Remediation of Cretaceous Sediments Affected by Uranium In-Situ Leaching (ISL) in the Czech Republic - 9108

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ABSTRACT

During the last 60 years since the end of Second World War uranium mining and milling in the Czech Republic has caused an enormous devastation of the environment by means of waste dumps accumulation, waste dumps left after uranium prospection, tailings impoundments and other workings. All these negative impacts influenced the quality of the environment and affected mainly surface and ground waters, soils and simultaneously polluted great areas of land and endangered the sources of fresh water.
The situation of this region here is more critical because the fact that the area belongs to the nature reservoir of drinking water protection in North Bohemian Cretaceous Platform.
Stráž pod Ralskem mining district which was chosen as the most heavily polluted locality geologically represents a stratiform sandstone type of deposit where acid in-situ leaching (ISL) has been applied as the mining method since 1968. A quantity of 4.1 million metric tons of H_2SO_4, 320 thousand metric tons of HNO_3 and 111 thousand metric tons of NH_4 have been injected into the leaching fields of the Stráž deposit in the last 35 years which has affected about 370 million m^3 of Cenomanian water over an area of 27 km^2. To solve the negative influence of coexistence of Uranium Hamr deep mine and ISL Stráž mine a protective hydraulic barrier has been built between these two deposits.
To suppress above mentioned negative impact of ISL a desalination plant (vaporization station) with a capacity of 5 $m^3.min^{-1}$ has been built and started to operate in 1996.

The objectives of the remediation activities have been as follows:

- to restore the geological environment to a condition guaranteeing the continuing use of Turonian drinking water of Northern Bohemia Cretaceous region,
- to decommission bore holes and surface installations,
- to incorporate the surface of leaching fields into the ecosystems taking into account regional systems of ecological stability and urban plans.

The entire remediation process is expected to finish in 2035. During this period 3.7 million metric tons of contaminants (2.8 million metric tons SO_4^{2-}) will be withdrawn from the ground. The total cost of remediation process is expected to be on the approximate level of 40.9 billion Czech crowns (1.55 billion EUR, 2.55 billion U.S. dollars).
Decommissioning of chemical leaching of uranium is a long lasting and complex process that must be continuously evaluated and specified. Until the end of remediation process the extensive monitoring, verification and modeling work will be carried out. Respective Czech authorities continuously approve the process of remediation and its individual components.
The report describes the contemporaneous stage of the governmental remediation and recultivation program.

HISTORICAL REVIEW

The Bohemian Massif belongs to a very important uranium bearing province. Uranium mineralization has been connected with post-variscan hydrothermal activity and emplacement of carbonatic dikes with uranium mineralization. Uranium ores have been mined in Jáchymov (Joachimsthal) since 1840 first for making paints and later when radium and polonium have been discovered by Mme Curie even for Radium producing. During the years 1907-1939 a total of 2.5 – 5.5 g of radium per year have been produced there.

The years 1945 - 1960 started the period of exploitation of uranium ores for army purposes and from uranium ore from Jáchymov the first Soviet atom bomb has been probably manufactured.
After the Second World War the uranium exploration grew rapidly as a large scale program in support of Czechoslovak uranium production industry. A systematic exploration program including geological, geophysical and geochemical surveys and related researches was carried out to assess the uranium potential of the entire country. Subsequently there have been opened and exploited following uranium districts:

West Bohemian District
Příbram and Central Bohemian District
Rožínka District
North Bohemian Uranium District

URANIUM MINES IN THE CZECH REPUBLIC

The majority of uranium deposits in Bohemian Massif are of vein type but this contribution pay attention to the stratiform, sandstone type deposit which is situated in North Bohemian Cretaceous Sedimentary Basin. It is the newest uranium producing district in the Czech Republic and its exploitation has been under way since 1967.
Because of relatively large deposit (about 200 thousand metric tons) the U-production was meant to cover all the long term needs of Czechoslovak nuclear power plant supply, including export to former COMECO countries.

