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Chapter 26

PILOT EXPERIMENT OF IMMOBILIZATION OF CONTAMINANTS IN-SITU

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ABSTRACT

At the end of 2006 the project of pilot experiment of immobilization of contaminants in-situ was prepared in DIAMO, s. p., o. z. TUU. Realisation of the experiment is planned for years 2007 and 2008.

The principle of immobilization of contaminants in-situ 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 (using sulphuric acid) of uranium on the deposit Straz 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, Fe) in pores. This process is followed by co-precipitation and sorption of other toxic contaminants as As and Be.

The results of the pilot experiment will be used for design of application of method of immobilization in-situ in the frame of remediation after chemical mining of uranium on the deposit Straz with the aim to decrease time and costs of the whole remediation process.

Keywords: Immobilization of Contaminants; Uranium Mining; Remediation

1. INTRODUCTION

Uranium mining using in-situ acidic leaching method was performed from 1967 to 1995 on the uranium deposit Straz located in North Bohemian cretaceous table in Czech Republic. During this period an enormous amount (almost 5 mil. tons) of chemicals, mostly sulfuric acid as the leaching medium, was injected into the ground. The products of leaching and the rest of injected chemicals cumulated in the groundwater of the Cenomanian horizon. The influenced volume of groundwater is more than 300 millions m$^3$ in the area of 27 km$^2$.

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In 1996 the injecting of sulfuric acid was finished and then the remediation of contaminated groundwater started. The groundwater is cleaning using classical pump and treat technology. The contaminants are abstracted from groundwater in surface technologies, and then they are reprocessed into industrially usable products (alum, aluminum sulfate, ammonium sulfate) or deposited in tailings pond of former chemical treatment plant. The part of cleaned water is discharged in the river and the rest is injected back in the ground.

In original remediation conception the use of the method of in-situ immobilization of contaminants was planned for the last years of a remediation process when concentration of contaminants in groundwater will sufficiently decrease.

The updated remediation conception assumes more extended use of neutralization cleaning technologies to accelerate contaminants’ abstraction from groundwater. It seems to be optimal to take advantage of the basicity of solutions after neutralization and to inject these solutions into the ground to neutralize less acidic groundwater (see scheme on Fig. 1) before it was originally planned. It means not only at the end of the remediation process but also during the remediation.

![Diagram of injection of alkaline solutions after neutralization](image)

*Figure 1. Scheme of injection of alkaline solutions after neutralization (decontamination)*

2. PRINCIPLE OF IMMOBILIZATION

The principle of immobilization of contaminants in-situ 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 (using sulphuric acid) of uranium on the deposit Straz, it means injecting a suitable alkaline medium and its spreading in contaminated sandstone aquifer. It will lead to decreasing of acidity of contaminated groundwater and precipitating of contaminants (SO42-, Al, Fe) in pores. This process is followed by co precipitation and sorption of other toxic contaminants as As and Be.

Since 1996 when remediation process began, the evaluation of changes in rock environment on the former deposit Straz have been continuously performed. The results of the evaluation have formed a view on target and effect of immobilization. In the beginning of the remediation process the massive decrease in porosity and permeability was considered as the main effect of immobilization. Now we see the effect in following:

- neutralization of acidic contaminated groundwater;
- decreasing of salinity of groundwater in consequence of transformation of contaminants into the form of insoluble precipitate;
- creating an environment suitable for long-lasting positive chemical changes in groundwater;
- liquidation of contaminants from inactive pores and sediments with low permeability;
- creating protective and reactive barriers and isolating of rest contamination in small volume of groundwater in central part of leaching fields;
- creating of conditions suitable for natural attenuation.

Since it is possible to use immobilization from the beginning of remediation, we expect a shortening of the complete remediation process. We also expect a decrease in total remediation costs.

The solutions after neutralization will be available during the whole remediation process in sufficient volume. There will be no need to use any other medium for immobilization which preparation is much more expensive.

