

Longevity Issues in Swelling Clay as a Buffer Material for a HLW Repository

고준위폐기물처분장 완충재물질로서 팽윤성 점토의 장기건전성과 주요 고려사항

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(Received July 18, 2007/Approved August 7, 2007)

Abstract

A swelling clay should remain physically and chemically stable for a long time to perform its functions as a buffer material of a high-level waste (HLW) repository. The longevity issues in the swelling clay were reviewed to evaluate their importance in the performance of a repository. The review results suggest that an elevated temperature due to decay heat, groundwater chemistry, high pH environment by concrete, organic matter and microbes, radiation, and mechanical disturbance might significantly affect the long-term performance of a swelling clay as a buffer material. This paper will be used as basic informations to design the swelling clay buffer for a HLW repository.

Key words : HLW repository, engineered barrier, buffer, swelling clay, long-term performance.

요 약

고준위폐기물처분장의 완충재 물질로 사용되는 팽윤성 점토는 방벽재로서 그 기능을 제대로 발휘하기 위해 오랫동안 물리, 화학적으로 안정해야 한다. 팽윤성 점토의 장기건전성 관련인자들을 검토하고, 처분장 성능에 대한 각 인자의 중요성을 평가하였다. 검토결과, 붕괴열에 의한 온도상승, 지하수 화학, 콘크리트에 의한 pH 증가, 유기물과 미생물, 방사선 조사 및 기계적 교란은 완충재물질로서 팽윤성 점토의 장기건전성에 중요한 인자임을 확인하였다. 본 연구는 고준위폐기물 처분장에서 팽윤성 점토의 완충재 설계를 위한 기초자료로 유용하게 활용될 것이다.

중심단어 : 고준위폐기물처분장, 공학적 방벽, 완충재, 팽윤성 점토, 장기건전성

I. Introduction

The concept for a disposal of HLWs in Korea is based upon a multi barrier system composed of engineered barriers and their surrounding plutonic rock. A repository is constructed in bedrock of several hundred meters in depth below the ground surface. The engineered barrier system (EBS) is made up of man-made barriers placed within the repository. This EBS consists of HLW disposal containers, and barriers constructed using swelling clays that is similar to the configuration considered by many other countries. In this configuration (Figure 1) the HLW are encapsulated in disposal containers, which are deposited into boreholes on the floor of the emplacement rooms. The gap between the container and the wall of a borehole is then filled with a buffer material and the inside space of the emplacement rooms with a backfill material.

A swelling clay, in the HLW repository, is favorably considered as a buffer material because it may minimize a water flux into the repository and restrict the release of radionuclides into the host environment. Also it may dissipate the decay heat from waste into the surrounding rock to avoid the possibility of a thermal stress on the container and a peak temperature resulting in a loss of the desirable functions of the buffer, and support the container and waste from external mechanical stress. However, the barrier performance of a swelling clay may be affected by a temperature elevation due to a radioactive decay heat and geochemical conditions, and consequently its longevity may be degraded when it is exposed to those environments for a long time. Therefore, a review of the longevity issues in a swelling clay is essential to evaluate the long-term barrier performance of a swelling clay and to design the buffer of a HLW repository.

The present paper, in this connection, reviewed the longevity issues of a swelling clay (e.g., elevated temperature, groundwater chemistry, high pH environment by concrete, organic matter and microbes, radiation, mechanical disturbance) to evaluate how they may affect the long-term performance of the swelling clay used for the buffer of a HLW repository.

II. Elevated Temperature

The decay of radionuclides in HLW wastes produces a significant amount of heat, which leads to an elevation of the temperature in a swelling clay buffer. As an elevated temperature adversely affects the integrity of a swelling clay as a barrier material, it should be maintained below a design value. In a Canadian concept[1], the maximum temperatures of the container and buffer material are limited to below 150 °C and 100 °C, respectively, and, in Sweden [2] and Japan[3], the maximum temperature of the buffer material is limited to 100 °C. This limitation of the maximum temperature would be set to avoid a mineralogical alternation of the buffer material which might occur at an excessively high temperature.

When a repository is closed, the swelling clay, due to an elevated temperature, undergoes a dry heating, hydrothermal or water vapor interaction,

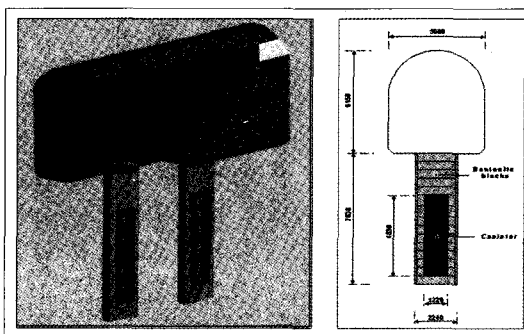


Fig. 1. Schematic picture of the engineered barrier system of Korean Reference Disposal System.