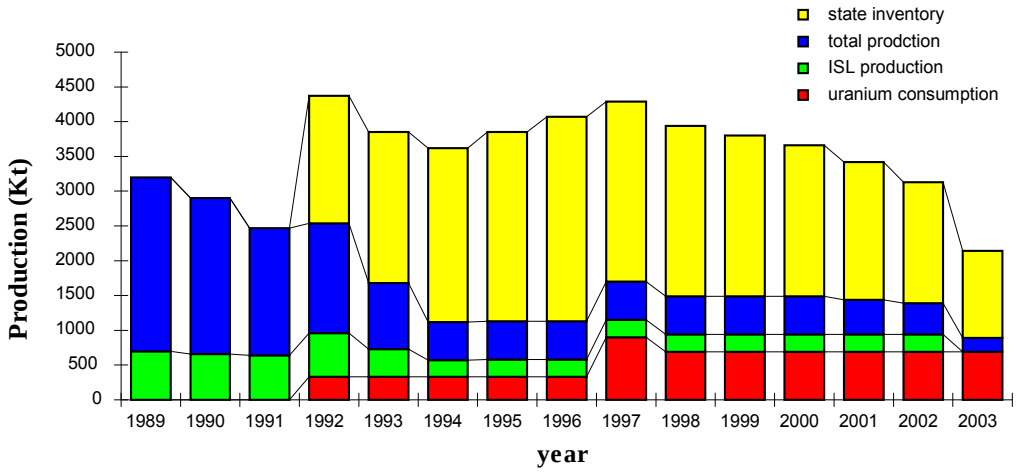

Fig. 1 Uranium production and use in the Czech Republic 1989 - 2003

NEGATIVE IMPACT ON THE ENVIRONMENT

After 1989 the political and economical situation in Europe has changed dramatically and finally attention has been paid to the evaluation of negative impacts of extensive mining activities of the past 50 years on the environment of the Czech Republic. There have been taken government decisions to close down majority of uranium mines and the subsequent assessment of negative impacts of uranium mining on the

environment have shown the necessity to install the remediation program of all areas where the uranium mines had operated.
The uranium districts in consideration were: West Bohemian District, Příbram and Central Bohemian District, Rožínka District and North Bohemian Uranium District.

Fig. 2 Map of uranium mines in the Czech Republic

Table I. Uranium mining waste dumps and milling tailings in the Czech Republic

Localities	Waste dumps	Tailings	
	Volume (thousand m^3)	**Extent** (ha)	**Volume** (thousand m^3)
Stráž pod Ralskem	1,137.0	187.0	19,236.0
Rožínka (GEAM)	3,290.0	90.1	9,827.4
MAPE Mydlovary		292.7	23,969.0
Příbram	30,072.0	44.1	238.3
West Bohemian Region	2,125.0	20.1	2,798.0
Jáchymov and Horní Slavkov	14,382.0		
Others	7,416.0		
TOTAL	**58,422.0**	**634.0**	**56,068.7**

The assessment of negative impacts of uranium mining and milling on the environment has shown the necessity to start immediately the wide remediation program of all environmental issues connected with uranium mining and milling as listed above.
The uranium deep mines in Hamr and ISL in Stráž pod Ralskem were chosen as the heavily affected area for the restoration program because in-situ leaching (ISL) and underground mining were operating together in the same geological unit i.e. Cretaceous Cenomanian sandstones which represent an important reservoir of drinking water. The mining activities, especially ISL are influencing the ground water regime and the pollution is enormous. Total quantity of chemicals injected to ISL fields in Stráž pod Ralskem during the period 1973 – 2005 is shown on Fig. 3.

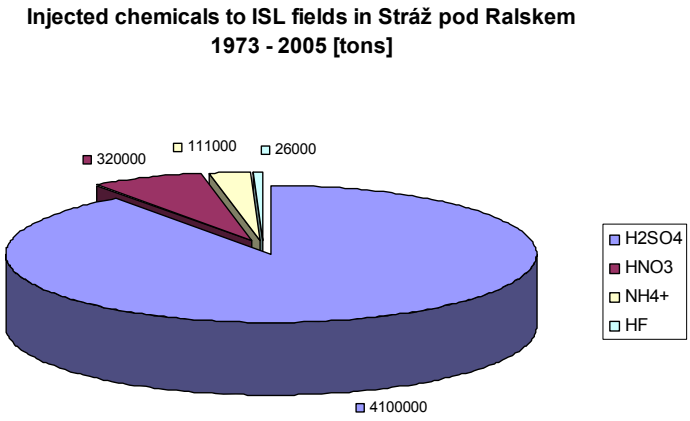

Fig. 3 Total quantity of chemicals injected to ISL fields in Stráž pod Ralskem 1973 - 2005

