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Analysis of Functional Criteria for Buffer Material in a High-level Radioactive Waste Repository

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Abstract

This study is intended to analyze the requirements of a buffer material that is one of the major components of the engineered barriers in a high-level radioactive waste repository. The characteristics of potential materials for the buffer in the repository were analyzed and a candidate material was selected. And, based on the current knowledge and the information from various sources, the requirements of a buffer material were evaluated. Finally its quantitative functional criteria on the generic viewpoint has been recommended to be supplied as a guideline for the development of the reference disposal concept and the related buffer material in Korea. The criteria are composed of seven major items, such as hydraulic conductivity, retardation capacity, swelling potential and swelling pressure, thermal conductivity, longevity, organic matter content, and mechanical properties.

1. Introduction

The high-level radioactive waste repository is usually room-and-pillar design constructed in the bedrock at a depth of several hundred meters. Waste containers are deposited in an array of large-diameter boreholes drilled on the floors of emplacement rooms. After the emplacement of a container the gap between the container and the wall of borehole is filled with a buffer material, and then the room is filled with backfill material (Fig. 1). Following closure of the repository, the borehole and room and the surrounding rock will eventually become water-saturated. In such circumstance, the buffer and backfill should control and minimize water flux into the repository, and restrict the release of radionuclides into the host environment. The buffer and backfill have to dissipate the decay heat from waste into the surrounding rock to

Key words : buffer, buffer material, functional criteria, high-level waste, repository, engineered barriers, bentonite

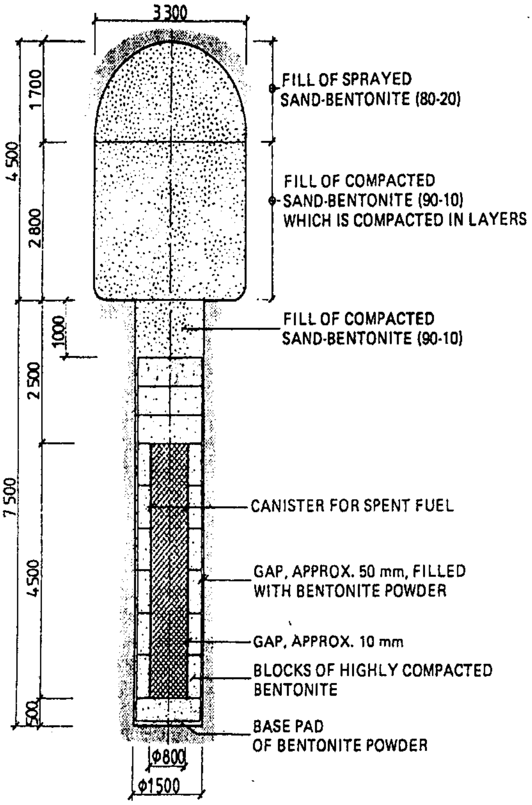

Fig. 1. Cross-Section of the Disposal Room in the High-level Radioactive Waste Repository [6]

avoid the possibility of thermal stress on the container and the high temperature resulting in the loss of the desirable functions of the buffer, and also have to support the container and waste from the external mechanical stress.

To meet these functions, the required properties of buffer material are low hydraulic conductivity, high retention capacity, high swelling potential and low swelling pressure, good thermal conductivity, good mechanical properties and longevity [1-3]. To develop the well qualified-buffer material, however, the quantitative values satisfying with functional criteria rather than qualitative requirements are necessary. To establish such quantitative functional criteria is not easy and requires the repository concept and the site-specific information. In Canada, some quantitative criteria are recommended for Reference Buffer Material (RBM) [4], and in USA, the functional criteria for buffer are not specified, but only the overall performance requirements for the engineered barriers composed of waste form, container and buffer are described in 10 CFR 60 [5]. The NRC criteria requires that "waste packages provide substantially complete containment of spent fuel for 300 to 1000 years, and radionuclide release rate after 1000-year containment should not exceed one part in 100,000/y of the inventory at 1000 years, at the engineered barrier boundary", In Sweden [6] and Japan[7], some quantitative functional criteria of a buffer material are suggested. In Korea, as the disposal concept for high level waste and the candidate disposal sites have not been selected yet, it is difficult to establish the detailed quantitative functional criteria for a buffer material. However to perform efficiently and systematically the related R&D programme and to optimize the development process of a buffer material, it is necessary to establish the quantitative functional criteria for a buffer material.

