treatment. They also deal with the possibilities of producing uranium as a secondary product in the course of mine water treatment.

Additional Key Words: uranium concentration, quasi-stagnant regime

INTRODUCTION

From 1945 to the mid 1990's, uranium mining was an important industry in the Czech Republic, and the Czech Republic occupied a prominent global position in the production of uranium concentrate. A marked phase-out of the industry in the first half of the 1990's, especially due to a marked reduction in possibilities of sales as a result of the economic-political changes taking place at the turn of the 90's of the last century, led to the closure of many underground uranium mines. At present they are closed and flooded (Michálek et al., 2005). Mining operations are only performed in one underground mine in the Rožná deposit, and are expected to finish after the year 2012.

In the recent past, the vast majority of underground uranium mines in the Czech Republic were decommissioned by naturally flooding the mines after underground mining operations and the necessary preparations in connection with mine decommissioning were complete. Mine waters that exceed the pre-determined level (henceforth referred to as overbalance waters) are at present drained from the flooded mines under control (by pumping or gravitationally). This mine water level is determined for each deposit by considering the terrain morphology in the surroundings and is crucial for avoiding the uncontrollable release of contaminated mine waters that could endanger the ambient environment. The absolutely fundamental task of uranium mining in relation to the environment, both in the course of mining the deposit and subsequently during mine decommissioning and mitigating the impact of mining, is to minimize the negative effects of released radionuclides on the environment and health of the population.

In the course of developing and exploiting individual uranium deposits, the chemistry of mine water changes depending upon the extent of the drawdown area surrounding the mine, the total surface area of exposed mine workings, the mineralogical composition of the rock environment (including residues of unmined uranium and other radioactive minerals in these mine workings), and also the depth of mining. During the flooding of underground mines as part of decommissioning, other significant changes occur in the content of dissolved matter in the waters. This involves a several-fold increase in such content (uranium, radium, iron, and others) due to the previous oxidation of minerals when the deposit was being exploited. With reference to the high concentrations of dissolved matter in mine water (above the limits determined for discharge), it is necessary to treat the overbalance water drained from the flooded mines and to retain contaminants before discharging to surface water. Nevertheless, owing to their considerable volumes, the mine waters from flooded uranium mines represent a secondary source of uranium that is only partially utilised in the Czech Republic.

URANIUM DEPOSITS AND THEIR EXPLOITATION

Altogether about 110 000 t of uranium have 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 geological exploration in other regions. Uranium deposits in the Czech Republic (Fig. 1) occur in a geological system that is larger than the Czech Republic itself, namely in the Bohemian Massif, which represents a denudation remnant of the Variscan mountains and 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, and the younger, exogenous deposits confined to Permian-Carboniferous, Cretaceous and Tertiary platform formations.

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

The endogenous deposits are situated in the areas of Příbram, West Moravia and West Bohemia 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 in places of uranium mineralisation by sandstones and siltstones. Here we find subhorizontally laid ore bodies of great thickness at a depth of about 250 metres. Stráž and Hamr are two such deposits that were exploited in the Czech Republic. The deposit of Hamr was exploited by underground mining with backfilling the worked-out stopes with a consolidated material, and the deposit of Stráž was exploited by the acidic leaching of uranium from the ore directly in ore bodies (in situ leaching). With regard to the specific hydrogeological conditions and their different methods of decommissioning, this contribution below will not deal with the deposits of Hamr and Stráž.

THE TREATMENT OF MINE WATERS

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 basin area and the hydrogeological conditions of the deposit, this process took up to several years (Hájek et al., 2006). During this time, conditions for proper mine water management in the "collection – controlled draining from underground spaces – treatment – discharge" mode had to be created in advance. Only this would ensure that shallow groundwater as well as surface water would not be threatened by uncontrollable leakage of contaminated water from the flooded mine in future.

Among the most significant uranium deposits exploited by underground mining in the Czech republic are the following:

- **Deposit of Okrouhlá Radouň**

Altogether 1 340 t of uranium were exploited; flooding began in the year 1990; waters rose to their permited level in 1993; about 50 000 m^3 of mine waters is drained annually from underground cavities; the total volume of waters in the deposit is about 1.2 million m^3.

