CHEMICAL ENGINEERING TRANSACTIONS

VOL. 71, 2018

A publication of
AIDIC
The Italian Association
of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Xiantang Zhang, Songrong Qian, Jianmin Xu

ISBN 978-88-95608-68-6; **ISSN** 2283-9216

DOI: 10.3303/CET1871131

Solute Transport in Deep Geology Disposal of High Radioactive Waste

Qiuling Wang, Shuqin Ni
School of Physics and Electronic Engineering, Fuyang Normal Colloge, Fuyang 236037, China
qiulingwang39102@163.com

This paper evaluates and analyzes the safety of solute transport as applicable to the deep geology ground water situation at the disposal of high-level radioactive wastes. This basis is consideration for basic safety assessment. Given the coupling effect from T-H-M-C, the EOS1 and EOS2 can simulate the interactions between groundwater and backfill material when brine and freshwater intrude in the vicinity of field, respectively. With the granite in a certain area in the northwest territories the migration media, the 1D multi-channel solute migration model simulates how the nuclides Cs-134, Co-57 and Tc-99more often involved in the study of granite in China migrate in the media. The results show that Cs-134 can migrate at the fastest rate under the specific conditions, while Tc. 99 is at the slowest rate.

1. Introduction

Along with continuous development of the nuclear industry, how to safely dispose of the high radioactive wastes is one of the important topics in the industry. Today, China has witnessed the rapid development in the economy, bringing with it the increasing demand for social energy resources. Our country strongly supports the development of nuclear power industry in order to eliminate the energy supply and demand contradiction. as early as the 1970s, the high-level radioactive wastes have aroused the common concern in the world. Many scholars at home and abroad devoted to studying the nuclides. In the disposal of high radioactive wastes, the atomization may be reached only after the evolution of eons. Therefore, how to dispose of radioactive wastes has become a staggering problem in nuclear technologies. For the deep geology disposal of high radioactive wastes, it is required to fully consider some spiny issues, for example, surrounding environmental protection and project safety in an attempt to avoid nuclear radiation and pollution. In the solute transport process of deep geology disposal of high radioactive wastes, the safety of the disposal of the radioactive wastes should be considered.

Given the above, this paper evaluates and analyzes the safety of deep geology disposal system for high radioactive wastes, and throws light on what is the possible impact of solute transport on the near field groundwater. According to the relevant studies of foreign nuclear fuel development organizations, the nuclides of deep geology disposal of this high-level radioactive waste are determined to inquire into the feasibility of solute transport.

2. Literature review

The near field of the deep geological system of high-level waste includes high-level waste solidified body, packaging container, engineering backfill material, that is, the engineering barrier system of the warehouse. Before the waste is disposed, the groundwater in the warehouse is generally drained and ventilated to allow the warehouse to receive waste in a dry state (Alexander et al., 2015). When the warehouse is closed, as time goes by, the groundwater will eventually return to the treasury and begin to erode and destroy the engineering barrier until the engineering barrier finally disintegrates. At this time, various substances in the solidified body of the waste are leached by the groundwater, released to the near field and gradually migrated to the far field. For various physical and chemical effects in the near field, temperature changes, fluid flow, contaminant transport, hydration and ice decomposition, mechanical deformation, physical-chemical exchange, etc. are mainly considered. In fact, these processes are interactive and influential. However, it is very difficult to

Please cite this article as: Wang Q., Ni S., 2018, Solute transport in deep geology disposal of high radioactive waste, Chemical Engineering Transactions, 71, 781-786 DOI:10.3303/CET1871131

combine all of the above phenomena in the same numerical model (Chen et al., 2015). For the various processes in the near field, many scholars have carried out related experimental research and a large number of numerical simulation studies. Avis et al. studied the composition of pore water in bentonite. The results show that these components are mainly controlled by the decomposition or precipitation of less active components in bentonite (Avis et al., 2014). Lopez-Fernandez et al. conducted diffusion experiments on the migration characteristics of solute such as (HTO, Se, Cs, Sr) in high-compressed bentonite used in the FEBEX project, and obtained the partition coefficients and diffusion coefficients of HTO, Se, Cs and Sr. Moreover, analytical methods and numerical analysis methods were used to explain these experimental results (Lopez-Fernandez et al., 2015).