In this paper, it is intented to analyze the requirements of a buffer material, and to recommend the quantitative functional criteria on the generic viewpoint without considering the disposal concept and site-specific conditions.

2. Survey of Potential Buffer Materials

To select the candidate material suitable for the buffer, the major properties of potential materials have been investigated in several countries, and clay-based and cement-based materials are proposed for preferable material

[8-10]. Use of cement-based material as buffer elevates the pH of groundwater to 12.5, the value for saturated lime solution, and the corrosion of container metals may be enhanced in these highly alkaline solutions than in the pH range from 7 to 10. Even corrosion-resistant metals such as titanium and titanium alloys for example have become more proven to localized crevice corrosion at high pH solution [11]. At high pH, the surface of copper would be passivated [12], and with the high Cl^- content in groundwater, passivated copper surface would be more likely to degrade by pitting than by uniform corrosion. High pH could also enhance the dissolution rate of vitrified reprocessing waste [13]. Hence clay-based materials which could adjust the pH of groundwater to neutral and slightly alkaline values ($7<pH<9$) are preferred as buffer.

Buffer material should limit the release of radionuclide from the breached containers to the rock. Clays have higher capacity to sorb cationic radionuclides from solution as compared to cement, and high swelling capacity to seal the void and the fracture in buffer. The long-term stability of clays under the high radiation and thermal field as natural material have also been proven. Considering these properties, the clays are preferred over cement and concrete as buffer material. Therefore the discussion was focused to clays and the possibility of use for buffer material has been investigated.

Clays can be defined as particulate materials with particle size less than 2μm. Clay could be classified into several major mineral groups with respect to the crystal structure and the chemical composition. Most common clay minerals are kaolinite, illite, and montmorillonite [14]. These clay minerals are evaluated for the hydraulic conductivity and radionuclide sorption properties which are most of the important parameters

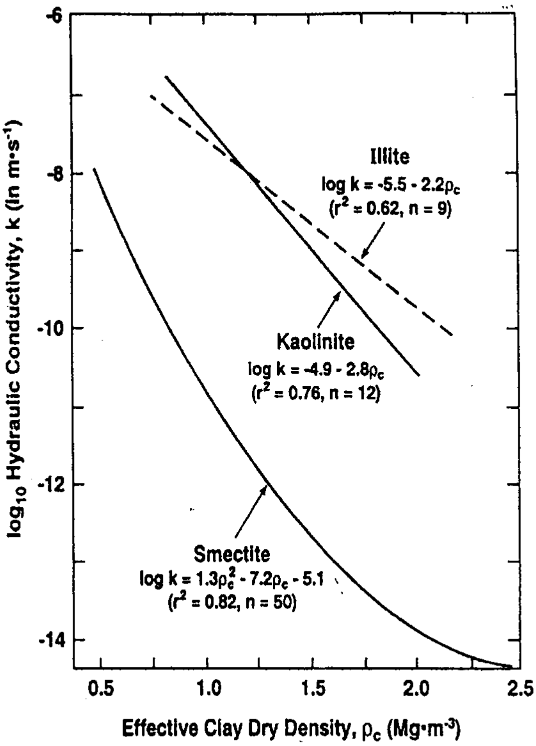

Fig. 2. Hydraulic Conductivities for Major Clay Minerals [4]

from the viewpoint of the function of the buffer material.

Hydraulic Conductivity : The typical relationships between hydraulic conductivity and dry density for kaolinite, illite and smectite(montmorillonite) are shown in Fig. 2 [4]. As shown, even at the dry density lower than 0.8 Mg/m^3, smectite (montmorillonite) has a hydraulic conductivity as low as 10^{-9} m/s, and at the same dry density, its hydraulic conductivities are several orders of magnitude less than those of illite and kaolinite [4,15].

