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Study on Environmental Impact of Uranium Mill Tailings on Groundwater

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As more and more uranium mining and metallurgy facilities are facing decommissioning, studies on environmental impacts of uranium mill tailings are given much attention. It has become an important part of environmental impact assessment of decommissioning uranium mining and metallurgy facilities. On the basis of many researches at home and abroad, this paper generalizes previous studies, presents the deficiencies existing and the research direction in the future.

KEYWORDS: uranium mill tailings; groundwater; environmental impact

I. Brief Introduction to Uranium Tailings

Since the middle of 1940's, along with the development of nuclear weapons and nuclear industry, the need to natural uranium has been increasing, therefore, uranium mining and milling has been rapidly developed, thus giving rise to a large amount of uranium mill tailings. At present, there are about 22 countries engaged in uranium mining and milling production[1)], accumulated more than 2×10^{10}t[2)] of uranium mill tailings, most of which resulted in many adverse impact on the environment.

Tailings include diluted ore and varieties of pollutants mixed during milling, containing all uranium decay products, more than 99% of ^{230}Th and ^{226}Ra are almost concentrated in tailing mines[3)]. After uranium extracted, large amounts of ore materials become milling wastes and tailings. Liquid wastes are generally a kind of mud containing powder gangue. Such kind of mud were pumped into tailings impoundment for storage, when milling facilities operating, tailings may have little serious radiation hazards due to reasonable siting and normal operation. Even though, it may result in the release of lots of tailings and contaminants into the environment[4)] when the dam damaged or transporting piping broken, or tailings mud attacked and submerged the dam when raining.

In addition, several tailings impoundment may bring about serious leakage and contaminate ground and surface water due to unreasonably designing or long time service.

The radionuclides contained in tailings are generally more than 2-3 magnitudes greater than its natural background. Uranium tailings belong to radioactive materials of low specific activity, but having great volume of wastes, varieties of radionuclides and much higher of toxicity as well as about 36% long-life nuclides, these nuclides have formed long-term potential hazards to the environment. Uranium tailing is one of main external radiation sources which have higher γ exposure rate and stronger γ radiation because of its containing a large amount of radium. In addition, the uranium tailings may produce dust and liquid wastes to contaminate air and water, due to external influences such as wind, rain etc. Human body may receive internal exposure through inhalation of dust, drinking contaminated water or intaking contaminated food. Furthermore, except radioactive hazards, there also exist non-radioactive hazards such as heavy metal, ammonia nitrogen, sulphate and nitrate[3, 5)].

II. Research Status

At present, a wide spread attention has been paid to the environmental impact by each uranium-producing country. After many years of operation, many uranium mining and milling facilities had been facing decommissioning. A lot of research works have been developed on environmental impacts of uranium milling tailings both domestic and abroad in order to provide scientific evidence for post-decommissioning environmental management. In United State of America, the overall assessment had been performed on 24 uranium mining and milling facilities all over the country, and remediation measures had been taken to several facilities where groundwater was seriously contaminated[7)]. In China, since later start of nuclear industry, such kind of research work was relatively behind some other large uranium-production countries such as USA, Russia, Canada, Australia, etc.

Uranium tailings may contaminate groundwater by seeping from tailings impoundment. Seepage water may vertically move to water level and flow to lower reaches along with groundwater. The main factors affecting the concentration of contaminants in groundwater include in pH, Eh (redox potential), absorption, sedimentation, complexing and chelation of organic and non-organic coordination compound, precipitation and evaporation[8, 9)]. The following summarizes present research from different views.

1. Geochemical Processes related to Activity of Radionuclides and Heavy Metal

The environmental impact of metal and radionuclides is closely related to its activity. So it is important to study on the factors affecting its activity. It was shown according to

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present research that such factors as pH, Eh, absorption and sedimentation had important influence on the migration of contaminants.

pH and Eh mainly affect existence form and chemical reaction performance of contaminants in groundwater. The different existence form may decide its different nature of solubility, absorption and sedimentation. Therefore the migration characteristics are also different. When pH>6, the activity of Th was restricted due to forming of insoluble $Th(OH)_4$[10]. As for pH impact on chemical reaction such as absorption, it had been described in many papers[8, 11, 12]. Under oxidization, uranium exists in the form of UO_2^{2+}, but the activity of $U^{6+}$is more than that of U^{4+}. So the activity of uranium under oxidization increases[8, 10, 12, 13]. On the other hand, under oxidization, Fe^{3+} sedimentated in the form of $Fe(OH)_3$. The absorbability of $Fe(OH)_3$ is very strong, many metal and metalloid related with uranium mining and milling are absorbed on its surface and sedimentated[10,14]. Under reduction, the insoluble sulphides start to sedimentate, a lot of metal and metalloid are removed in the form of insoluble sulphides[14].

Absorption and sedimentation is an important mechanism of radionuclide reduction in groundwater. For example, in the water enriched with sulphates, uranium generally sedimentated along with barite and gypsum[8, 10, 15], thorium formed $Th(SO_4)_2$[8, 10]. Clay and $Fe(OH)_3$ have strong absorbability on radionuclides. As for the absorption of uranium on ferrhydrite and magnetite, many researches had been done[11, 16].