However, the duration of these experiments is too short compared to the time required to secure the repository. Therefore, many scholars focus on numerical simulation. Mcevoy et al. simulated the interaction between bentonite and super alkaline water. The simulation results show the complex front of mineral dissolution and precipitation and its sensitivity to temperature, solution composition, dissolution mechanism of montmorillonite (Mcevoy et al., 2016). Mohammed et al. studied the reaction process in partially saturated bentonite. The results show that the chemical properties of pore water depend on the soluble matter in bentonite. In addition, the change in porosity is less affected by the precipitation and dissolution of minerals, and is mainly affected by the swelling of bentonite (Mohammed et al., 2015). Reijonen and Alexander used KIRMAT to simulate the geochemical and cation exchanges that consider the diffusion of chemical elements in the engineering barrier under the conditions of the repository. The results show that the engineering barriers associated with groundwater vary greatly after 10,000 years. The most important chemical processes are cation exchange, illitization and saponification. The change in expansion pressure is estimated by the volume balance in the saturated engineering barrier (Reijonen and Alexander, 2015).

The far field of the high-level waste repository is the natural geological body around the repository, i.e., the geological barrier system. This is not only the last line of defense for harmful radioactive substances in high-level radioactive waste, but also an important line of defense. Therefore, the selection of suitable surrounding rock geological bodies is an important prerequisite for ensuring the safety of high-level waste disposal repositories (Chopra et al., 2015). At present, combined with the status quo of high-level waste disposal, relevant laboratory experiments have been conducted for single cracks. The effect of gap width variation, surface roughness and surface adsorption on solute migration in single cracks was studied, and the relevant parameters describing solute transport were determined by tracer experiments. Wang conducted tracer experiments in natural fissures and used X-ray imaging techniques to track solute transport. The migration parameters under the variation of the gap width were measured to obtain the effect of the change of the gap width on the solute transport (Wang, 2014). Internationally, on the basis of laboratory experiments, combined with the needs of geological disposal of radioactive waste, a series of field experiments were carried out. The typical tracer field migration experiment in earlier fractured rock masses was carried out in Stripa, Sweden, which lasted for three years and revealed the main features of water flow and solute transport in fractured rock masses. Then, a diffusion experiment was carried out at a depth of 360 meters in the mine. The experimental results show that the soluble matter diffuses into the matrix domain under natural stress conditions, and the diffusion coefficient and hydraulic gradient are obtained (Follin et al., 2014).

In summary, the above studies were carried out in a single fracture, and then developed to study the flow and solute transport in the plane cracks and network cracks composed of plane cracks. Therefore, the characteristics of radioactive material release and the random distribution of cracks in the high-level waste repository are analyzed. The Laplace transform and multi-path transport model are used to study the solute transport law in single fracture and fracture networks. The nuclide migration simulation of the deep geological disposal system for high level radioactive waste was carried out. It provides a basis for the safety evaluation of high level waste disposal.

3. Method

3.1 Safety assessment of deep geology disposal system for high radioactive wastes

For the high radioactive waste, due to a long time and spatial span of its geology disposal system, there are many uncertain factors included in the safety evaluation. it is thus rather cumbersome to evaluate the safety on this system using the traditional models. Ultimately, the deep geology disposal of high radioactive waste aims to ensure its long-term safety. It is predicted that the risk to the human health and the environment should not be greater than the current acceptable level whenever possible in the future. Therefore, the safety evaluation generally includes the following three components: 1) determine what phenomena can cause or are likely to cause the release of radioactive materials, or can affect the release rate, or play an effect on the migration rate of radioactive materials into the environment. 2) estimate the probability that these phenomena occur and quantitatively express what’s impact they play on the nuclear waste disposal system. 3)calculate the radiation consequences from the release of radioactive materials, i.e. individual and collective doses, and then re-estimate what's the risk of it to the human health, in order to ultimately assess what are the impacts of it on human health and the environment. The Organization for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA) proposed to analyze the sequences by using the scenario analysis model in 1991. Based on the experiment, the physical/chemical model and the mathematical model are built to conduct a safety evaluation on the high-level radioactive waste disposal system. (see Fig.1).