Sorption of Radionuclides : Buffers are ultimately intended to retard the release of radionuclides. To evaluate their ability to perform this function, the cation exchange capacity and the distribution coefficient of

Table 1. Radionuclide K_d Values for Major Clay Minerals

Material	Cation exchange capacity (meq/100g)	K_d values (mL/g)				
		Th	Tc	Am	Pu	Sr
Illite	15-40	$120\text{–}4\times10^2$	0.0–11	-	129	2-200
Kaolinite	5-10	$600\text{-–}\sim2\times10^5$	0.4–3	4.3	352	55-257
Montmorillonite	70-100	$4\times10^{-2}\text{–}2\times10^2$	1.5–0.2	34	630	10-700

radionuclides in the buffer material are required. The specific surface areas of three clay minerals are increased in order of kaolinite (20 m^2/g), illite (80 m^2/g) and montmorillonite (800 m^2/g) [16]. Values of the cation exchange capacity and the distribution coefficient (K_d) of three clay minerals for a number of different radionuclides are presented in Table 1 [16]. The data show that the cation exchange capacity and K_d are higher for montmorillonite than for kaolinite and illite., and montmorillonite is better than other two clay minerals on the viewpoint of retardation of radionuclide release.

These properties make montmorillonite particularly attractive for buffer material of a repository. Bentonite, the clay which originates from volcanic ash, has therefore been suggested as a candidate buffer material in a high-level waste repository. It is mainly composed of montmorillonite, and also contains minor amounts of quartz, feldspar, hallosite, clinoptilorite, and cristobalite. Bentonites are classified into two groups, Na-bentonite and Ca-bentonite depending on the type of exchangeable cations existing in interlayers of bentonite particle.

The types of bentonite produced in every country are different. The abundant deposits of Na-bentonite rest in USA and Canada, and Ca-bentonites are deposited in Europe [16]. Na-bentonite is generally considered to be preferable for the buffer material because of high swelling potential and high sorption capacity. But every country considers the use of domestic bentonite without distinction of types of bentonite for buffer material in a high-level waste repository because the required quantities of bentonite would be large. The candidate buffer materials for high-level waste repository considered in several countries are presented in Table 2.

3. Analysis of Requirements for Buffer Material

3.1. Hydraulic Conductivity

Hydraulic conductivity should be low so that water movement from the surrounding rock to the waste container is restricted, resulting in that the diffusion be principal mechanism of radionuclide release through the buffer. If hydraulic conductivity is high, the principal mechanism of radionuclide release could be advection rather than diffusion. Gillham and Cherry[17] showed that if the hydraulic conductivity is less than 10^{-8} m/s when the hydraulic gradient and the porosity are 10^{-2} and 0.35, respectively that are the typical values for host rock in deep granite, radionuclide migration would be controlled by diffusion. Therefore the

Table 2. Reference Buffer Materials for High-level Waste Repository

Country	Reference buffer material	Dry density (Mg/m^3)	Compaction method
Canada	50:50 sand : Na-bentonite	~ 1.67	In situ
Sweden	Na-bentonite	1.8-2.0	Precompacted blocks
Finland	Na-bentonite	1.8-2.0	Precompacted blocks
France	Ca-smectite	1.4-1.8	Precompacted blocks
Switzerland	Ca-bentonite	1.65-1.75	Precompacted blocks
Spain	Ca-bentonite	1.4-1.8	Precompacted blocks
Japan	Na-bentonite	1.8	Precompacted blocks /In situ

Table 3. Hydraulic Conductivity of Compacted Bentonite

Dry density (Mg/m^3)	Hydraulic conductivity (m/sec)	Bentonite	Reference
2.1	4.6×10^{-15} - 6.7×10^{-15}	Na-bentonite	18
1.7	1.5×10^{-14}	Na-bentonite (MX-80)	19
1.60- 1.75 1.4	1.0×10^{-13} - 5.0×10^{-13} 1.0×10^{-12}	Ca-bentonite	20
1.3 - 1.5	1.0×10^{-13} - 1.0×10^{-12}	Na-bentonite (Avonlea)	67

hydraulic conductivity of buffer material should be, at least, less than 10^{-9} m/s considering safety margin. The hydraulic conductivity of impermeable deep bedrock such as granite rock mass is in order of 10^{-10} to 10^{-12} m/s [4,6], and if the hydraulic conductivity of buffer material is less than these values, the groundwater flowing in the surrounding rock will bypass and not penetrate through the buffer. Such a low hydraulic conductivity can be obtained by compacting bentonite to high density.

