

Uranium mill tailing sites reclamation - comparison of approaches in the world uranium producing countries and the Czech Republic - Uranium Mine Dolni Rozinka

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Abstract: The submitted contribution deals with environmental impacts of uranium mill tailings, remediation of tailings sites and legislation in this subject area. The experience with uranium mill tailings reclamation in the large uranium producing countries is briefly presented. Czech legislation does not define naturally occurring radioactive material as a special category of radioactive waste and no special standards for uranium mill tailings control and reclamation exist. Abandoning of mining activities at the Uranium Mine Dolni Rozinka called preparation of the project of uranium mill tailings reclamation. The exploitation is permitted until the year 2002. According to the project of reclamation water released from the uranium mill tailings should be discharged to the deep horizons of the mine (600 to 300 m under the sea level). Mill tailings dams will be covered by inert material and the layer of clay sealing. The great depth of horizon, radioactive wastewater will be discharged to, and good sealing properties of rock massif (metamorphic rocks of Moldanubikum) ensure stable disposing of wastewater out of range of environment. This method of reclamation will make processing plant liquidation (after abandoning of mine) less cost demanding and faster.



1 GENERAL CHARACTER OF WASTE PRODUCED IN THE COURSE OF EXPLOITATION AND TREATMENT OF URANIUM ORES IN WORLD DEPOSITS

Traditionally, uranium ore is exploited in both open-pit and underground mines. The content of uranium usually ranges between 0.1 and 0.4 %. However, uranium deposits discovered recently in Canada and Australia contain not less than several per cent of uranium.

Due to the decrease of uranium prices in the world market after 1980, a number of mines have been closed by reason of their non-profitability. This also happened in the Czech Republic after 1990.

During the treatment process, conventional exploitation technologies create considerable amount of waste uranium mill, because the utilizable part usually represents less than one volumetric per cent of ore. For instance, the total volume of mill tailings sites in the U.S. represents more than 95% of the volume of entire radioactive waste produced in the process of the production of power, nuclear weapons, etc. In last decades, the method of in-situ uranium extraction has often been applied. U_3O_8 of up to 90% concentration is extracted in the treatment process and this method minimizes the production of solid waste.

Notwithstanding the fact that uranium is eliminated from milled rock in treatment plants, created mill contains all elements of the original ore. Since disintegration products such as Ra, etc. are not eliminated, the mill contains up to 95% of the original radioactivity of the ore. Besides, due to technical limitations, uranium itself contained in the ore cannot be extracted completely and the mill always contains 5 to 10 % of uranium contained originally in the ore. In addition, the mill also contains heavy metals and other contaminants from chemical substances used during the treatment. A number of other components (Mo, V, Se, Fe, As, etc. - according to foreign experience) extracted in the process of treatment of uranium ores can be environmentally dangerous in high concentrations.

Untreated waste rock deposited in waste dumps is another problematical waste of ore rocks from underground and open-pit mines. This waste product often shows increased radioactivity.

Uranium exploitation and treatment dislocates dangerous ore elements from their natural underground position into the environment.

2 POTENTIAL RISKS OF URANIUM MILL TAILINGS SITES

Uranium mill tailings sites contain radioactive ^{226}Ra (which is formed by uranium disintegration from minerals) and heavy metals that can migrate into groundwater. Levels of some contaminants in water specimens taken in the vicinity of mill tailings sites exceed acceptable levels for drinking water up to 100 times. Therefore, seepage from mill tailings sites represents a great risk for both underground and surface water.

Radionuclides contained in uranium mill tailings sites emit 20 to 100 times more of γ radiation than the level of natural deposit background. The radiation level decreases with distance intensively.

Radium - ^{226}Ra - disintegrates continually in mill tailings sites into radon (^{222}Rn). Radium with regard to its long half-life represents the main risk after the enclosure of treatment plant and mine.

Radon - ^{222}Rn - its half-life is 3.8 days (a product of α , γ radiation). It seems to be very short time, but regarding the long-term radon production (a disintegration product of radium ^{226}Ra with half-life of 1622 years - a product of α , γ radiation) it represents a long-term risk. It is disseminated by wind very fast,

therefore it increases radiation only very slightly. In fact, people can be affected only in the vicinity of its release.