- **Deposit of Vítkov**

Altogether 3 970 t of uranium were exploited; flooding began in the year 1990, waters rose to their permited level in 1994; about 50 000 m^3 is drained from underground cavities annually; the total volume of waters in the deposit is about 2.5 million m^3.

- **Deposit of Zadní Chodov**

Altogether 4 150 t of uranium were exploited; flooding began in the year 1993; waters rose to their permited level in 1995; about 420 000 m^3 is drained from underground cavities annually; the total volume of waters in the deposit is 3.1 million m^3.

- **Deposit of Olší**

Altogether 2 920 t of uranium were exploited; flooding began in the year 1989; waters rose to their permited level in 1996; about 240 000 m^3 is drained from underground cavities annually; the total volume of waters in the deposit is about 2.3 million m^3.

- **Deposit of Příbram**

Altogether 50 990 t of uranium were exploited; flooding began in the year 1990; waters rose to their permited level at the end of 2005; about 2 300 000 m^3 is drained annually from underground cavities through two points of abstraction; total volume of waters in the deposit is about 23 million m^3.

In the overwhelming majority of cases the process of mine flooding was left to take its natural course. After flooding the underground cavities and achieving a certain level, mine water can discharge to surface water and shallow aquifers. From the environmental point of view, it is thus necessary to maintain the mine water level below this altitude (by pumping) and to treat pumped waters, i.e. to remove those contaminants from mine waters which exceed levels determined by the relevant water authorities for discharging.

From the hydrological point of view, the Czech Republic occupies a significant position in Europe, because there are watersheds of three 1st order European watercourses in its territory: the Elbe (discharge to North Sea), the Oder (Baltic Sea) and the Danube (Black Sea). The protection of surface water quality is thus subject to international agreements. The mine waters from uranium deposits produced in the Czech Republic provide significant loads to the Elbe River Basin and the Danube River Basin (Table 1).

Table 1. Discharge of mine waters in the Czech Republic (in thousand m^3 annually) – Grmela, Rapantová (2005).

		Elbe watershed	Oder watershed	Danube watershed	total
Total - Czech Republic 1975		122000	31000	5000	158000
1998	non-metallic deposits	5652	953	6390	12995
1998	**radioactive materials**	**19219**	**185**	**1906**	**21310**
1998	crude oil			83	83
1998	ores	1740	2838	19	4597
1998	coal-underground exploitation	20739	16849	1932	39520
1998	coal-open-pit exploitation	17750			17750
1998	Utilization of mine water	drinking water management			6614
1998	Utilization of mine water	industry			4680
1998	Utilization of mine water	agriculture			500
1998	Utilization of mine water	balneology			6
1998	Total - Czech Republic 1998	65100	20825	10330	96255
Total - Czech Republic 2004		45024	8891	8379	62294

The basic technology for the treatment of mine waters drained uranium mines is based on uranium sorption on ion exchange resin and the simultaneous precipitation of radium contained in mine waters (simplified diagram in Fig. 2).