The concentration of sulphides in the tailings mainly impact the activity of metal and radionuclides[8, 10, 15]. The acid tailings are generally neutralized with lime before its piling up or entering into tailings dam during which most of uranium, other radionuclides, salt and metal are sedimentated. This is only a short-term measure. When sulphides existed in tailings, the long term effectiveness of lime became a problem. The oxidization of sulphides provides low pH conditions, and makes migration of metal and radionuclides again along with seeping water.

In aerated zone and groundwater, the complexation of organic and non-organic coordination compound is another factor affecting the migration of contaminants. UO_2^{2+}reacted with SO_4^{2-} and CO_3^{2-} and formed stable anionic complex[9], the latter is not easy to be absorbed, so the activity of uranium becomes more and more strong, but the complexation of UO_2^{2+} with organic acid reduces its activity[17].

Perry[8] and Daniel[14] carried out research on the impact on the migration of contaminants due to neutralization in ground water by geochemical model. In the areas rich in calcite, the acid seeping water in tailings were neutralized; In a very extended scope of Eh, each kind of sulphites, hydroxides of iron and of aluminum are sedimentated from solution. The oxides of iron and aluminum may also cause the sedimentaton of many contaminants related with uranium mining and milling, so many contaminants are cut down in neutralizing process.

Although it may have reached a certain place above groundwater using the average velocity of groundwater to calculate the concentration of contaminants. Actually, the concentration of contaminants here is probably lower than background due to the above-mentioned factors.

2. Environmental Impact on Groundwater

Three methods applied to study on the environmental impact on groundwater include: the first, Study on the status of groundwater contamination at site based on monitoring data; the second, Forecasting impact on groundwater by quantitative analysis model; and the third, Study on the migration of contaminants by the solution of mathematic model.

(1) Current Status of Research on Groundwater Contamination

The main assessment methods include composite index and probabilistic method. Robert[18] carried out preliminary environmental impact assessment of uranium mining and milling on groundwater in Grants district in New Mexico and put out further monitoring plan. Severe leakage occurred to tailings impoundment and waste water treated by deep-well injection, the surrounding ground water had been contaminated to different extent. It was shown in the research that the main contaminants are Ra, Se, NO_3^-. Horst[19] had conducted statistic analysis on a uranium mining and milling facilities with its ten year monitoring data in Brazil. It was found that the neutralization of waste water in tailings impoundment could reduce effectively concentrations of each kind of contaminants in seeping water. Except SO_4^{2-}, other contaminants have little impact on groundwater. In American, the risk assessment had been taken to groundwater restoration project on uranium tailings district, the comparison had been made between water quality data of the site and its background, thus, the main contaminants had been identified and its risk analysis on human body had also been carried out. As for carcinogen, the assessment is performed on its carcinogenesis risk with certain exposure dose. But for non-carcinogen, the assessment is carried on its risk. With sufficient data, the probability distribution of its risk assessment might be drawn up and could be performed on exposed probability distribution by Monte-Carlo method. During the assessment, the maximum concentration value and intake rate are taken. The main exposure pathways considered include: drinking contaminated groundwater, intaking animal and plant products contaminated by groundwater and skin contact by bathing with groundwater.

(2) Quantitative Analysis Model

Perry and Deborah[8] had studied the migration of seeping water and contaminants in uranium tailings impoundment in aerated zone and groundwater. The leak off flow was calculated by hydrological forecasting in tailings pound district. The water flow velocity was directly obtained by Darcy Law. The migration velocity of contaminants was got by water flow velocity divided by delay coefficient.

The migration velocity of contaminants might be calculated by Hajek formula：

$$V_i = \frac{v}{1+K_d\rho_b/\theta} \quad (1)$$

where, V_i:migration velocity of contaminants ;

v :water flow velocity ;

K_d:distribution coefficient ;

ρ_b:dry weight of soil ;

Θ :water content.

By calculating, it may obtain the concentration of contaminants at certain position when contaminants moved to lower reaches.

(3) Mathematic Model[20-26]

(a)Migration Model of Radionuclides in Aerated Zone

Water Flow Equation

Basic differential equation (water content as variable)

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial x}[D(\theta)\frac{\partial\theta}{\partial x}] + \frac{\partial}{\partial y}[D(\theta)\frac{\partial\theta}{\partial y}] + \frac{\partial}{\partial z}[D(\theta)\frac{\partial\theta}{\partial z}] - \frac{\partial K(\theta)}{\partial z} \quad (2)$$

where, $D(\Theta)$:diffusion coefficient ;

$K(\Theta)$:permeability coefficient.

Defining appropriate initial value and limit, getting solution to water flow equation, then flow field distribution result could be obtained.