or a combination of all three depending on the rate at which the groundwater saturates the repository. As the temperature in the swelling clay is increased, the bound water of a hydration and the structural water in the clay might be driven off and then the swelling and sorption capacity of the clay might be decreased, resulting in a reduction of its radionuclide retardation capacity[4]. Allen et al.[5] investigated the stability of bentonite exposed to a dry heating at an atmospheric pressure. Their results showed no permanent change in the bentonites which were heated to temperatures as high as 370 °C for 340 days. Eberl[6] reported that the bentonite exposed to a dry heating showed a considerable dehydration at a temperature of 200 °C and a loss of structural water at 300 °C. When the bentonite was cooled and treated with water, the dehydration proved to be fully reversible and the bentonite swelled with no permanent change.

However, the hydration of a swelling clay due to an elevation of the temperature is more important in a hydrothermal condition than in a dry heating. Two alteration mechanisms[1] have been identified in the hydrothermal conditions: an illitization by which the swelling clay (smectite) is chemically converted into non-swelling clay (illite) and a silicification by which silica in the groundwater, which is either dissolved from the minerals in the rock or from the clay itself, is precipitated and simultaneously cements the swelling clay mass. An illitization is known to dominate a hydrothermal reaction and to reduce its swelling capacity resulting in a loss of its ability to self-seal any cracks in a swelling clay buffer[1, 4]. These changes depend on environmental conditions. Pusch[7] and Kamei et al.[8] conducted natural analog studies to clarify the temperature, time history and chemical conditions. The former study showed that bentonite existed without an illitization for about a thousand years at 100-150 °C

in the Kinnekule bentonite deposit. The latter study concluded that an illitization did not occur in an environment similar to seawater which had been cooled down for 2 million years from 160 °C to 100 °C. However, when a swelling clay is hydrothermally exposed to a potassium-containing solution condition, there may be an enhanced illitization of the swelling clay. It is because a supply of potassium is essential for a conversion of smectite into illite and this controls the illitization rate. Also this illitization affects a radionuclide sorption. The interlayer cations are fixed in illite and, as a result, its cation exchange capacity is lower than that for unconverted smectite, This may result in a degraded sorption capacity of an illitized clay.

III. Groundwater Chemistry

The chemistry of a groundwater intruded into a repository is changed by chemical reactions with the components of the engineered barriers. Studies[9-13] have shown that the groundwater was buffered to a high pH and low Eh and that there was a noticeable change in the concentration of some ions by taking into account the reactions among the groundwater, the swelling clay buffer, the container corrosion products, and the sealing materials. The chemistry of the porewater in a water-saturated clay buffer is important for its radionuclide-retarding capacity. The ionic concentration, pH, and Eh of groundwater influences the chemical reactions and processes such as a dissolution/precipitation, complexation, oxidation/reduction reactions and sorption/diffusion processes, which consequently affect the transport of radionuclides in a swelling clay buffer. The groundwater chemistry also affects the stability of a swelling clay under disposal conditions. As mentioned above, given the availability of *Al* and *K* in a pore solution (provided by the dissolution of

co-existing phases and/or present in the solution flowing through the clays), smectite which is a major component of a swelling clay undergoes a continuous series of transformations through illite/smectite (I/S) to illite[3]. However, the presence of Na, Ca, and Mg in a groundwater slows down this conversion reaction. Oscarson et al.[14] and Roberson and Lahann[15] explained this as follows: the K is readily dehydrated in the interlayer region of 2:1 layered clays because of its relatively low hydration energy. It thus promotes a collapse of the layers of high-charge clays such as illite. The cations such as Na, Ca, and Mg compete with K for exchange sites on the swelling clay and, consequently, a collapse of these high-charge clays is inhibited in the solutions with low $K/(Na + Ca + Mg)$ ratios. Johnston and Miller[16] examined the effect of a solution pH on the stability of smectite. It was revealed in his examination that the smectite-to-illite conversion was dominant in the near-neutral pH region; in the mildly acid region, there was an extensive formation of aluminum hydroxyl interlayers in the swelling clay; and in the alkaline region, framework silicates (zeolites and feldspars) were produced.

IV. High pH Environment by Concrete

Radioactive waste repositories require concrete for plugging or construction purposes, the concrete being in direct contact with the swelling clay. In this case, the performance of a swelling clay may be affected by two main chemical processes: a high pH produced by the concrete or rather by the dissolution of $Ca(OH)_2$ in the pore water of the cement mortar, which attacks the smectite crystal lattices, and the Ca which migrates from the pore water of the cement into the clay buffer, where it occupies exchange sites and then influences the inter-particle spacing.