Many investigators [18-20,67] have reported that the hydraulic conductivities in Na- and Ca-bentonite with the dry densities of 2.0 to 1.3 Mg/m^3 are in the range of 1.0×10^{-14} to 1.0×10^{-12} m/s (Table 3). The hydraulic conductivity for Canadian RBM(50:50 sand-bentonite mixture) with the dry density of 1.7 Mg/m^3 is less than 1×10^{-11} m/s [4]. The hydraulic conductivity of Ca-bentonite originated from the eastern coast of Korea is about 1×10^{-12} m/s in case of the dry density of 1.6 Mg/m^3 [21]. Hence the requirement of hydraulic conductivity for diffusional mass transport could be achieved easily, if the bentonite is compacted to the dry density of 1.6 Mg/m^3.

As a repository in Korea is expected to be located in deep crystalline bedrock, the hydraulic conductivity of a buffer material should be less than 1×10^{-12} m/s in order to bypass the groundwater around the deposition holes. However this tentative criteria would be reviewed and modified when the site characterization for the repository is performed

Table 4. Apparent Diffusion Coefficients for Some Radinuclides in Compacted Bentonite

Radionuclides	Bentonite density (Mg/m^3)	D_a (m^2/sec)	Bentonite	Reference
C (as HCO_3)	1.4 - 1.6	5.1×10^{-11} - 8.2×10^{-11}	Avonlea	22
Cl	1.6 - 1.8	1.1×10^{-10} - 2.3×10^{-10}	Avonlea	23
	1.5 - 2.0	3.0×10^{-11} - 9.4×10^{-11}	MX-80	24,25
Sr	1.6 - 2.0	2.1×10^{-11} - 5.2×10^{-11}	Avonlea, MX-80	23,26
	1.5 - 2.0	4.3×10^{-12} - 9.1×10^{-12}	Kunipa	27,28,29
Tc	1.3 - 1.4	1.6×10^{-12} - 1.2×10^{-10}	Avonlea(oxidizing)	30,31
	1.6 - 2.0	1.0×10^{-11} - 1.8×10^{-10}	Kunigel	27,32
	1.3 - 1.4	6.3×10^{-12} - 7.0×10^{-12}	Avonlea(Reducing)	30
	2.0	8.4×10^{-14} - 5.3×10^{-11}	MX-80(Tc(VII,VIII))	33,34,35
I	1.5 - 1.6	2.6×10^{-11} - 2.1×10^{-10}	Avonlea	36
	1.4 - 2.0	2.4×10^{-11} - 8.5×10^{-11}	Kunipa	27
Cs	1.4 - 2.0	4.0×10^{-13} - 7.8×10^{-12}	Kunipa, Kunigel	32,37
	1.8 - 2.0	2.5×10^{-13} - 8.5×10^{-13}	MX-80	28,29
Am	1.4 - 1.6	9.0×10^{-16} - 2.8×10^{-15}	Kunigel, Kunipa	27,32
	1.9 - 2.0	2.0×10^{-15} - 1.4×10^{-14}	MX-80	34,35,38
U	1.6 - 2.0	4.6×10^{-14} - 8.2×10^{-13}	MX-80	24,26,34,35
Np	1.4 - 2.0	3.0×10^{-14} - 1.5×10^{-13}	Kunigel, Kunipa	27,32
Pu	2.0	1.9×10^{-15} - 7.0×10^{-14}	MX-80	34,35,38

in future.

3.2. Radionuclide Retention Capacity

The buffer materials should have high equilibrium distribution coefficients (K_d) and low diffusion coefficients for radionuclides in order to retard radionuclide release from waste to surrounding rock after the failure of waste container.

The ionic diffusion coefficient in the porous medium such as a bentonite could be expressed as an effective diffusion coefficient (D_e) as follows:

$$D_e = D_o \tau \varepsilon \qquad (1)$$

where D_o is a molecular diffusion coefficient in free water, τ and ε are tortuosity, and porosity respectively. The K_d represents the chemical retardation capacity of medium due to the sorption, and D_e means the physical retardation capacity of medium due to the complexity of geometric structure of medium. Because it is however difficult to separate the chemical retardation and the physical retardation, the apparent diffusion coefficient (D_a) is usually adapted to represent the overall retardation capacity of medium. D_a is defined as follows;

$$D_a = D_o \tau \varepsilon / (\varepsilon + K_d\ \rho_d) \qquad (2)$$

where ρ_d is the dry density of medium.