Mill tailings sites are affected by many types of degradation and decomposition of substances. In regard of long half-life of some radioactive components of tailings sites, their safety must be guaranteed for a long time. If ore contains pyrite, then the access of precipitation water and oxygen enables sulphuric acid to form, which leads to the change of pH and the intensification of further process of contaminant extraction. In addition, mill components are not in geochemical balance with environment, which causes new reactions that create a further source of environmental danger.

A surface uranium mill tailings site is exposed to exogenic processes. Precipitation can form erosion beds, floods can destroy whole deposition sites, vegetation can penetrate deep into deposition sites and disperse material toward the surface, thus increasing radon emanation further and creating a deposit that is prone to climatic erosion. When the surface of a deposition site dries up, fine sands are wafted into wider environment and increased values of ^{226}Ra and arsenic are detected in it (example: uranium mill tailings site Wismut). The collapse of dams of deposition sites represents another possible risk. The collapse enables release of mill tailings water and solid mill components.

These negative phenomena of exogene processes can be prevented with technical means during reclamation and recultivation of tailings sites (creation of difficultly flushable covering layers sowing of grass, trees, bushes).

3 LEGISLATIVE APPROACHES TO THE HANDLING WITH PRODUCTS OF URANIUM EXPLOITATION AND TREATMENT

Even after World War Two, mine companies and uranium ore treatment plants left waste dumps and uranium mill tailings sites contaminated. Treatment plants and facilities were often not liquidated. In Canada, uranium mill was commonly disposed off into lakes.

Although the risk per unit of uranium mill is relatively low (compared to other radioactive waste), big volume of it and the lack of regulations dealing with its safeguarding and liquidation have led to extensive environmental contamination. In addition, half-lives of the main components of uranium mill are very long.

One of the first legislative acts to solve this untenable situation was the Uranium Mill Tailings Radiation Control Act, approved in the U.S. in 1978. This act defined basic juridical requirements for the remediation of uranium mill tailings sites. In 1983, the U.S. federal government formulated control standards for the contamination from both active and abandoned mill tailings sites. The main subject of these federal standards are the limit values for the seepage of radionuclides and heavy metals into groundwater, as well as radon ^{222}Rn emissions into atmosphere.

The U.S. legislation is the most detailed one in the area of the management of uranium mill. It includes some very specific codes and norms that are not at

disposal in the Czech (and European) legislation. Where there are no equivalents, it is, therefore, suitable to use the American norms and deal with them as with recommended values (e.g. "Summary of the U.S. Uranium Mill Tailings Standards").

The Czech legislation, classification and terminology does not declaratorily define natural radionuclides and their environmental risk. Only the types of exploitation waste are classified in a general way, according to the main types of raw materials (coal, ores, oil, etc.). The legislation does not define nor classify products of exploitation and treatment of natural radionuclides. Therefore, waste rock exploited from ore deposits or treatment mill after the application of sodium extracts cannot be filed and classified, for instance. This is the reason why problems occur when assessing the suitability of various methods and liquidation methodologies both in surface waste dumps and underground deposition sites (including deep geological structures). The legislation gradually creates the conditions under which it will be possible to dispose with radioactive mill after closure of chemical processing plant.

4 PRESENT CONCEPTIONS OF URANIUM MILL MANAGEMENT

In general, several methods of mill management are used in practice:

- a) surface deposition,
- b) shallow or deeper deposition,
- c) underwater deposition.

Surface deposition is usually used in site of the mill production (in-site type) or, if necessary (by reasons of settlement, flood areas, etc.), deposition sites can be moved to a more distant place (off-site type). This deposition method necessitates covering and stabilization of waste dumps. In shallow depositions, mill is deposited in natural or artificial depressions and covered by up to 3 m thick layer of soil to reduce erosion and radon emissions. Otherwise, mill can be deposited in the form of waste dumps with solid mill substance. The biggest waste dumps of this type in the U.S. and Canada contain up to 30 million tons of solid material. In Germany (Helmsdorf near Zwickau), such a waste dump contains 50 million tons.

Shallow or deeper deposition describes mill deposition in underground mines in the form of backfill, or on the bottom of deep open-pits. This approach is being applied mainly in Australia (uranium open-pit mine Ranger or mine Olympic Dam), France and Canada.