Pusch[17] stated in his report that the latter process leads to stronger bonds and smaller distances between the smectite lamellae in the aggregates, by which the swelling potential was reduced and the permeability is increased to 2 to 5 times that for the latter process. And he added that the former process involved the transfer of $Ca(OH)_2$ in the solution into the clay, and thus, if there was an excess of $Ca(OH)_2$ from the cement mortar, the reaction between smectite and $Ca(OH)_2$ created framework silicates like zeolites and feldspars. These may yield no swelling potential and thus adversely affect the desirable swelling and transport properties of compacted clay buffers. However, if a highly compacted clay is contacted with concrete, the limited available pore space and the rather extreme swelling pressure exerted by the swelling clay will probably prevent a noticeable growth of the zeolite nuclei and consequently the permeability will be reduced. Oscarson[18] examined the effect of $Ca(OH)_2$ (portlandite) on the swelling and hydraulic properties of compacted bentonite. His results indicated that, although additions of up to 5 wt.% $Ca(OH)_2$ to compacted bentonite decreased the swelling pressure and increased the hydraulic conductivity of the bentonite, when compared to those of untreated bentonite, the compacted bentonite still had a hydraulic conductivity that was low enough to ensure that a molecular diffusion would be the principal transport mechanism through the bentonite. However, he added this was particularly true if low-pH, high-performance concretes were used. It has been observed in laboratory tests, that smectite dissolved in a high-pH environment and formed C-S-H gel and zeolite as secondary minerals[19], which resulted in an decrease of the swelling potential and thus the hydraulic conductivity. Therefore, in a disposal concept where large amount of cementitious materials are used for waste forms, fillings,

structure, and lining, it is necessary to consider the chemical interaction between the swelling clay and cement which will be one of the important phenomena when one evaluates the long-term performance of an engineered barrier system.

V. Organic Matter and Microbes

A clay may generally contain organic matters which will act as a nutrient to a microorganism, and it also contains other nutrients such as nitrogen, phosphate, and sulphate[20, 21]. The direct effects of a microbiological action on a swelling clay buffer may not be important for several hundred years immediately after a closure of a repository, because the radiation level and the temperature around the waste container are high.

However, the complexation of radionuclides by an organic complexing agent[22, 23] and under a low pH condition by sulphate and iron oxidizing bacteria may increase the mobility of the radionuclides in a swelling clay, and reduce the stability of high-level wastes. The organic matters in a swelling clay also adversely affect the corrosion of copper containers[24]. The corrosion of copper takes place in the presence of corrosive substances, and the corrosive substances are dissolved oxygen under oxidizing conditions and dissolved sulfide under reducing conditions. The sulfide is supplied from the clay and the groundwater, and theoretically it is formed by a microbiological reduction of sulfates in the groundwater and clay. And a high content of organic matter may have a bad influence on the physical and mechanical properties of a swelling clay[25, 26].

It is desirable to maintain the organic matter content in a swelling clay buffer as low as possible. Sweden[24] and Canada[1] recommended that the maximum allowable content of organic matter in a swelling clay buffer should be 0.5 wt.%, below

which the effect of organic matter on the physical and mechanical properties of a swelling clay might be negligible and a migration of the microbes is prevented by a filtration effect of the compacted swelling clay, because the size of most microorganisms is of the order of one micron[25, 26].

VI. Radiation

The radiation field within the buffer of a repository is a function of the time and the distance from a waste, and also depends on the shielding effect of a waste container. This radiation is almost exclusively gamma ray and the contribution of alpha and beta rays and neutron is relatively insignificant, as long as the container remains intact. But in the event of a container failure after a long time, the alpha and beta radiations from released radionuclides may significantly contribute to the radiation field. The radiation from high-level wastes has an influence on a swelling clay via two kinds of processes: one is radiation damage of the crystalline structure of a swelling clay and the other is a change of the pH and Eh of the pore water by radiolysis. And the barrier properties of the swelling clay may be affected by these processes.

Krumhansl[27] exposed a bentonite sample to 3×10^{10} rad of Co-60 gamma radiation at room temperature. The optical and XRD results of the irradiated bentonite showed no crystallographic changes. The bentonite sample heated to 300 °C and then exposed to a total absorbed dose of 3.5×10^9 rad from a Co-60 source, showed no detectable change in its clay particle morphology, chemical composition, crystallography, and swelling ability[28, 29] who investigated the effect of radiation on the hydraulic conductivity and swelling potential of bentonite which was exposed to a dose of 9.5×10^9 rad from a Co-60 source. They showed

no change in the hydraulic conductivity and a slight decrease in the swelling potential.