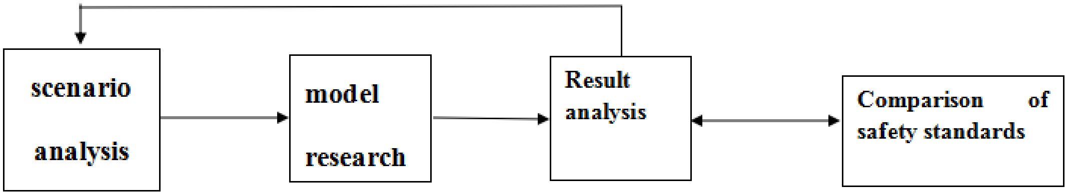

Figure 1: Safety assessment method for geological disposal system of high-level radioactive waste

3.2 Simulation of groundwater and solute transport in near field

The near-field of the high radioactive waste disposal depot refers to its engineering barrier part, namely the waste drum, the backfill materials, the containment material and the like. Based on the classification of analytic situations as described in Chapter 2, it is considered that the release of radioactive materials from waste drums to backfill materials, geological barriers and even to biospheres is mainly attributed to the groundwater dissolution. Especially under the deep geology disposal conditions for high radioactive wastes, when the temperature and pressure rise up, the dissolution process will be greatly accelerated.

In the EOSI program, the basic conservation equation in the integral form is directly used to make spatial discretization, rather than the equations (3.8), that is, the integral finite difference method. This method was first applied to fluid flow and heat transfer by Edwards (1972). As the integral finite difference method can discretize the study area using irregular element meshes, and more apply to the interaction between the flow, migration and fluid multi-regional heterogeneity media and intermedia. Time discretization uses a full implicit first-order backward finite difference. Discretized strongly coupled nonlinear algebraic equations are sync solved from iterations by the Newton. Raphson. In the process of simulation computation, the time step can be automatically adjusted according to the convergence speed in iterative process. Regardless of the number of fluid phases and the nature of the components presented, the multiphase fluid and heat flow have the same mathematical form. The structure of EOSI program mode is shown in Fig. 2.

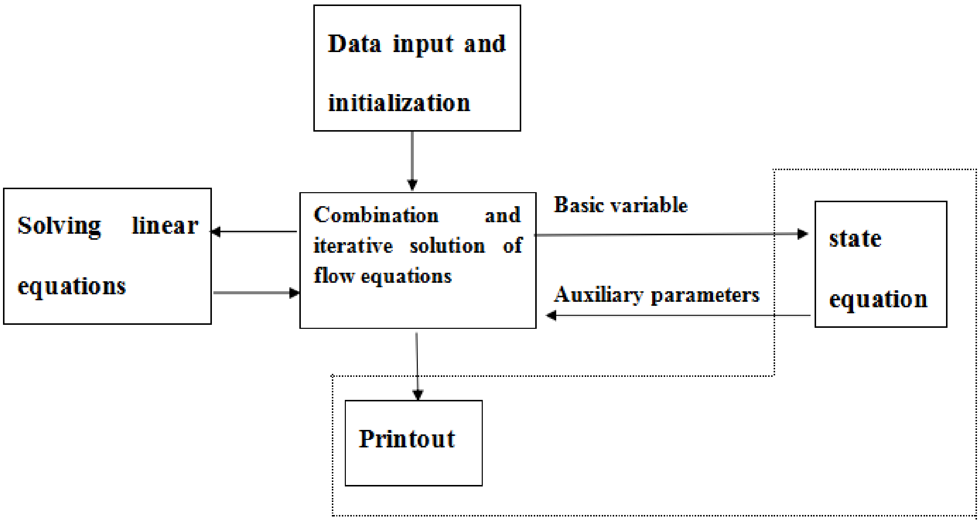

Figure 2: EOSI program pattern structure

The basic simulation parameters mainly include the density, porosity, thermal conductivity, etc., of the backfill materials. The initial conditions: average pressure 1．654×1 07Pa, volumetric water content 8.4%, the sodium chloride as a percentage of the initial pore water is 0.07% in the whole backfill material, and the soil water potential at the top is ψ=0m (Akira Ito et al.， 2003). At the top boundary, there is a certain concentration of aqueous solution (where the water weight accounts for 96.7%, the mass fraction of NaCl is 3.3%), while the bottom and the periphery are closed, i.e. there is no water and solute transport, as shown in Table 1.