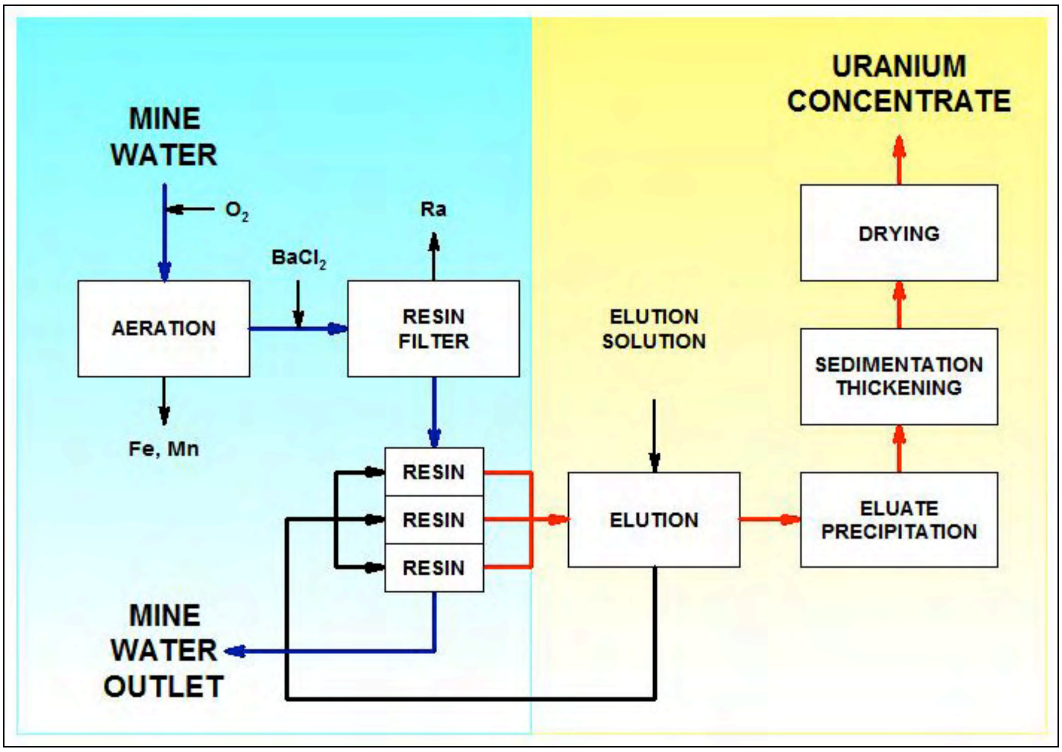

Figure 2. Diagram of treatment technology for mine waters with uranium and radium.

This technology for mine water treatment makes it possible to obtain uranium or uranium concentrate from mine waters as a secondary product of water treatment even long after the classical exploitation of the deposit has been completed (see example of the Olší deposit, Table 2 and Fig. 3). The duration of the mine water treatment process will vary from locality to locality; still, in all cases it will be a matter of decades. The duration of treatment depends on overall water inflow into the mine, the range of water flows in the former mine, the mineralogical composition of rocks in the deposit, and last but not least, on the conditions determined for water discharging into public watercourses, which depend in part on the volume of through-flow in the given watercourse. With regards to the above-mentioned uranium deposits, mine water treatment has already been completed in the localities of Okrouhlá Radouň, Vítkov and Zadní Chodov. Here, the treatment of drained mine waters was carried out for about 13 to 15 years and, at present, it is possible to discharge waters to the watercourse directly, through natural wetlands. As to the process of mine water treatment in other localities, i.e. Olší and above all Příbram, it is supposed that concentrations of dissolved matter in the upper part of the aquifer will reach permissible values for directly discharging mine waters after 30 years at the earliest.

Olší Deposit

The deposit of Olší was mined between 1959 and 1989. At the time when exploitation of the deposit 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 deposit were characterised by their irregular shape, variable thickness and uranium content. Small and medium-sized bodies were predominant and were developed vertically for two or three levels. With depth, the size of ore bodies diminished, as did the mineralisation coefficient. The richest ore bodies were localised on levels 2 to 5 of the deposit. The spontaneous flooding of the underground mine in the Olší deposit began in April 1989 after its exploitation had ended. The volume of mine waters accumulated underground is about 2.3 million m^3. Since 1996, the treatment of excess mine waters pumped and drained through a drainage adit from the deposit has taken place. The treated mine waters have been discharged into a surface stream (Hadůvka creek). The quantity of mine waters treated annually in the deposit of Olší amounts to 200 000 – 280 000 m^3. Altogether 23.3 t of uranium was obtained through the process of mine water treatment (from the flooding of the mine until December 2008); see Table 2.