Approximation Method: Water movement in aerated zone is a complicated process, many parameters are difficult to be precisely defined. In a certain cases, some approximations might be used, to assume that the average downward flow velocity is in direct proportion to feed rate of surface water and in inverse proportion to average volume water content, and also to assume conservatively that average volume water content is equal to capillary water content in soil, then

$$v = \frac{r}{n-n_e} \quad (3)$$

where, v :vertical average flow velocity, cm/s ;

r:feed rate of groundwater, cm/s ;

n:total porosity of soil, without dimension ;

n_e:effective porosity in soil, without dimension.

Solute Transfer Equation

$$\frac{\partial}{\partial t}(\theta c+\rho_b s) = \nabla\cdot(\theta\overline{D}\cdot\nabla c) - \nabla\cdot(\overline{V}c) - \lambda(\theta c+\rho_b s) + M \quad (4)$$

where, c:concentration of radionuclides in liquid phase, Bq·cm^{-3} ;

s:concentration of radionuclides absorbed by solid phase, g·cm^{-3} ;

$\overline{D}$:diffusion coefficient tensor cm^2·s^{-1} ;

$\overline{V}$:permeable velocity, cm·s^{-1} ;

λ :decay constant of radionuclides, s^{-1} ;

M :source all sink。

Considering that absorption and desorption is reversible inequilibrium process, so c and s meet the following relation :

$$\frac{\partial s}{\partial t} = \alpha(K_d c - s) \quad (5)$$

The above is inequilibrium absorption relation, in which α is equilibrium rate, s^{-1}. When absorption and desorption reach equilibrium,, $\frac{\partial s}{\partial t}=0$,at this time $s = K_d c$, which is equilibrium absorption model. It was shown that equilibrium absorption is a special case of inequilibrium absorption.

(b)Migration Model of Radionuclides in Aquifer

Water Flow Equation

Basic equation

$$\frac{\partial}{\partial x}(T_x\frac{\partial h}{\partial x}) + \frac{\partial}{\partial y}(T_y\frac{\partial h}{\partial y}) + \varepsilon = S\frac{\partial h}{\partial t} \quad (6)$$

As for phreatic water aquifer, $T = Kh$, S is feed water ; As for pressure bearing aquifer, $T = KM$, M is the thickness of aquifer ;S is water storing coefficient ;ε is source all sink.

Approximation method: In the simplified case, the water flow velocity along main stream could be obtained by Darcy Law.

$$u_x = -\frac{K}{n_e}\frac{\partial H}{\partial X} = -\frac{K}{n_e}\frac{\Delta H}{\Delta X} \quad (7)$$

where, u_x :flow rate of groundwater along main stream, cm/s ;

$\frac{\Delta H}{\Delta X}$:hydraulic gradient along main stream.

Solute Transfer Equation

$$R_d\frac{\partial C}{\partial t} - \nabla\cdot(\overline{D}\cdot\nabla C) + \nabla\cdot(\frac{\overline{V}C}{n_e}) + \lambda R_d C = 0 \quad (8)$$

where, R_d :delay conffieicent of radionuclides, without dimension, $R_d = 1+\frac{\rho_b}{\theta}K_d$;The meaning of the other symbols are the same as before.

III. Conclusion

In general, certain research achievements had been obtained, especially for reduction mechanism and migration law of certain radionuclides in groundwater, but there still exist many problems. It was shown from present research that the deficiencies are mainly as follows:

- The observing and monitoring at site are mostly limited to contamination status research, it needs study on long term potential hazards according to forecasting contamination trend based on long term data.
- Site experiment is one kind of research means which is a must, but monitoring of groundwater is relatively difficult and highly costly. Some essential parameters might be obtained in laboratory, but must be revised according to site information where it is actually applied. Otherwise, the conclusion derived is meaningless and unbelievable.
- Mathematic model is an important research method, but when it is used, the actual conditions must be simplified, and the more factors considered, the more difficult model

parameters obtained and equation solved. In present research, the approximation is applied to flow field, this could avoid the difficulty in obtaining parameters, but may increase calculation deviation. With the development of computer technology, the more attentions should be paid to numerical simulation of groundwater contamination by uranium tailings.

- There exists both water and air in aerated zone, but based on present papers, when studying on migration of contaminants in aerated zone, the air impacts are neglected. Only one phase is considered. Studying on the migration of radionuclides in aerated zone from points of two phases would be a front subject.
- Absorption and desorption is important mechanism of the migration of radionuclides in porous media, and is an equilibrium process. In present research, it is generally assumed that absorption and desorption instantaneously reach equilibrium. Therefore, such model could not reflect dynamic state. There is less research on radionuclides migration model in inequilibrium absorption. In addition, chemical reaction in the process of nuclide migration is pending further research.
- Once radioactive wastes are mentioned, more attentions are paid to their radioactive hazards, their non-radioactive hazards are neglected. In fact, the non-radioactive impact might be higher than their radioactive impact, which should not be neglected.
- The radiation environmental impact assessment on nuclear industry over 30 years in China had been carried out in 1980's. It was shown from radiation environmental impact assessment on 12 uranium mines and 3 nuclear research units that the impacts of uranium mines and nuclear research on groundwater are still a weak link, some of which such kind of assessment had nearly not been performed. Impacts from Radionuclides on groundwater are pending further research.

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