Table 1: basic parameters, initial and boundary condition parameters

Basic parameters	Backfill material density	1600kg/m3	porosity	0.403
	Thermal conductivity	2.5W/m·℃	permeability	2.0×10-9m2
	Specific heat of rock	1443.72J/kg·℃		
initial condition	Top temperature	80℃	Bottom temperature	100℃
	initial temperature	80℃	pressure	1.654×107Pa
	volumetric water content	8.4%	NaCl concentration	0.07%
boundary condition	Top NaCl concentration	3.3wt%	Top initial water potential	Ψ=0
	Bottom and surrounding	close		

3.3 Simulation of solute transport in single fracture media

According to the reference case adopted by the Japan Nuclear Cycle Development Institute (JNC) in the study of the migration of nuclides in the deep geology disposal depot of high radioactive wastes, it is assumed that the nuclide released from the engineering barrieris released into the biosphere via the Major Water-Conducting Fault (MWCF), and the three radionuclides, Th-229, Cs-135 and Se-79, control the total dose of radioactivity released (JNC, 2000c). Three species of nuclides are used as simulation objects, and the relevant parameters are shown in Table 2.

Table 2: considered the corresponding parameters of nuclides

nuclide	Decay constant	partition coefficient	Hysteresis coefficient
Th-229	9.443×10-5	1	132001
Cs-135	3.01×10-7	0.05	6601
Se-79	1.066×10-5	0.01	1321

4. Analysis of results

The migration characteristics of nuclides Cs-134, Co-57 and Tc-99 in granite single fracture (i.e. single fractured granite column) are studied experimentally, and the relevant diffusion coefficients and hysteresis coefficients are obtained. When selecting parameters, for the sake of safety, the values in favor of nuclide migration are chosen, such as hysteresis coefficient, matrix diffusion coefficient, etc., take the minimum values in the literature, as shown in Table 3:

Table 3: considered nuclides and their characteristics

Nuclides and carriers	half life	Decay constant	Hysteresis coefficient	Matrix diffusion coefficient
Cs-134（CSCI）	2.06	0.336	2791.12	6.96×10-7
Tc-99（NH4TcO4）	2.13E5	3.254E-6	761.4	8.537×10-6
Co-57（CoCI2）	0.7446	0.931	419.8	1.31×10-5

In the same position, the relative concentration of nuclide increases over time during the migration. Comparing the nuclides, it is known that the relative concentration of Tc-99 in the fracture domain is the minimum at the same position and time, that is to say, it also migrates at the slowest rate in the one medium, while Cs-134 is the fastest; comparing these data, it can also be found that, under given conditions, the relative concentrations of these nuclides in the fracture domain are not distributed in terms of hysteresis coefficients at the same simulation time and position, which shows that the relative concentration of nuclide in the fracture domain also has a bearing on the diffusion coefficient in its half-life period and matrix domain. (the relative concentrations of Cs-134 as a function of simulation time are shown in Table 4)

Table 4: relative concentration values of Cs-134 at different simulation time

	100m	200m	400m	800m	100m	1200m
T=1×106y	0.7143	0.5213	0.2832	0.1013	0.07064	0.0539
T=1×105y	0.5275	0.2896	0.1014	0.03199	0.02296	0.01734
T=1×104y	0.3101	0.1108	0.03124	8.903E-3	5.208E-3	3.015E-3

5. Conclusion

This paper focuses on the “multi-barrier system” used in the geological disposal of high radioactive waste. A simulation runs on how nuclides in the geological barrier against groundwater when it returns to the depot migrate. The results from this simulation show that the steam movement caused by the temperature gradient seems not obvious, and no salt precipitation occurs everywhere throughout the whole simulation process. Under the conditions discussed herein, the calcite precipitation does not cause great changes in porosity in the near-field backfill material. Ming Yh-229 migrates at the slowest rate, at the shortest distance, and diffuses to the shallowest layer in the matrix domain; at 0.1x the maximum migration distance, the relative concentrations of individual nuclides tend to be stable in the next 4×106 years. Comparing the simulation results, it is found that under the simulated conditions, the hysteresis coefficient (distribution coefficient) has a negative correlation with the nuclide migration distance and the diffusion depth.