Table 2. Uranium yield from the treatment of mine waters from the Olší deposit in the period from January 1996 to December 2008.

period	U concentration in inlet water	U yield
	$mg \cdot l^{-1}$	t
1996 - 2000	10.6	13.2
2001 - 2005	7.7	6.3
2006 - 2008	6.1	3.8
total		**23.3**

A declining trend in the uranium concentration of mine waters drained through the drainage adit from the flooded Olší deposit (Figure 3) is completely logical because, in the upper part of the aquifer, (the so-called shallow circulation of mine waters), formed by underground cavities developed as a result of the deposit exploitation, there is a significantly higher proportion of waters infiltrated from atmospheric precipitation or infiltrated from surface water courses. Thus this trend is not representative of the uranium content in the accumulated waters in abandoned mine workings as a whole. The extent of shallow circulation depends on hydrogeological conditions and the method by which the deposit was developed, as well as the flow of waters induced by controlled drainage of mine waters (either by pumping or gravitationally). In contrast, quasi-stagnant waters are impounded in the mine, almost without movement, and the concentration of dissolved matter is markedly higher than in shallow circulation waters. Once the new hydrological regime of the deposit has been stabilised, a distinct vertical stratification of mine waters develops, i.e. one that is especially governed by the minimisation of mine water flow after cessation of the hydraulic gradient induced earlier by actively draining the mine. These principles are evident in practically all ore and also coal mines (Rapantová and Grmela, 2004).

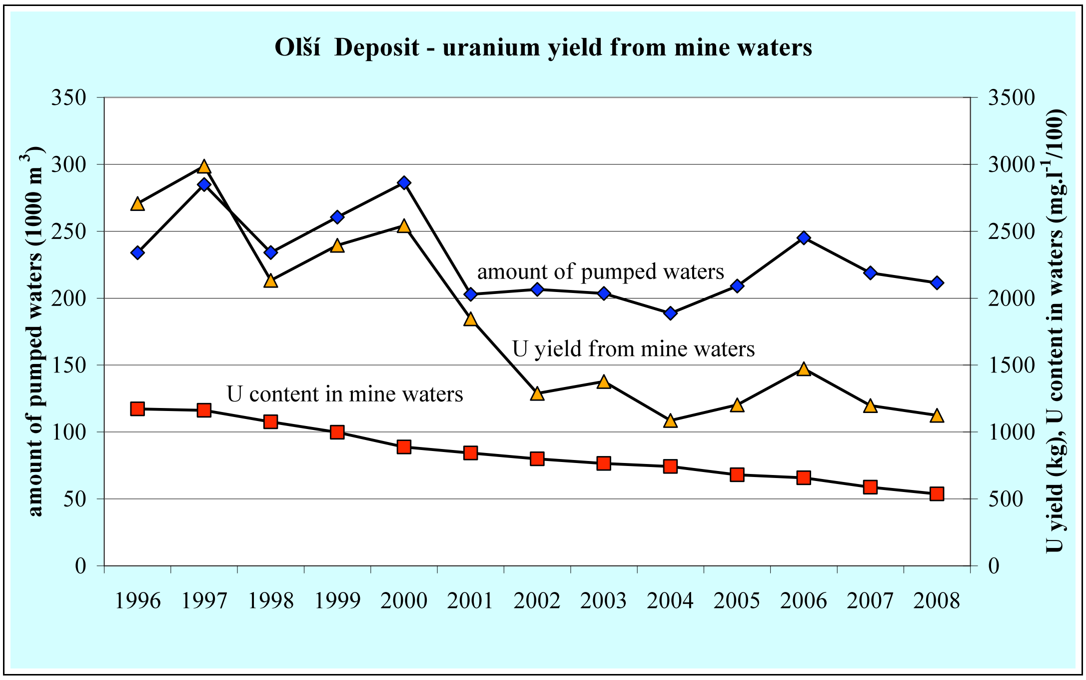

Figure 3. Amount of mine waters and uranium concentration in mine waters drained from the flooded Olší deposit to the treatment plant, and the uranium yield from mine waters.

Příbram Deposit

From the deposit of Příbram, which was the largest exploited uranium deposit in the Czech Republic, mine water has been drained under controlled conditions, treated and discharged into the Kocába River since the flooding of the deposit in October 2005. The amount of water drained in this way is determined at any particular moment by the volume of seepage from precipitation and surface water into the underground mine cavities. The draining of overbalance mine waters is carried out at two points through shafts Nos. 19 and 11A, which have not yet been decommissioned by backfilling. The pumped water is piped to the mine water treatment plants ČDV Příbram I and ČDV Příbram II, where contaminants are removed.