The main problem for uranium exploitation in this area is the coexistence of two large production complexes - classical deep mining at the Hamr deposit and ISL at the Stráž deposit. Both deposits are situated near each other and they have negative influence on the hydro-geological situation of the whole area.
Two methods of extraction have been applied within the Stráž block since 1973:

a) Classical underground mining in Hamr mine and Břevniště. A stable depression of the water table in Cenomanian aquifer has been achieved due to the long term water pumping out from the mine at a rate of about 50 $m^3.min^{-1}$.
b) The ISL has been in operation for about 35 years and so far 32 ISL claims have been commissioned covering a total area of about 6 km^2.

A quantity of 4.1 million metric tons of H_2SO_4, 320 thousand metric tons of HNO_3 and 111 thousand metric tons of NH_4 have been injected into the leaching fields of the Stráž deposit in the last 35 years. This has affected a total of more than 370 million m^3 of Cenomanian water over an area of 27 km^2. Leach solutions from ISL fields have dispersed horizontally and vertically not only to the Cenomanian horizon but along the extraction boreholes and tectonic lines to the Turonian drinking water aquifer above.
To solve the negative influence of coexistence of Hamr mine and ISL Stráž mine a protective hydraulic barrier has been built between these two deposits. To suppress above mentioned negative impact of ISL the construction of a desalination plant (vaporization station and alum crystallization unit) with a capacity of 5 $m^3.min^{-1}$ has been later projected and started to operate in 1996. The positive results were already achieved:

- polluted underground waters were stabilized,
- the water level in Cenomanian aquifer went down,
- overlaying Turonian aquifer has been not affected by Cenomanian polluted liquors.

ENVIRONMENTAL REMEDIATION PROGRAM

Remediation – present state

Remediation of the consequences of chemical leaching of uranium in Stráž pod Ralskem is a complex problem, consisting of a variety of partial solutions. The Czech government adopted two separate resolutions with respect to this matter, namely the Governmental Decree of 20th May 1992 No. 366, determining a transitory period between 1992 - 1994, including the characteristics of special regime of

exploitation, and the Governmental Decree of 6th March 1996 No. 170, presenting the decision on terminating chemical leaching of uranium in Stráž pod Ralskem.
The assessment of the situation led the Czech government to the decision to terminate chemical leaching as of 1st April 1996 and to perform active remediation work.
The objectives of the remedial activities are:

- to restore the rock environment to a condition guaranteeing continuing usability of Turonian water of Northern Bohemia Cretaceous,
- to decommission bore holes and surface installations,
- to incorporate the surface of leaching fields into the ecosystems taking into account regional systems of ecological stability and urban plans.

For remediation the pump and treat remediation approach is used and innovative in-situ immobilization approach is planned to use. The pump and treat approach is following. The dissolved uranium is separated on chemical station from the abstracted Cenomanian solutions. After uranium sorption the part of the solutions is led to evaporation station where it is concentrated and during crystallization process the alum ($NH_4Al(SO_4)_2.12H_2O$) is produced from the concentrate. Alum is reprocessed onto aluminum sulfate and ammonium sulfate, which is returned back into the crystallization process. The distillate from evaporation is discharged into the Ploučnice River. The mother liquor after crystallization of alum is injected back to the ground. Another part of the Cenomanian solution after the uranium sorption is decontaminated on neutralization station NDS 6. This station decontaminates not only Cenomanian solutions but also a part of Turonian water and water from inner drainage of tailings pond. The filter cake from neutralization process is deposited in the tailings pond. The scheme of above-mentioned existing surface technologies is shown on Fig. 4.