The apparent diffusion coefficients of

radionuclides depend largely on the type of and the dry density of bentonite. The values of apparent diffusion coefficients of several radionuclides in the compacted bentonites with dry densities of 1.4 Mg/m^3 to 2.0 Mg/m^3, which are collected from the literatures, are represented in Table 4 [22-38]. As shown in the table, there are not big differences between data in the range of dry density of 1.4 Mg/m^3 to 2.0 Mg/m^3, and the apparent diffusion coefficients of cationic nuclides excluding strontium and actinides are less than 10^{-11} m^2/s, and those of strontium and anionic nuclides are less than 10^{-10} m^2/s and 10^{-9} m^2/s respectively. The safety assessment results for Canadian [4] and Swedish disposal concept [6] showed that the disposal safety can be assured when the apparent diffusion coefficients of cationic nuclides including actinides and anionic nuclides are less than 10^{-11} m^2/s and 10^{-9} m^2/s, respectively.

If Korea chooses the similar disposal concept, the values of apparent diffusion coefficients of radionuclides in buffer material should be less than 10^{-11} m^2/s for cationic nuclides including actinides, 10^{-10} m^2/s for strontium and 10^{-9} m^2/s for anionic nuclides in order to assure the disposal safety. These are however the tentative criteria and would be reviewed and modified when the final disposal concept is fixed in future.

3.3. Swelling Properties

Buffer material should have a high swelling potential to fill the void in deposition holes and the fracture in the surrounding rock and should have a low swelling pressure to avoid the over-burden on the waste container and the surrounding rock when it is saturated with groundwater after closure of the repository.

Bentonite has a very high swelling potential and swelling pressure and the bentonite with exchangeable cation of sodium shows the highest swelling potential. The swelling pressure increases with increasing dry density of bentonite. Westsik et al. [18] reported that the swelling pressure of Na-bentonite with dry density of 2.1 Mg/m^3 was 57 to 58 MPa. The swelling pressures of Wyoming bentonite, MX-80 and Avonlea bentonite with dry density of 1.75 Mg/m^3 were 13 MPa [39] and 16 MPa [40], respectively, and those of Ca-bentonites with dry density of 1.75 Mg/m^3 were 11 MPa [20] and 13 MPa [41]. The high swelling pressure developed by compacted bentonite would act as imposed load on the waste container, and to reduce the swelling pressure, the use of bentonite-sand mixture as a buffer material has been suggested. The swelling pressure of bentonite-sand mixture depends on the ratio of bentonite and sand, and increases with increasing bentonite content. The swelling pressure of 50:50 sand/Na-bentonite mixture with dry density of 1.75 Mg/m^3 was much lower than that of 100% bentonite with the same dry density, and the maximum swelling pressure was about 2 MPa [4].

The criteria for swelling pressure depends on the repository concept and the material of waste container in order to avoid the failure of the container. In Canada, the candidate material for waste container is titanium and the maximum allowable swelling pressure of RBM with dry density of 1.75 Mg/m^3 comprising 50% sand and 50% Avonlea bentonite is 2.5 MPa. On the other hand, the swelling pressure of Wyoming bentonite with dry density of 2.0 Mg/m^3 which has been suggested as a buffer material in Sweden is about 50 MPa, and it may not be easy to sustain the waste container without failure under such a high swelling pressure. In Switzerland [42] and Japan [7], the

Table 5. Thermal Conductivity of Compacted Bentonite

Dry density (Mg/m^3)	Thermal conductivity (W/m °K)	Moisture content (%)	Bentonite	Reference
1.7	0.6 - 0.8	13 - 15	Na-bentonite (Ibeco)	44
1.8	0.9 - 1.2	17 - 22		
1.9	0.9 - 1.2	21 -25		
2.0	1.3	28		
2.1	1.0 - 1.2	5 - 14	Na-bentonite (Wyoming)	6
1.4	0.4 - 0.7	5 - 15	Na-bentonite (Kunigel V1)	7
1.6	0.6 - 1.4	5 - 24		
1.8	0.8 - 1.6	5 - 17		
2.0	1.0 - 1.4	4 - 11		
2.1	0.6 - 0.9	air dry	Na-bentonite	43
			Na-bentonite	46
1.0 - 1.1	0.8 - 1.2	35 - 45	(Black Hill)	
1.25	0.7 - 1.1	20 - 30	(Pembina)	
1.5 -1.7	1.7 - 1.8	12 - 18	(Sealbond)	

intermediate dry density of ~ 1.8 Mg/m^3 was chosen for buffer material, and in this case the swelling pressure is about 20 MPa. The swelling pressure of Ca-bentonite originated from the eastern coast of Korea is about 15 MPa in case of the dry density of 1.8 Mg/m^3 [68].