Underwater deposition is very suitable to suppress sulphate oxidation and radon release into atmosphere (Canada). Uranium mill is most commonly deposited in artificial or natural basins (ponds, lagoons), in surface depressions or in artificial basins with one tightening wall (deposition site being above the original terrain level).

Waste deposition in mines and open-pits is a subject of intensive debate worldwide. According to some authors, deposition of mill as a product of uranium ore treatment back in mines or open-pits is not an acceptable solution.

This is based on the fact that waste material does not become less risky (although most uranium has been extracted from the original ore). On the contrary, most contaminants have not been extracted (approx. 95% of radioactivity plus all chemical treatment pollutants remain in place). Moreover, such a material has been transformed by mechanical and chemical processes into a state in which it is prone to migrate into surrounding environment. After the uranium mill is deposited into an underground mine, pumping will stop, the waste will be flooded and the pollutants extracted from it. If conditions for uncontrolled migration of extraction water through rock environment are satisfied on such a locality - whether by natural or anthropogeneous reasons (as a result of previous mining activity) - the fear of such a liquidation method is well-founded.

The situation is similar to the situation in case of waste disposal into abandoned uranium open-pit mines. A direct contact with shallow circulation groundwater exists in most cases, as well as the risk of increased seepage of precipitation or surface water into groundwater. Groundwater contamination can be prevented only if an impermeable environment (natural or artificial) exists in place. A highly permeable layer is formed in some cases around the body of the deposited mill, which enables free circulation of percolated water around the waste. Since the mill permeability is by an order lower than this artificial drainage, practically no contaminants are transferred between the waste and groundwater. A similar method is being tested in Canada as a part of uranium mill disposal into lakes (the method is called "*pervious surround disposal*"). Other conceptions deny the necessity to create an artificial permeable layer (drainage) around the waste, providing that surrounding rocks are sufficiently permeable.

In most cases, mill is deposited on the surface, as there are no other options. The advantage of such a deposition method lies in better control of protection requirements. Special measures must be taken, however, to prevent erosion, transport into environment, radon release, etc. The main drawback of this method is the fact that uranium waste remains directly in populated environment.

The advantage of mill deposition in open-pit mines lies in its relatively good protection against erosion (good examples of this can be mill depositions in the Bellezane open-pit mine in France – Marschalko, 1997, in the Ranger open-pit mine in Australia, etc.). On the other hand, time-demanding consolidation of mill prior to the final terrain adjustment is a big problem, which necessitates application of some technical measures (vertical drainage and other constructional adaptations).

5 BASIC INFORMATION ON THE DEPOSITS OF URANIUM ORES IN THE CZECH REPUBLIC

Czech Republic is relatively rich in deposits of uranium ores. These strategical raw materials were utilized mainly between 1950s and 1970s. The damping of exploitation that started in 1990s has been so radical that there is only one active

deposit of uranium ores on the territory of the Czech Republic at present. This deposit - Rožná - will be exploited until 2002, according to the government decree.

This means that the center of activity has shifted from research and exploitation activities to the problems connected with the liquidation of uranium mines, different exploitation methods, recultivation of areas affected by ore exploitation and treatment and revitalization of affected environment.

Rožná is a hydrothermal uranium deposit located in tectonic zones and veins. It is tied to Pre-Cambrian rocks (biotite gneiss) of Moldanubicum in Bohemian Massif. The veins are deposited monoclinally, 50° to 70° to the west. Ore bodies of large dimensions containing uranium in the form of uranium minerals of uraninite and coffinite are exploited in an underground way.

The deposit has been opened by shafts, main and bottom crosscuts and offsets. The exploitation is realized by the method of underhand top slicing from raises driven along veins or in their bedrocks.

A chemical treatment plant was built near the mine in 1968, in which approximately 12,5 million tons of uranium ore have so far been processed. Ore is treated by the so-called alcalinous extraction - it is ground in the chemical treatment plant and then extracted by the solution of soda (Na_2CO_3) at temperature ranging around 85°C . The uranium concentrate obtained by this process is ammonium diuranate $(\text{NH}_4)_2\text{U}_2\text{O}_7$.