The above-mentioned technology of sorption on ion exchangers is used for the collection of uranium. This enables the production of uranium concentrate as a by-product of treatment (Table 3). With regard to the pumped and treated volumes of mine waters (2 200 000 to 2 400 000 m^3 annually, of which ČDV Příbram II purifies up to 1 700 000 m^3 annually), the production of uranium is markedly higher than in the case of mine waters from Olší deposit.

Table 3. Uranium yield from the treatment of mine waters of the Příbram deposit in the period from November 2005 to December 2008.

year	**ČDV Příbram I**		**ČDV Příbram II**		**Total**
	U concentration in inlet water	U yield	U concentration in inlet water	U yield	U yield
	$mg \cdot l^{-1}$	t	$mg \cdot l^{-1}$	t	t
2005	2,3	0,1	8,5	1,7	1,8
2006	3,3	0,6	8,3	14,2	14,8
2007	4,6	2,1	7,3	11,0	13,1
2008	7,5	3,4	7,4	8,4	11,8
total		**3,2**		**35,3**	**41,5**

The by no means negligible amount of uranium obtained as a by-product of mine water treatment markedly improves the economics of operating treatment plants. However, from the point of view of maximizing the uranium yield, it would be desirable to drain mine waters accumulated in the deeper parts of the former mine because they have significantly higher uranium concentrations.

MINE WATERS IN THE DEEPER PARTS OF FORMER MINES

One possibility for acquiring data on the composition of mine waters beyond their shallow circulation is represented by shafts, provided they have not been fully backfilled as part of mine decommissioning, and providing they have remained accessible for sampling of waters at various depths even after the mine was flooded. With regard to former uranium deposits, before now only some shafts in the Příbram deposit have been accessible and suitable for taking samples of waters. Here, the properties of mine waters have been observed in this way periodically since 2004 (Kalous et al., 2006), see Figure 4.

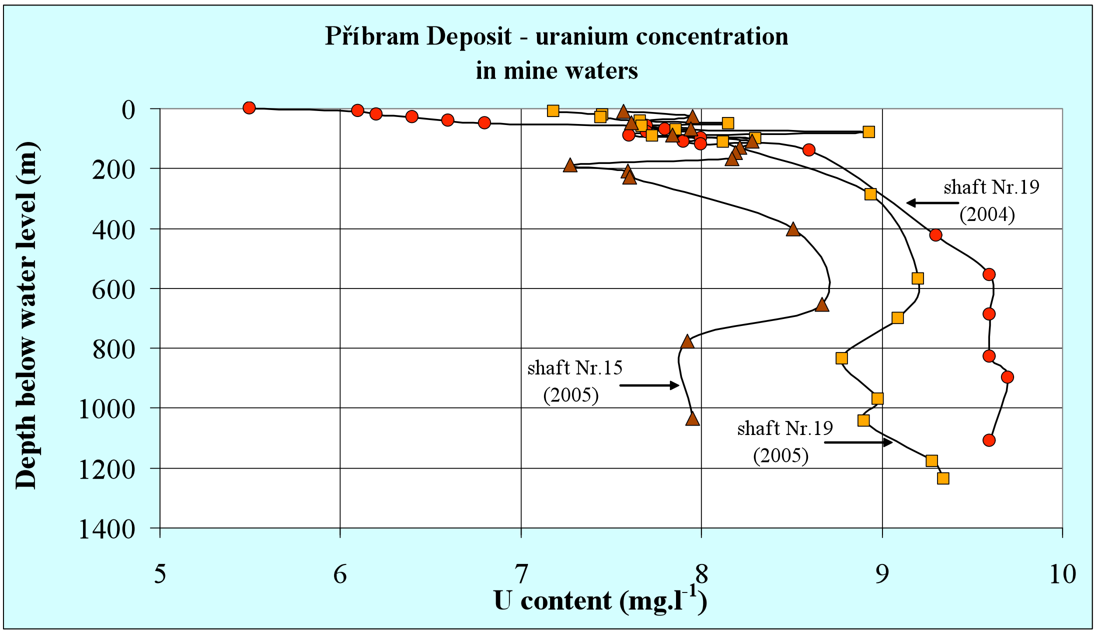

Figure 4. Uranium concentration in mine waters of the flooded Příbram deposit relative to depth.