Therefore, considering the integrity of waste container and the sealing ability for void and fracture, 20 MPa, the swelling pressure of Ca-bentonite with dry density of ~1.8 Mg/m^3 plus the safety margine was chosen as a maximum allowable swelling pressure for buffer material. This value would be continued to be revised and optimised by the development of a buffer material and the design concept of waste container.

3.4. Thermal Conductivity

The thermal conductivity of buffer should be similar to that of the host rock to conduct efficiently heat from the waste to the surrounding rock. If the decay heat is not dissipated efficiently, the heat would be accumulated in the container and in the interface of container and buffer, which affects adversely on the long-term integrity of waste container and waste form as well as buffer. The physical and chemical properties of buffer could be changed by the elevation of temperature. Bentonite buffer consists of various inorganic elements as a porous media of which thermal conductivity is not good. Therefore it is desirable to improve the thermal conductivity of buffer as far as possible.

Several investigations [6,7,43-46] on the thermal conductivities of compacted bentonites have been reported and the results are summarized in Table 5. The thermal conductivities of bentonites with dry density of 1.0~2.1 Mg/m^3 are in the rang of 0.4~1.8 W/m °K, and increase with increasing dry density of bentonite but largely depend on the moisture content. The thermal analyses of the repository based on the KBS-3 [6] and PNC [7] design concepts indicate that the expected maximum temperature in buffer is below 100 ℃ when the thermal conductivity of buffer material is greater than 1.0 W/m °K. As shown in Table 5, this value of thermal conductivity can be obtained if the dry density and the moisture content of bentonite are greater than 1.8 Mg/m^3 and 20 %, respectively. As the buffer will be fully saturated with groundwater after closure of the repository, 1.0 W/m °K is the reasonably achievable thermal conductivity of bentonite buffer material. To improve thermal conductivity and thermal capacity, the use of sand-bentonite mixture was suggested [45,46]. Suzuki et al. [7] reported that the thermal conductivities of Kunigel V1 bentonite with dry density of 1.4~1.8 Mg/m^3 were increased to 1.0~2.0 W/m °K with increasing sand content to 30 wt%.

Therefore the minimum thermal conductivity of buffer material was set at 1.0 W/m° K to maintain the temperature of buffer below 100 ℃.

3.5. Long-term Integrity

The function of buffer material should be maintained over the whole isolation period of waste under disposal environment. The decay of radionuclide produces significant amounts of heat, which could lead to increase in temperature of the buffer material. As high temperature affect adversely on the integrity of buffer material, the temperature of buffer material should be maintained below the predetermined value. Thermal modeling based on the BWIP(Basalt Waste Isolation Project) design indicates that the expected peak temperature in the buffer material is approximately 200℃ shortly after repository closure, and will be maintained above 150℃ for approximately 500 years [47]. But, in Canadian concept [4], the maximum temperatures of container and buffer material are limited below 150℃ and 100℃, respectively, and, in Sweden [6] and Japan [7], the maximum temperature of buffer material is also set at 100℃. This limitation of maximum temperature would avoid the mineralogical alternation of bentonite that might occur at excessively high temperature.