Two tailings sites have been built for the mill deposition. The K1 site (in operation since 1968) has an earth dam, a mill tightening on the shell and a drainage system to catch seepage water that is then discharged, back into the tailings site. The K2 site (in operation since 1978) is of a dam construction (tailings site dams are very little permeable and the drainage system only serves to catch little amount of contaminated water – Bujok et al., 1996). The tailings sites cover the area of 90 hectares and contain roughly 12 million tons of mill.

In regard of the fact that the treatment plant was situated in the protection zone of drinking water for the city of Brno, a closed water cycle (treatment plant \leftrightarrow tailings site) is dealt with. Since the sum of precipitation water and treatment plant water deposited in the tailings site exceeds its natural evaporation rate (by 90 to 100 mm per year), so-called "*overbalance water*" accumulates in place. The deposited mill bonds only about 0.37 volumetric per cent of water. The remaining proportion is free water that is contaminated by both radioactive minerals and chemical substances and products of the treatment process. The volume of overbalance water is about $300,000 \text{ m}^3$ per year. To liquidate it, a multi-stage evaporation station was built in 1976. It operates simultaneously with electrodialysis. The overbalance water is pre-processed (reduction of the content of unsolved substances, heavy metals, calcium and magnesium) and then it goes through the electrodialyser. The cleansed part is discharged into water stream, while the concentrate goes to the evaporation station. A part of evaporation residue is used for the production of a by-product - chemically clean and non-radioactive sodium sulphate that, in turn, is used for the production of detergents.

It is a unique technology that works in closed cycle. The remaining part of the residue in liquid condensate is deposited in tailings sites.

A system operating in this way induces the increase of concentration of substances in accumulated waste water in tailings sites. It is expected that approx. 1,2 million m³ of free overbalance water will be left untreated in the tailings sites in the area of Dolní Rožínka at the moment of termination of exploitation and treatment.

The main problem at present is to solve the liquidation of products of treatment of exploited ore, i.e. the liquidation of both solid products (uranium mill) and liquid products (overbalance water). From the environmental viewpoint, it is not possible to discharge overbalance water into recipients. Water discharge of streams is so low that further deterioration of water quality would be unjustifiable. Induced contamination of surface water would endanger the quality of water resources utilized in water works, because natural protection of the resources is low.

A) Two liquidation methods have been assessed for **liquid products of chemical treatment**, enriched with precipitation water in tailings site :

1. Electrodialysis and evaporation to separate salts from overbalance water (treatment of mill tailing water):

a) Concentrate to be disposed back into tailings site,

b) Condensate (clean water) to be discharged into water stream,

2. Deposition of overbalance water in deepest levels of the mine in the Rožná deposit.

Ad 1) Increase of electrodialysis and evaporation capacity

Regarding the total water balance in tailings sites, it is necessary to treat 370,000 m³ of water per year after the termination of the treatment plant operation. It is estimated that this would take 15 to 20 years, because the drainage system of tailings sites will work even after their total safeguarding. Utilization of already operating power-demanding electrodialysis and the evaporation station is planned for the treatment. Cleansed water will be discharged into water streams. In this case, the area of tailings sites would diminish, but mineralization and contamination of tailings site water would increase.

Ad 2) Deposition in deep horizons of the Rožná deposit

The first documentation concerning the problem of deposition of overbalance water from tailings sites of the chemical treatment plant into the deepest parts of mine workings of the Rožná deposit was made by VÚGI Brno in 1993 (Pelikán, et al, 1993).

The study was then supplemented with the evaluation of safety of deposition of overbalance water from the viewpoint of the transport of contamination in mine water. The problem was later dealt with by ČVUT Praha (Havlík et al, 1993, 1994, 1995). In 1997 this variant was approved as a plan for the liquidation of overbalance water.

It has been proved on the basis of experimental measurements that the fissure system in depths exceeding 450 m under the surface is almost ineffective from hydraulic point of view. Permeability of the massif is much lower than the values of the classification of permeable rocks ($K \ll n \cdot 10^{-8} \text{ m s}^{-1}$). No hydraulically active tectonic faults have been detected in deeper parts of the opened deposit of Rožná.