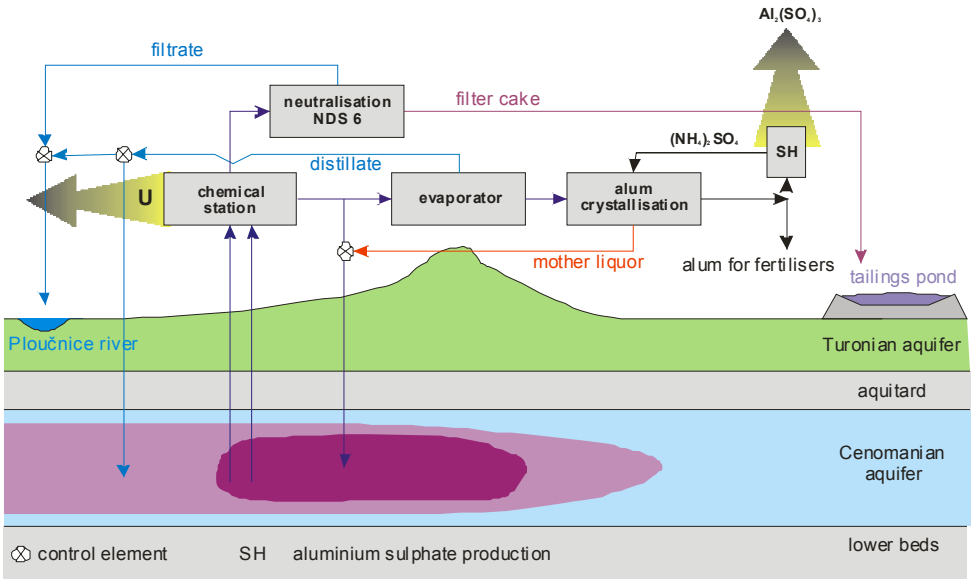

Fig. 4 Scheme of present configuration of surface remediation technologies

Remediation – future plans

The present configuration of surface technologies does not satisfy the demands on the whole remediation process. So that it is necessary to complete the chain of surface technologies as it is shown on Fig. 5.