After closure of the repository, depending on the rate at which groundwater saturates the repository, the buffer material may experience dry heating, hydrothermal or water vapor interaction, or combination of all three. As the temperature of bentonite is increased, the bound water of hydration and structural water in the bentonite might be driven off and the sorption and swelling capacity of the bentonite are decreased resulting in the reduction of radionuclide retardation capacity [48]. Allen et al. [49] investigated the stability of bentonite exposed to dry heating at atmosphere pressure, and the results showed no permanent change in the bentonites which were heated to temperature as high as 370℃ for as long as 340 days. Eberl [50] reported that the bentonites exposed to dry heating showed the considerable dehydration at temperature of 200℃ and the lose of structural water at 300℃. When the bentonite was cooled and treated with water, the dehydration proved fully to be reversible and bentonite swelled showing no permanent change. The hydration of clay due to the increase of temperature is known to be more important in hydrothermal condition rather than in dry heating [48]. Two mechanisms have been identified through which a bentonite-based buffer might lose its swelling capacity [4]. These are illitization, in which the swelling montmorillonite in bentonite can be chemically converted into non-swelling clay (such as illite), and silicification, in which silica in the groundwater, which is either dissolved from the minerals in the rock or from the clay itself, is precipitated in and cements the clay buffer mass. These reactions might reduce the swelling capacity resulting the loss of the ability to self-seal any cracks in the buffer. Both illitization and silicification have been studied extensively in Canada and Sweden [4,51,52]. It has been shown that provided temperature do not exceed about 120℃, illitization and silicification are not a concern.

Allen et al. [49] and Wood [53] investigated the reaction between bentonite and groundwater at temperature of 200℃ and 300℃ and pressure of 30 MPa. The 60-days experiment yielded little change in the bentonite, other than minor dissolution and limited substitution of calcium and iron for sodium in the montmorillonite. The results of Wyoming bentonite plus groundwater experiment were also similar to those discussed above [53]. When 25:75 mixture of bentonite and sand or crushed basalt were exposed to water vapor at temperature as high as 260℃, the hydraulic conductivities of the mixture were increased sharply, but below 150℃ the hydraulic conductivity changed negligibly [54,55].

Therefore considering the silicification, the illitization and the reduction of integrity due to the hydrothermal reaction between bentonite and groundwater, it is desirable that the temperature of buffer keeps less than 100℃ in order to maintain the long-term integrity.

3.6. Radiation Stability

The radiation field in buffer is a function of time and distance from waste, and also depends on the shielding effect of waste container. This radiation will be almost exclusively gamma ray as long as the container remains intact, and the contribution of alpha and beta radiation and the neutron flux will be relatively insignificant. But in the event of container failure after long time, the alpha and beta radiation emitted from the radionuclides leached out from waste will contribute the radiation field. The radiation from high-level waste may have an influence on buffer material by two kinds of mechanism; one is radiation damage on the crystalline structure of buffer material and the other is the change of pH and Eh of pore water by radiolysis. Both can affect the properties of buffer material.

Krumhansl[56] exposed a bentonite sample to 3×10^{10} rad of Co-60 gamma radiation at room temperature. Optical and XRD studies of the irradiated material showed no crystallographic changes. The bentonite sample heated to 300℃ and exposed to a total absorbed dose of 3.5×10^{9} rad from Co-60 source showed no detectable change in clay particle morphology, chemical composition, crystallography, and swelling ability [57]. Bradley et al. [58] investigated the radiation effect on the hydraulic conductivity and swelling potential of the bentonite exposed to a dose of 9.5×10^{9} rad from Co-60 source, and the results showed no change in hydraulic conductivity and slight decrease in swelling potential. To investigate the influence of radiation on the radionuclide sorption on clay mineral, the Boom clay (mainly composed of illite, smectite and vermiculate) was exposed to a gamma ray of 3×10^{9} - 3×10^{10} rad before sorption experiment [59]. The results showed a slight decrease in sorption capacity of Cs, Sr and Eu and it might be due to the decomposition of organic matter in the clay by radiation. Haire and Beall [60] studied the effects of high alpha doses on the bentonite using Es-252 under the condition of total absorbed alpha doses ranged from 4.8×10^{9} to 4.8×10^{11} rad. TEM analysis indicated that the crystallinity of bentonite was lost only at the total absorbed dose of 4.8×10^{11} rad.

However the expected radiation dose rate is $\sim$ 2.5×10^{5} rad/y at the surface of disposal container for PWR spent fuel [61]. And so, in the period of 105 years after the disposition of bentonite with the fuel, the radiation damage of bentonite should be negligible. Therefore the radiation criteria for a buffer material would not be necessary at present.