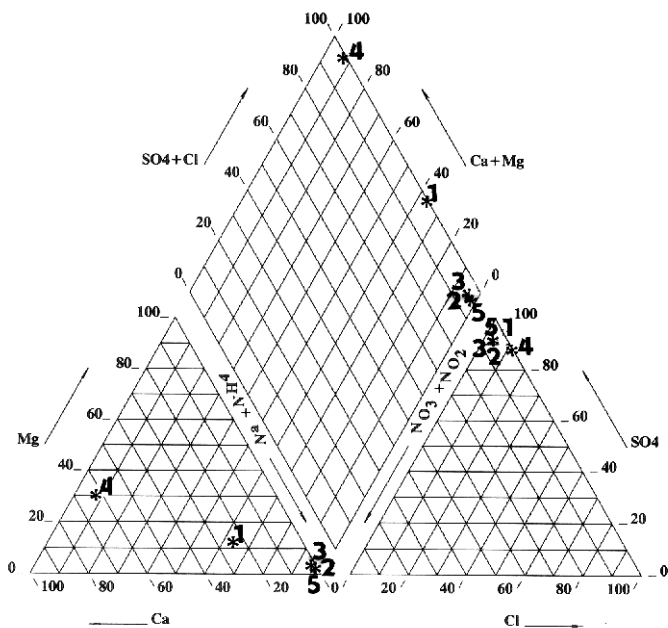


Figure 1: Piper diagram of waste and mine waters.

1-Mine water - Mine Rožna -before flooding; 2- Simulation of mine waters chemism after disposing of waste water after 6 month, 3- after 12 month; 5- Condesate to be disposed in mill tailing site

Mother gneiss rocks of the deposit are geochemically very stable from the viewpoint of aggressiveness of water-bearing environment. Regarding the chemism of water from tailings sites (increased content of U, Ra, Fe and Mn only) and natural mine water resources in the deposit, hydrochemical influence of water deposited 900 to 1200 m below the surface will be insignificant. Owing to natural radioactivity of rock environment of the deposit, residual radioactivity of overbalance water will not be an important contaminant in the deposition site, either. Geothermic gradient of the deposit (approx. 55 m per 1°C) is also favourable - temperature ranges around 29°C in the depth of 1200 m.

B) Two liquidation methods have also been assessed for **solid products of chemical treatment**, including water bonded in mill:

a) Deposition in deep parts of the mine (below the level of deposited overbalance water) :

a) Flushing of untreated mill and tailings sites,

- b) Mill to be used as a basis for suitably formed backfilling mixtures,
- b) Mill is left in tailings sites (the surface of tailings sites to be insulated and recultivated after overbalance water is liquidated). This is a preferred solution at present.

6 GENERALIZATION OF HITHERTO KNOWLEDGE CONCERNING THE CONDITIONS FOR THE LIQUIDATION OF URANIUM TREATMENT WASTE BY ITS DEPOSITION IN MINES

In connection with direct application of global hitherto experience, it must be realized that it is not possible to apply some of the methods directly and without previous adaptation because of different (variable) natural, demographic and other conditions (e.g. climatic conditions in the U.S. and Australia are similar - semiarid and arid areas - yet the approach is different). Furthermore, inveterate methodology, deposition strategy, experience and routine come into the decision making as an important factor. Policy of local and state authorities and organizations also play significant role - mainly in connection with civic attitudes and initiatives. Preferred liquidation methods of uranium mill in certain countries are as follows: Canada - lakes, U.S. - surface deposition sites - waste dumps, Australia - open-pit and underground mines, Europe - surface deposition sites and open-pit mines.

Opinions differ only in case of mill deposition in underground mines - from totally negativist to accepting. In different countries - in developed countries on one hand and the so-called Third World countries on the other hand - the approach to such an evaluation is totally different from the viewpoint of both legislation and control. Different risk criteria are applied, which often coheres with living and social standards of individual societies, level of technology, health care, population density, etc. (Fendeková & Némethy, 1998)

In case of waste deposition in rock environment in the Czech Republic, deposition of **selected waste types** into **selected mine workings** is accepted. This method of liquidation of selected types of waste can be agreed of only on the pre-condition that four basic requirements are fulfilled :

1. Concrete site and type of waste will be approved by local Mining Office.
2. Deposition will be discussed with the water management organisation which on basis of hydrogeological assessment decides whether waste disposing at a specific site cannot threaten environment (especially from the point of view of groundwater). Hygienic service will assess the impact of manipulation with waste on human health.
3. Deposition will be discussed with local administration (according to conditions given by water management organisation, environmental institutes) and complex impact on environment will be assessed.