The mechanism of the solute transport process in the high radioactive waste from disposal system underlies the establishment of its conceptual and mathematical models. In particular, the solute transport in near-field involves multi-field coupling, how to build a mathematical model that incorporates various effects for consideration based on the typical groundwater situation requires further study in the future.

Acknowledgement

1. Project of National Natural Science Foundation of China: theoretical study on electronic structure and spectral properties of halide molecules of zinc and cadmium. Project number: 11604052

2. Anhui platform talent introduction and support project: soil improvement of sand ginger black soil in Northern Anhui Province.

3. National Undergraduate Innovation Training Project: application of biochar to improve hydraulic properties of sandy black soil. Project number: 201710371060

Reference

Alexander W.R., Reijonen H.M., Mckinley I.G., 2015, Natural analogues: studies of geological processes relevant to radioactive waste disposal in deep geological repositories, Swiss Journal of Geosciences, 108(1), 1-26, DOI: 10.1007/s00015-015-0187-y

Avis J., Suckling P., Calder N., Walsh R., Humphreys P., King F., 2014, T2ggm: a coupled gas generation model for deep geologic disposal of radioactive waste, Nuclear Technology, 187(2), 175-187, DOI: 10.13182/nt13-83

Chen K.K., 2018, Study on the Mineral Materials' Adsorption Properties of Radioactive Chemical Elements, Chemical Engineering Transactions, 66, 139-144, DOI: 10.3303/CET1866024

Chen L., Wang J., Zong Z.H., Liu J., Su R., Guo Y.H., 2015, A new rock mass classification system q hlw, for high-level radioactive waste disposal, Engineering Geology, 190(2), 33-51, DOI: 10.1016/j.enggeo.2015.02.006

Chopra M., Nair R.N., Sunny F., Sharma D.N., 2015, Migration of radionuclides from a high-level radioactive waste repository in deep geological formations, Environmental Earth Sciences, 73(4), 1757-1768, DOI: 10.1007/s12665-014-3525-x

Follin S., Hartley L., Rhén I., Jackson P., Joyce S., Roberts D., Swift B., 2014, A methodology to constrain the parameters of a hydrogeological discrete fracture network model for sparsely fractured crystalline rock, exemplified by data from the proposed high-level nuclear waste repository site at Forsmark, Sweden, Hydrogeology Journal, 22(2), 313-331, DOI: 10.1007/s10040-013-1080-2

Lopez-Fernandez M., Cherkouk A., Vilchez-Vargas R., Jauregui R., Pieper D., Boon N., 2015, Bacterial diversity in bentonites, engineered barrier for deep geological disposal of radioactive wastes, Microbial Ecology, 70(4), 922, DOI: 10.1007/s00248-015-0630-7

Mcevoy F.M., Schofield D.I., Shaw R.P., Norris S., 2016, Tectonic and climatic considerations for deep geological disposal of radioactive waste: a uk perspective, Science of the Total Environment, 571, 507-521, DOI: 10.1016/j.scitotenv.2016.07.018

Mohammed M.H., Pusch R., Warr L., Kasbohm J., Knutsson S., 2015, Interaction of clay and concrete relevant to the deep disposal of high-level radioactive waste, Applied Clay Science, 118(38), 178-187, DOI: 10.1016/j.clay.2015.08.008

Reijonen H.M., Alexander W.R., 2015, Bentonite analogue research related to geological disposal of radioactive waste: current status and outlook, Swiss Journal of Geosciences, 108(1), 101-110, DOI: 10.1007/s00015-015-0185-0

Wang J., 2014, On area-specific underground research laboratory for geological disposal of high-level radioactive waste in china, Chinese Journal of rock mechanics and geotechnical engineering, 6(2), 99-104, DOI: 10.1016/j.jrmge.2014.01.002