GROUNDWATER CONTAMINATION AND SELF-PURIFICATION AT URANIUM PRODUCTION BY THE IN-SITU LEACHING PROCESS

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1. INTRODUCTION

An important condition of uranium mining industry development is environmental monitoring and ecological security of field development. The in situ leaching (ISL) method of uranium mining has less impact on the environment than traditional underground and open pit mining methods. The uranium extraction is carried out by the engineered well system that accesses a productive horizon containing the orebody. Injection wells are supplied with leaching solutions capable of selectively dissolving uraniferous minerals. The sulphuric acid ISL method is the most widespread in the world. The pregnant solution is returned to the surface by pumping wells and is discharged into the processing complex for uranium sorption extraction. Thus, mining is carried out without lifting ore to the surface through selective dissolution of uranium minerals directly within the mineralized host strata. At the same time, deposit development is not accompanied by the formation of overburden and tailings dumps, drainage of underground aquifers, formation of wastewaters from hydrometallurgical plants, etc. However, during field development of the ISL method there is some contamination of underground waters with petrogenic and technogenic substances due to the leach solution injection and its interaction with the host rock [1–3].

To monitor the productive horizon state and assess the geo-ecological effects of ISL, it is wise to use mathematical modelling methods. This is due to the process complexity occurring during ISL and the lack of information on the productive horizon state and the high cost of observation well construction. The report presents a mathematical model of uranium sulphuric acid leaching and software for forecasting the groundwater state during deposit development by the ISL method. The results of the epigenetic and predictive modelling of the change in the productive horizon state during the development of the Khokhlovsk uranium deposit by the ISL method are presented.

2. METHODS AND RESULTS

In sulphuric acid ISL, sulphuric acid is injected into the productive aquifer together with the working solution. An oxidizer can also be added to the working solution. As a result of the interaction of the leaching solutions with uranium-bearing and rock forming minerals, a sufficiently large number of different chemical elements pass into lixiviant solution. According to their characteristics and ecological significance, all polluting components can be divided into three groups. The first group includes the radionuclides of both the uranium and thorium series (U-234, U-235, U-238, Th-230, Th-232, Th-228, Ra-228, Ra-226). The second group involves items passing into processing solutions in amounts exceeding permissible limits (Be, Al, Fe, V, Cd, Zn, Pb, Ti, Tl, Ni (1–3 orders of magnitude), and Na, Ca, Mg (several multiples)). The third group consists of elements whose concentration in the lixiviant solution does not exceed the acceptable
limits (Co, Mo, Sr, Se, Hg, Ag, Sb, Te, Cu). From the point of view of groundwater pollution monitoring, only the elements in the first two groups are of interest. The movement of pollutants in the aquifer occurs as a result of convective mass transfer, hydrodynamic dispersion and molecular diffusion. In solutions, pollutants migrate in the form of ions, neutral molecules and complex compounds. The form in which there is a polluting component in the liquid phase is conditioned by the geochemical conditions. The geochemical situation is determined by the following main factors: hydrogen index (pH), oxidation–reduction potential (Eh), solution ionic strength and the presence of a large number of complex agents [4]. In the ISL process, the geochemical state of the productive horizon within the well units varies significantly. In sulphuric acid ISL, the pH values within the well unit decreases and Eh increases compared with the reservoir waters. This can change the oxidation degree of polyvalent elements, such as iron and uranium, and as a result, their migration properties are changed. Contaminants can also be in the form of complex compounds. The main ligand in sulphuric acid leaching is the sulphate ion. In the form of complex compounds, the components can increase their migratory ability or form sulphate insoluble compounds. It is not possible to describe the migration of all components of the ISL process in detail. Therefore, when creating a model, it is necessary to determine the main physical and chemical processes in order to choose a limited number of minerals and components, the descriptions of which are sufficient for adequate modelling of pollutant migration.

This paper presents a model describing the main hydrodynamic and physicochemical processes that determine pollutant behaviour in uranium sulphuric acid ISL [5]. The hydrodynamic processes include filtration of solutions within porous media and hydrodynamic dispersion. Physicochemical processes include complexation, diffusion, and single phase and multi-phase oxidation–reduction and acid base processes, sorption, mineral precipitation–dissolution and coprecipitation. The following components are contained in lixiviant solutions in significant quantities and determine the geochemical environment: $\text{Fe}^{3+}$, $\text{Fe}^{2+}$, $\text{Al}^{3+}$, $\text{Ca}^{2+}$, $\text{H}^{+}$, $\text{OH}^{-}$, $\text{K}^{+}$, $\text{Na}^{+}$, $\text{Mg}^{2+}$, $\text{S}^{2-}$ and $\text{SO}_{4}^{2-}$. These are considered in the model as well as radioactive contaminants ($\text{U}^{4+}$, $\text{UO}_{2}^{2+}$, $\text{Ra}^{2+}$, $\text{Th}^{4+}$). The model does not examine the components of the third group (Co, Mo, etc.) as these are present in the ISL process in only small amounts and do not affect the geochemical situation. Also, some of the elements of the second group present in small amounts in technological solutions are not taken into consideration. This is due to the fact that they precipitate in the form of hydroxides during acid neutralization as a result of interaction with the host rock and therefore do not extend beyond well units.