Mayfield and Baker[20] exposed Boom clay (mainly composed of illite, smectite and vermiculate) to a gamma ray of $3 \times 10^9 - 3 \times 10^{10}$ rad, and then conducted a sorption experiment to investigate the influence of a radiation on a radionuclide sorption onto the Boom clay. The results showed a slight decrease in the sorption capacity of Cs, Sr and Eu, which was said to be due to a decomposition of the organic matter in the clay by a radiation. A possibility that an alpha radiation may damage the crystal structure of buffer constituents such as smectite and affects its sorption properties[30, 31] has been reported. Haire and Beall[21] studied the effects of high alpha doses on bentonite using Es-252 under the condition of total absorbed alpha doses which ranged from 4.8×10^9 to 4.8×10^{11} rad. TEM analysis indicated that the crystallinity of the bentonite was lost at a total absorbed dose of 4.8×10^{11} rad. But its effect on a radionuclide migration was negligible because at most only a few percent of the total smectite may be affected and the altered product also sorbs nuclides[3, 31].

The radiolysis of pore water due to radiation from vitrified wastes generates oxidizing agents such as H_2O_2 , and thus affects the redox conditions. If an oxidation front develops in a swelling clay, the solubility and sorption of the radionuclides in that clay may change. However, even if these oxidizing agents are generated, they will be buffered by a sufficient amount of reducing materials including the overpack and its corrosion products, and the pyrite in the clay buffer[3, 32]. And, if local oxidizing regions which mobilize some radionuclides occur, they will be precipitated in adjacent reducing regions. Accordingly, the existence of oxidizing agents will have no significant effect on a radionuclide migration in a swelling clay

buffer.

VII. Mechanical Disturbance

The mechanical disturbances in a swelling clay buffer include a hydrogen gas generation, the erosion of clay particles, and the corrosion of carbon steel overpacks. The corrosion of carbon steel under reducing environments produces hydrogen gas, which dissolves in a groundwater and then diffuses through a water-saturated swelling clay buffer into a surrounding rock or accumulates within a clay buffer. The stress generated by the accumulation of hydrogen gas is relieved by the deformation of a clay buffer and a surrounding host rock. However, since the equilibrium pressure of the corrosion reaction is high[33], if the gas permeability of the clay buffer is assumed to be low, then there is the possibility that the host rock will be influenced by a pressure increase which exceeds the ground pressure. Pusch and Forsberg[34] and PNC[3] showed in their study that compacted bentonite (MX-80) was found to be permeable to hydrogen gas at a pressure between 10-90 % of the swelling pressure, and the gas permeability of the bentonite was 100 to 1000 times the diffusion rate at which the hydrogen gas was accumulated in the clay buffer and thus escaped through the fluid phase in the pores of the clay. In this case, a hydrogen gas production may accelerate a radionuclide transport.

A swelling clay has a plasticity and stress-buffering capacity against a rock pressure. Due to its plasticity, it has a self-sealing capacity in the form of intruding into fractures in a surrounding host rock. However, if the following process occurs, the favorable properties of the swelling clay as a buffer material are damaged: the front of the intruding swelling clay forms a soft gel, the soft gel is eroded by a groundwater flow, the amount of the eroded

soft gel is replenished from the deposition hole, and in the end a significant amount of the swelling clay in the deposition hole keeps on decreasing[35]. Pusch and Forberg's experiments[36] showed that the water flow rate required to remove a bentonite particle was 10^{-4} m/s and the intrusion of the bentonite stopped after the fractures were filled with a certain amount of the bentonite, depending on the width of the fractures.

A corrosion of the carbon steel overpacks leads to a volume expansion (i.e. corrosive swelling), because the specific gravity of the corrosion products is smaller than that of the carbon steel. This may lead to a failure of a buffer material, resulting in a degradation of its mechanical stability.

VIII. Conclusions

The longevity of a swelling clay was dependent upon environmental factors such as the elevated temperature, groundwater chemistry, high pH environment by concrete, organic matter and microbes, radiation, mechanical disturbance in a HLW repository. The elevated temperature was an important variable for a hydrothermal reaction and thereby degraded the integrity of the swelling clay. The groundwater chemistry was changed by contacting with EBS components, which affected the stability of the swelling clay and the physical and chemical reactions in the swelling clay. Large amount of cementitious materials yielded an extremely high pH, under which the swelling pressure of the clay was decreased and thus its hydraulic conductivity was increased. The organic matter and microbes, when in a low temperature and radiation field, might increase the mobility of radionuclides and reduce the stability of high-level wastes. Also they adversely affect the corrosion of copper containers. The radiation effect on the swelling potential and the radionuclide-retarding

capacity of the swelling clay was minor. The mechanical disturbances such a hydrogen gas generation, the erosion of clay particles, and the corrosion of carbon steel overpacks had a rather significant influence on the integrity of the swelling clay as a barrier material. This paper suggests that the longevity issues of a swelling clay are important factors when evaluating the long-term performance, and designing a swelling clay buffer, for a HLW repository.

IX. Acknowledgement

This work has been performed under the Nuclear R&D Program by the Ministry of Science and Technology.

X. References

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