But results obtained in this way still do not necessarily represent the typical characteristics of mine waters for the given deposit. Shafts are usually situated in the underlying layers of the deposit and not in parts of the deposit exposed to intensive mining, i.e. outside those areas where, even after previous exploitation, non-mined parts of ore bodies have remained exposed to oxidation. Instead they primarily represent a drainage system through which surface and near-surface waters are brought into the former mine, causing a change in the properties of mine waters accumulated in the shafts and their near surroundings.

The only possibility for obtaining a representative sample of mine waters impounded in the deeper parts of a former mine is 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. Such a hydrogeological borehole was drilled in the Czech Republic for the first time as part of a research project in the Olší deposit (Michálek et al., 2007). The hydrogeological borehole for research purposes passes through the complex of overlying rocks consisting of amphibolites and biotitic gneisses, and below level 3 of the former mine (about 160 metres below ground level) reaching earlier extracted vein structures. It passes beyond mine workings that serve to actively drain the former mine. Its mouth is located in a mine working on level 5, i.e. at a depth of 245 metres below ground level.

To document changes in the chemical composition of mine waters in the closed mine, a comparison can be made of the concentrations of dissolved substances at particular stages of the mine's existence or also, as the case may be, from various parts of the mine. A comparison of selected ions, is shown in Figure 5.