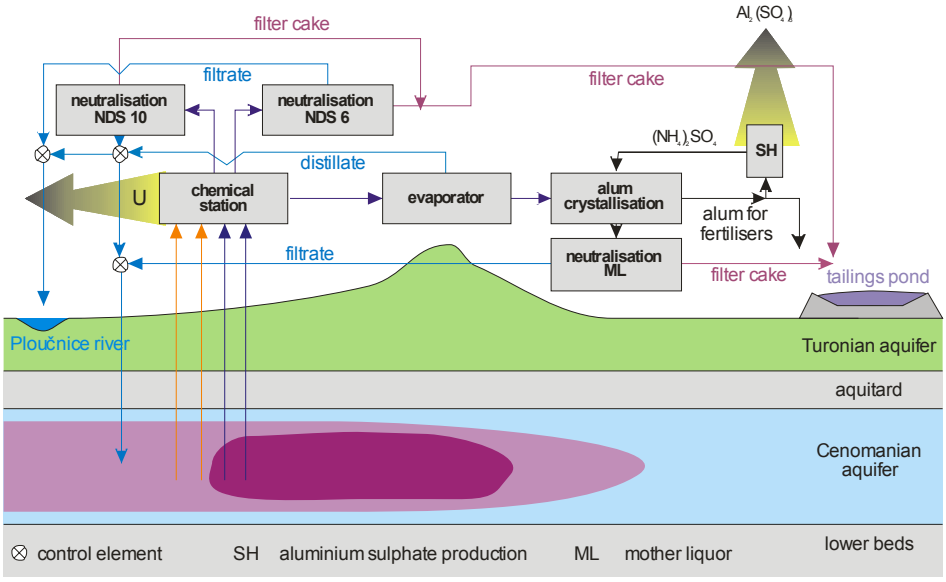

Fig. 5 Scheme of complete configuration of surface remediation technologies

At the beginning of 2010 the liquidation of mother liquor by neutralization process will start. The stage II of drilling of remediation wells will be finished at the end of the same year. Another neutralization station NDS 10 will start its operation in 2014.
The principle of innovative in-situ immobilization approach is to develop special conditions in water bearing sandstone sediments when transformation of contaminants from mobile form to immobile form can happen. Under the conditions of remediation after chemical mining of uranium on the deposit Stráž it means injecting suitable alkaline medium and its spreading in contaminated sandstone aquifer. It will lead to decreasing of acidity of contaminated groundwater and precipitating of contaminants (SO_4^{2-}, Al and Fe) in pores. This process is followed by co-precipitation and sorption of other toxic contaminants as As and Be. It seems to be optimal to take advantage of basicity of solutions after neutralization and to inject these solutions in the ground to neutralize less acidic ground water.
In 2008 and 2009 the pilot experiment of immobilization in-situ is realized. The results of the experiment will be used for design of application of this method in the frame of remediation process with the aim to decrease time and costs.

Environmental Impact Assessment (EIA)

Final Report has been prepared for the Czech Government and the Environmental Impact Assessment (EIA) has been concluded evaluating not only the negative impact of uranium mining and milling on the environment but also the impact of activities proposed by DIAMO State Enterprise in designed remediation program.

The Statement of the Ministry of Environment with Respect to Proposed Environmental Restoration Program

The Ministry of the Environment taking in consideration the projects regarding the restoration program of the area affected by uranium in-situ leaching in Stráž pod Ralskem agreed with the concept of remediation program and EIA submitted to the Ministry of the Environment within the following conditions:

1. The first stage of the remediation program will start in1996 and will be followed by the operation of the vaporization station to stabilize and control the leach liquors in the Cretaceous aquifers. This stage

will represent the beginning of the complex program of remediation of the environment in the whole area. Uranium extraction from the leaching fields will continue as subordinate process of restoration program.
2. At the same time the monitoring system will be modified according to the results gained to assure the system will register the effectivity and progress of the restoration process.
3. The restoration steps will be coordinated with the liquidation of the Hamr underground mine and tailing impoundments in the Stráž region. Simultaneously the geochemical contamination of the surface sediments and waters along the river Ploučnice will be solved.
4. The reclamation of land will be coordinated with the technological regime of rehabilitation and parts of natural reservation (Ralsko, Velký a Malý Jelení vrch, Lipka) have to be protected as the systems of ecological stability.
5. DIAMO State Enterprise will arrange a complex socio-ecological study which will consider the program of revitalization of the area.
   Taking in consideration the difficulty of the problem The Ministry of the Environment required from DIAMO State Enterprise to accomplish:
6. A complex risk analysis of proposed concept of the remediation program with special attention to the contamination of the Turonian aquifer.
7. A report on the possibility of immobilization of the contaminants underground in the Cenomanian aquifer as the follow-up program of decontamination.
8. To define and prepare a follow-up project dealing with the products of the vaporization station extracted from underground and particularly dealing with the remaining constituents (incl. radioactive wastes).
9. Simultaneously conduct processes to produce materials which will be suitable for further technological use or for sale.
10. Products - minerals potentially possible for reuse have to be safely stored.
11. To prepare the study regarding the minor elements content which have been enriched during the circulation of salinated liquids underground (REE, Al, Be, Ni etc.) as future non-traditional mineral resources.
12. To prepare a broad research program and follow up studies with the aim to finalize the technological program and verification of executed steps.

CONCLUSION

The remediation program of DIAMO State Enterprise supervised by The Ministry of Environment is carried out according to the Environment Protection Law No. 17/1997 Coll. Monitoring Progress Reports are issued every year and describe the progress in the remediation of all components of the environment (i.e. air, ground water, surface water, mine waters, soil and even the parameters of radioactive safety) in the whole area of Stráž pod Ralskem ISL Mine. It follows the regulation norms as ČSN EN ISO 9001:2001 and Notice No. 214/1997 Coll. of State Office of Radioactive Safety.
Long-lasting evaluation of monitoring results of the remediation program shows that many particular localities within the area have reached sufficient figures of contamination limits. There starts to appear a positive effect of different remediation technologies followed by the reclamation of land for industrial and even agricultural purposes. There were also registered natural processes of attenuation causing natural degradation of contamination. According to this effect the management of DIAMO State Enterprise prepared a system of optimal steps of further monitoring. The main aim of remediation of the negative impacts of the industrial activities on the environment is to return the affected land to its natural use.

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