The process of legislative requirements on the establishment of underground waste repositories is under present conditions very demanding.

Geological and hydrogeological requirements on locality.

A locality is geologically and hydrogeologically suitable, if:

- it is hydraulically insulated from aquifers used for water supply purposes,
- it is not situated in area with priority hydraulic pathways interconnecting it with other hydrodynamic systems (natural : permeable tectonic zones, artificial : mine workings, wells, permeable areas disturbed by fissures opened in consequence of mining, etc.),
- it is situated in a sufficient depth below the surface where groundwater flow is minimized or water is stagnant (sufficient rock massif thickness between aquifer with intense water exchange with surface and quasi-stagnant hydrodynamic system),
- present properties of rock massif will not be changed in the future in consequence of mining or other artificial impact on created natural regime. The area is not situated in the zone of occurrence of extreme geomechanical phenomena- natural (earthquakes) or induced (rock bursts),
- dumpings must be hydraulically sealed to prevent the transport of contaminants to surrounding hydrodynamic systems after flooding of repository. Hitherto geological and hydrogeological knowledge can be, with respect to the possibility to deposit uranium treatment mill in mine workings of uranium mines, specified and summarized into the following main items :
 - Disposing will be carried out in the depths larger than 400 m below surface (hydraulically closed primary joint systems),
 - Mine workings, used for waste disposing, must not go through aquifers (tectonic zones or layers with hydraulic conductivity $K > 10^{-8} \text{ m.s}^{-1}$),
 - Mill in mine working will be confined and insulated by flushed-ash dam with a tightening insertion made of clay. Dams will be dimensioned for pressure of up to 10 MPa, and resistant against brittle failure,
 - Repository must be leak-proof to ensure that pumped water from the level or mine will not contain pollutants leached from deposited mill in concentrations exceeding the limits.

Requirements on the deposited uranium mill

1. Uranium mill will not produce toxic substances under both present mine conditions and future natural conditions,
2. Uranium mill and its extracts will not increase contamination (neither chemical, radiological nor biological) of mine water above the present state (or an otherwise determined limit state) of water discharged into water stream or other surface water system.
3. Insulation of deposition must permanently prevent pollution of surrounding hydrodynamic systems over the limit (the quality of mine water discharges is a decisive factor). Sealing must be efficient even under extreme conditions and after possible strong geomechanical events

4. Deposited uranium mill must be pre-processed so that its physical-chemical and sensoric properties do not endanger mine environment or labourers that transport and handle it, until it is insulated by its enclosing in the deposition site.

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Rekultywacja obszarów składowania odpadów z kopalń uranu – porównanie rozwiązań stosowanych w krajach wydobywających uran i w Republice Czeskiej – kopalnia uranu Dolni Rozinka

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Streszczenie: Artykuł zajmuje się wpływem odpadów z kopalń uranu na środowisko, rekultywacją składowisk i obowiązującymi w tym zakresie przepisami prawnymi. Omówiono również doświadczenia związane z rekultywacją tego typu odpadów w krajach wydobywających uran na szeroką skalę. Czeskie przepisy prawne nie definiują naturalnie występującego materiału radioaktywnego jako specjalnej kategorii odpadów radioaktywnych i nie istnieją specjalne standardy określające kontrolę i rekultywację tych odpadów. Zamknięcie kopalni uranu Dolni Rozinka spowodowało konieczność przygotowania projektu rekultywacji. Zgodnie z projektem woda ze stawów poflotacyjnych będzie odprowadzona do głębokich izolowanych wyrobisk kopalni położonych na głębokości 600 do 300 m p.p.m.. Stawy poflotacyjne zostaną przykryte warstwą uszczelniającej gliny. Znaczna głębokość poziomów,

do których odprowadzona będzie woda oraz dobre właściwości uszczelniające metamorficznego masywu skalnego niwelują zagrożenie ze strony radioaktywnych wód. Metoda ta uczyni likwidację kopalni tańszą i szybszą.