On the basis of the mathematical model generated, problem oriented software was developed to predict the migration of pollutants within underground water and to evaluate the effect on the environment as a result of uranium mining by the ISL method [6]. The software performs calculations, taking into account actual operating modes of injection and extraction wells, working solution compositions, hydrogeological structure of the productive horizon and regional groundwater flow. The software is developed in C++ programming language and intended for the use on personal computers with Microsoft Window XP-10 operating systems. Interaction with geological and technological databases is carried out through SQL enquiry.

3. DISCUSSION AND CONCLUSIONS

Groundwater contamination and self-purification of the productive horizon after uranium mining is considered in the example of the central part of the Khokhlovsk uranium deposit, which is located in the Zauralsk uranium ore district. Deposit development is carried out by Dalur, which is a part of the ARMZ uranium holding company. The simulation was carried out on the basis of the deposit’s hydrogeological model, which was constructed with the help of a mining and geological information system, in turn based on data from exploration and geophysical investigations [7]. The information on working solution compositions, injection and extraction well operating modes was imported from the technological database of the mining complex’s information system [7]. Simulations have been carried out since the beginning of operation up to the present-day but forecast calculations are for a twenty-year period after the cessation of uranium mining.
The simulation results show that, from the commencement of operations, the acid content in the lixiviant solutions within the operating unit region increases. After attaining a value of several grams per litre less than the acid concentration in the leaching solutions themselves, the acid content growth diminishes. The oxidation and reduction potential, $E_h$, which increases from $100–200$ mV to $350–450$ mV during the ISL process, behaves similarly, returning towards the original values. The total iron content also increases during the ISL process, reaching values about of $1$ g/L. The uranium content in the lixiviant solutions increases, reaches a maximum value, and then decreases as the orebody is worked out. The sulphate content in the process solutions of the operating unit reaches values of $15–20$ g/L up to mining completion. In addition, the contents of aluminium, potassium, sodium and magnesium are increased. Thus, the total mineralization of lixiviant solutions increases to $30–40$ g/L. During the ISL process, most of the lixiviant solutions remain within the operating unit region. This is due to the balanced rates of working solution injection and extraction, both for separate operational units and for the entire field. When the process solutions leave the operating unit region, acid neutralization and the formation of insoluble compounds of uranium and other pollutants takes place. As a result, the area of uranium distribution extends beyond the well units for distances of up to $20–30$ m. The sulphate ion, having the highest migration capacity, extends beyond the block outline by distances of up to $80–100$ m. To confirm the adequacy of the ISL process description by the generated model, the simulated and actual time dependences of the uranium concentration, bi- and trivalent iron, sulphuric acid and sulphate ion in productive solutions were compared. The comparison was carried out both for individual pumping and observation wells, and for the operational unit and the entire field. A good agreement of simulation results with real data confirms the adequacy of the proposed model and the correctness of the software operation.

Neutralization of the lixiviant acid and the subsequent increase in the pH of the residual process solutions have occurred for several years due to the chemical interaction with the host rock. As a result, insoluble hydroxides of uranium, iron and aluminium formed and their dissolved concentration in the residual solution decreased. At the same time, insoluble sulphate-containing minerals (gypsum, alunite, jarosite, etc.) formed. Over 20 years, the total mineralization of residual lixiviant solutions decreased from $30–40$ g/L to $7–10$ g/L. Having completed the operation, the stratigraphic lens of residual solutions remains within the operating unit region as a result of the very low rate of regional groundwater movement within the productive horizon of the Khokhlovsk uranium deposit.

The simulation results show that, in the case of uranium ISL, the region of groundwater contamination is local and situated mainly within the boundaries of the operational units. The main indicator of pollution is the sulphate ion because its content in processing solutions with sulphuric acid ISL greatly exceeds the concentrations of other components and it also has a high migration capability. Using its distribution in the underground waters, it is possible to evaluate the area of productive horizon pollution. Having completed uranium mining, there is self-cleaning of the productive horizon over a period of several decades. The reduction in pollutant concentration occurs as the result of the interaction of residual lixiviant solutions with rock forming minerals, formation of new minerals and dilution with groundwaters. The self-cleaning process speed depends on the mineralogical composition of the ore-bearing rocks in the productive horizon and the intensity of water exchange. In the case of low groundwater movement speed, the self-cleaning process takes place within a region extending a little beyond the boundaries of the well units.

REFERENCES