Performed static and dynamic laboratory experiments and chemical processes simulation calculations have brought important knowledge which helps to forecast (using numerical modeling) expected hydrogeological and geochemical trends. Unfortunately they cannot fully answer questions connected to large scale immobilization applications. To obtain such answers we prepared at the end of 2006 the project of pilot experiment of immobilization (DIAMO, 2006).

3. DESCRIPTION OF PILOT EXPERIMENT

The pilot experiment of immobilization will be realized in south-eastern part of leaching field VP 8C (see Fig. 2). This area has suitable geographic position, geological, hydrogeological and hydrochemical conditions.

It was designed using numerical models to use currently operating well network in chosen locality and to add five new wells (see Fig. 3). Four of five new wells together with six to eight wells from the currently operating well network will be used for monitoring of pilot experiment.
The fifth new well will be used for injecting of immobilization medium (solution after neutralization). The depth of wells is approx. 200 m.

**Figure 2.** Locality for realization of pilot experiment of immobilization

**Figure 3.** Location of five new wells for pilot experiment
As the immobilization medium, the filtrated solution after neutralization on decontamination station NDS 6 was chosen. The solution will lead from separation tank on NDS 6 to pilot experiment locality on VP 8C by 2500 m long pipeline.

The chemical composition of alkaline solution after neutralization and quality of groundwater influenced by chemical mining of uranium on locality VP 8C are documented in the following table, Table 1.

Table 1. Chemical composition of immobilization medium and quality of groundwater on locality VP 8C

<table>
<thead>
<tr>
<th></th>
<th>unit</th>
<th>immobilization medium</th>
<th>VP 8C groundwater</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH (20 °C)</td>
<td></td>
<td>11,5 –– 12</td>
<td>2,0 –– 2,1</td>
</tr>
<tr>
<td>Eh (abs) (20 °C)</td>
<td>[mV]</td>
<td>240</td>
<td>~ 800</td>
</tr>
<tr>
<td>Spec. conductivity (20 °C)</td>
<td>[mS.m⁻¹]</td>
<td>415</td>
<td>1 400 –– 1500</td>
</tr>
<tr>
<td>Density</td>
<td>[g.cm⁻³]</td>
<td>1,002</td>
<td>~ 1,016</td>
</tr>
<tr>
<td>RL</td>
<td>[mg.l⁻¹]</td>
<td>2 000 –– 2 100</td>
<td>20 000 –– 25 000</td>
</tr>
<tr>
<td>NH₄⁺</td>
<td>[mg.l⁻¹]</td>
<td>80 –– 100</td>
<td>300 –– 400</td>
</tr>
<tr>
<td>Na</td>
<td>[mg.l⁻¹]</td>
<td>140</td>
<td>10 –– 15</td>
</tr>
<tr>
<td>K</td>
<td>[mg.l⁻¹]</td>
<td>30</td>
<td>20 –– 25</td>
</tr>
<tr>
<td>Mg</td>
<td>[mg.l⁻¹]</td>
<td>1,5</td>
<td>25 –– 35</td>
</tr>
<tr>
<td>Ca</td>
<td>[mg.l⁻¹]</td>
<td>600 –– 650</td>
<td>150 –– 200</td>
</tr>
<tr>
<td>Fe</td>
<td>[mg.l⁻¹]</td>
<td>&lt; 0,05</td>
<td>500 –– 550</td>
</tr>
<tr>
<td>Al</td>
<td>[mg.l⁻¹]</td>
<td>&lt; 0,5</td>
<td>1 800 –– 2 400</td>
</tr>
<tr>
<td>NO₃⁻</td>
<td>[mg.l⁻¹]</td>
<td>50 –– 60</td>
<td>230 –– 280</td>
</tr>
<tr>
<td>F⁻</td>
<td>[mg.l⁻¹]</td>
<td>1,0</td>
<td>60 –– 100</td>
</tr>
<tr>
<td>Cl⁻</td>
<td>[mg.l⁻¹]</td>
<td>20</td>
<td>~ 5</td>
</tr>
<tr>
<td>SO₄²⁻</td>
<td>[mg.l⁻¹]</td>
<td>100 –– 1 100</td>
<td>15 000 –– 20 000</td>
</tr>
</tbody>
</table>

The preparation and realization of the pilot experiment of immobilization will last two years (2007-2008). The whole experiment is divided into two phases – phase of preparation and phase of realization.