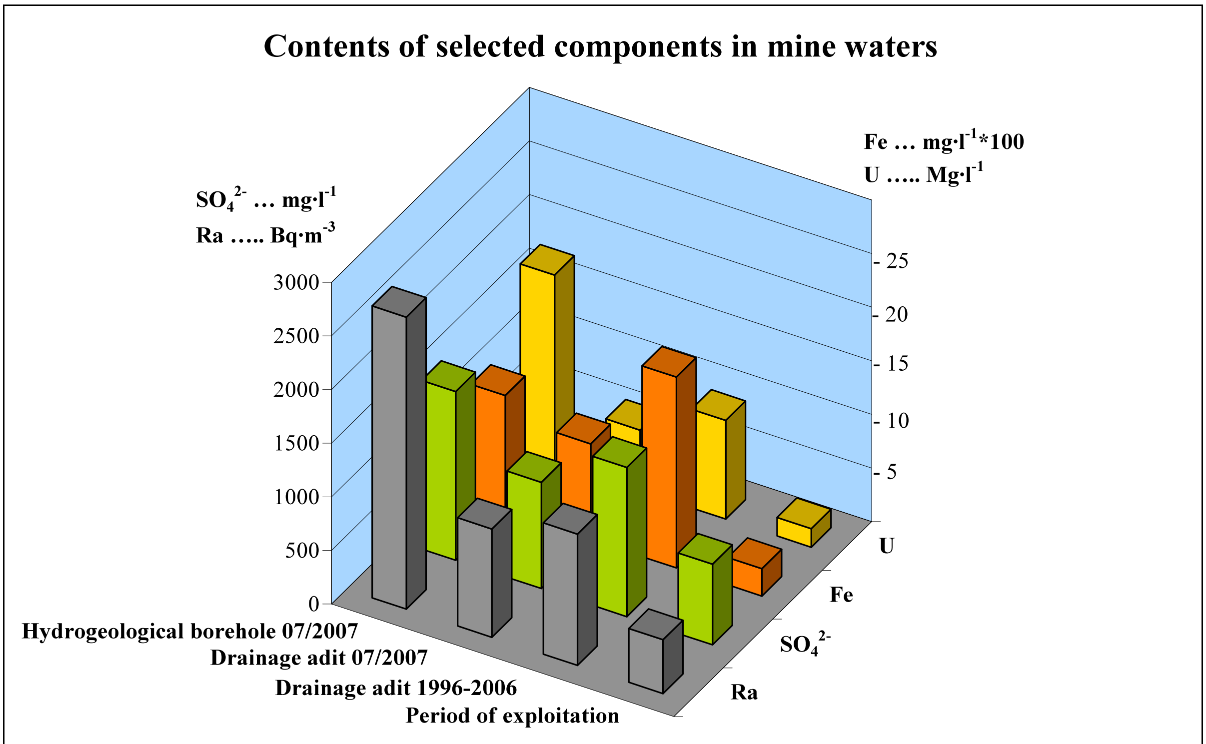

Figure 5. Comparison of the chemistry of mine waters from the Olší deposit.

With regard to the objectives of the research project, the hydrogeological borehole, or more specifically the pumping test, has confirmed the assumption that an accumulation of mine waters with uranium content suitable for utilisation as a secondary source of this raw material exist in deeper parts of the former mine. Whereas the concentration of uranium in mine waters during the period of exploitation did not exceed 4.5 mg·l^{-1}, after flooding the mine, the concentration of uranium in waters of shallow circulation increased to an average of

9.0 $mg \cdot l^{-1}$, but thereafter exhibits a declining trend (at present the concentration of uranium does not exceed 5.9 $mg \cdot l^{-1}$). However, after stabilisation of the water regime, a uranium concentration of 17.5 $mg \cdot l^{-1}$ was determined in quasi-stagnant waters outside the zone of active draining.

THE UTILISATION OF QUASI-STAGNANT MINE WATERS AS A URANIUM SOURCE

In the case of uranium mines, mine waters accumulated in the deeper parts of former mines, in a so-called quasi-stagnant regime, represent in view of their considerable volumes and high concentrations of dissolved uranium a significant secondary source of this raw material. The possible utilisation of these mine waters as a secondary uranium source is, to a certain extent, analogous to the mining method “in situ leaching”; the difference being that the process of uranium passing into solution is not intensified by the injection of acids into the rock environment. Instead, the natural process of dissolving uranium minerals in mine waters after their previous oxidation at the stage of deposit exploitation is utilised.

To verify whether or not the utilisation of quasi-stagnant mine waters as a secondary uranium source is viable, a pilot scale test is being prepared in the Olší deposit. The hydrogeological borehole will be used as a site for pumping mine waters with a high content of uranium. 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 back into the deposit at another point. The operation of active drainage of the flooded former mine through the treatment plant will not be changed in any way. On-going 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 treatment plant, will be kept under continuous observation. The engineering solution given here will not change present conditions in relation to the surrounding environment; all changes in mine water circulation will take place just inside the flooded deposit. The pilot test for the utilisation of quasi-stagnant mine waters with high uranium concentrations will provide necessary data for further theoretical work (mathematical modelling of flow and transport of matter in the flooded mine) and simultaneously will serve to verify theoretical knowledge.

CONCLUSION

After sufficient verification in the Olší deposit, it will be possible to apply the principle presented here to practically all uranium deposits that have been exploited by underground mining and where non-backfilled empty cavities, which have facilitated the creation of quasi-stagnant waters, have been left after exploitation. The specific engineering solution, especially the method of pumping quasi-stagnant water, must however rest on the conditions of the given deposit, its geological structure, hydrogeological conditions, the method of development and exploitation, as well as how the mine was decommissioned.

In addition to the Olší deposit, it is possible in the Czech Republic, at least in principle, to utilise quasi-stagnant waters in other former uranium-mining localities, such as the Okrouhlá Radouň, Zadní Chodov, Vítkov and, above all Příbram deposits. The first three deposits are comparable, in terms of geological structure and extent of mining, with the Olší deposit (accumulation of waters with a volume of 1.2 to 3.1.million m^3 in the underground parts); thus, a comparable yield of uranium from quasi-stagnant waters can be supposed. However, the extent of exploitation of the Příbram deposit is of a higher order (about 20 million m^3 of mine waters have accumulated in the underground parts). Therefore, if the above-mentioned technology is applied appropriately, the conditions exist for obtaining several times higher yields of uranium.

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