Phase of preparation will include:
- the building of a pipeline from NDS 6 to VP 8C locality and building of technological equipment;
- check of current status of existing wells;
- cleaning of wells;
- drilling of five new wells (one for injecting of immobilization medium, one core well for monitoring using special zonal sampling method and three wells for classical hydrogeological monitoring);
• logging, groundwater quality sampling, piezometric head measuring, chemical analyzing;
• numerical modeling.

Realization phase will include:
• injecting of the immobilization medium for at least 150 days in the volume rate of 200 l.min\(^{-1}\) (52.8 gal. per minute);
• logging, groundwater quality sampling, piezometric head measuring, chemical analyzing.

Care will be given also to long term monitoring of groundwater quality evolution in pilot experiment locality. It will mainly include:
• monitoring of time behaviour of chemical equilibrium setting in groundwater in the all influenced area using sampling and logging;
• monitoring of hydraulic parameters change using pumping tests;
• monitoring of vertical distribution of chemical composition of groundwater;
• drilling of control core well, core sampling and analyzing.

The results obtained during and after the pilot experiment will be evaluated continuously from the finishing of the injection until a steady state in the Cenomanian aquifer will be reached.

After finishing the pilot experiment the following activities will be performed:
• evaluation of chemical composition of groundwater;
• evaluation of retardation of monitored components;
• evaluation of permeability changes;
• evaluation of stability of immobilization effect;
• evaluation of diffusion intrusion of immobilization medium into low permeable sediments;
• evaluation of the practical realization of in-situ immobilization in large scale;
• risk identification with the view to large scale application of in-situ immobilization as the part of complex remediation process.

The specialized numerical model based on FEM for simulation of groundwater flow and contaminant transport was developed at DIAMO for evaluation of pilot experiment of in-situ immobilization results. It follows from testing calculations that the model is fully usable for pilot experiment simulation. During the preparation phase many numerical tests will be performed. They will help to predict expected rates of change of monitored parameters to set frequency of piezometric head measurements and frequency of groundwater sampling and analyzing. The calculations will not be performed only using the above mentioned numerical model, but we will use MODFLOW™ in parallel.

4. CONCLUSION

Results of the long term monitoring of stability of geochemical equilibrium setting in the ground will be the base knowledge for prediction of possibility of targeted influencing the quality of cenomanian groundwater. They would also help to determine the parameters of future planned large scale in-situ immobilization; for example, the quality of groundwater into which
the alkaline solutions can be injected, the volume of immobilization medium needed to inject in particular parts of deposit, final quality of groundwater after injection of alkaline solutions (pH, Eh, concentration of SO$_4^{2-}$, U, Al, Fe, ...), vertical distribution of chemical composition of groundwater caused by gravity effect or by diffusion effect on the contact areas of sediments with extremely different permeability and many others.

During the pilot experiment of in-situ immobilization together with commonly used methods of monitoring the special method of zonal groundwater sampling from several vertically separated profiles will be tested.

The results and evaluation of the pilot experiment will be used as a source for future design of large scale applications of in-situ immobilization method. It will be possible to determine:

- number of wells for injection of needed volume of alkaline solutions;
- location and dimension of feeding pipelines;
- process control and monitoring system;
- optimal timing and aerial distribution of injection (using numerical models);
- prediction of total cost of remediation with use of in-situ immobilization method in combination with pump and treat technology;
- expected risks of large scale immobilization method application.

5. REFERENCES