3.7. Organic Content

Clay contains generally organic matter, and the organic content of the bentonite for buffer material should be low. The organic matter can act as a nutrient to the microorganism. In the clay deposit there are also other nutrients (such as nitrogen, phosphate, and sulphate) [59]. For several hundred years immediately after closure of the repository, because the radiation level and the temperature around the waste container are high, the direct effects of microbiological action on the buffer may not be important. But the formation of soluble complexed radionuclides by organic complexing agent [62,63] and of low pH condition by sulphate and iron oxidizing bacteria would increase the mobility of radionuclide in buffer, and reduce the stability of waste. The organic matters contained in buffer can affect adversely on the corrosion of copper container [6]. The corrosion of copper can take place in the presence of corrosive substance, and the corrosive substance is a dissolved oxygen under oxidizing condition and is a dissolved sulfide under reducing condition. Sulfide can be supplied from the bentonite and the groundwater, and can theoretically be formed by the microbiological reduction of sulfate in groundwater and bentonite. This reduction requires the presence of degradable organic matter. At present it is impossible to predict exactly the effects of microorganisms on the buffer material. The high content of organic matter may also have a bad influence on the physical and mechanical properties of clay minerals [64,65]. It is therefore desirable to maintain the organic matter content in buffer material as low as possible.

In Sweden [6] and Canada [4], the maximum allowable content of organic carbon in the buffer material is 0.5 weight % and below this value, the effects of organic matter on the physical and mechanical properties of clay mineral might be negligible [64,65]. The organic carbon content of buffer material should not therefore exceed 0.5 weight %.

3.8. Mechanical Properties

Buffer material should be required to mechanically support the waste container without significant deformation resulting in the formation of void either under the weight of the container and other imposed loads. Also the cracks due to shrinkage of bentonite should not occur when the temperature is increased because of decay heat. The shear strength of clay depends on the mineralogical composition, the size and shape of clay particle and the content of non-clay minerals [66], and the dominant factor is the thickness of adsorbed water layer on clay surface. As the adsorbed water layer of bentonite is thicker than those of kaolinite and illite, the shear strength of bentonite is relatively low. The consolidation properties should be also considered for buffer material. But these mechanical properties depend directly on the type and weight of waste container, the waste container emplacement concept, and the design concept of the repository which have not defined yet. Therefore the criteria for mechanical properties are not quantified in this paper.

4. Recommended Functional Criteria for Buffer Material

From the previous discussion, it is intented to establish the functional criteria for buffer material. These are however not final criteria and should be reviewed and if necessary, modified when the design concept of the repository is fixed. Here the criteria are intented to be quantified as far as possible, but some criteria which are difficult to be quantified at present are expressed in the qualitative form.

4.1. General Requirements

- hydraulic conductivity : The buffer material should have low hydraulic conductivity so that water movement from the surrounding rock to the waste container is restricted and the diffusion should be principal mechanism of radionuclide release through the buffer.
- radionuclide retention capacity : The buffer material should retard the release of radionuclide into the surrounding host rock after the failure of waste container.
- swelling potential : The buffer material should have a high swelling potential to seal the cracks in host rock and the voids in buffer. It should not also generate excessive swelling pressure to the system.
- mechanical properties : The buffer material should retain its physical integrity without significant deformation resulting the formation of void either under the weight of the container and other imposed loads.
- radiation safety : The radiation safety criteria are not necessary to define.

4.2. Functional Criteria

- hydraulic conductivity : The hydraulic conductivity of buffer material should be below 1×10^{-12} m/s.
- retention capacity : The apparent diffusion coefficients of cationic nuclides (excluding strontium) and actinides in buffer material should be less than 10^{-11} m^2/s. Those of strontium and anionic nuclides should be less than 10^{-10} m^2/s and 10^{-9} m^2/s respectively.
- swelling pressure : The swelling pressure should be less than 20 MPa to avoid the excessive external load on the canister.
- thermal conductivity : The thermal conductivity of buffer material should be higher than 1.0 W/m °K to effectively dissipate heat from the decay of radionuclides in the wastes.
- long-term integrity : The peak temperature of bentonite based buffer material should be less than 100℃ to assure its long-term integrity.
- organic content : The organic carbon content should be less than 0.5 weight % to minimize any bad influence on phyical and chemical properties of clay minerals and the formation of any soluble complexes of radionuclides.
- mechanical properties : The buffer material should have good mechanical properties such as compressive strength and consolidation properties in order to mechanically support the waste container without significant deformation.

5. Summary

A clay is a preferable material for the buffer, and bentonite that the predominant clay mineral is montmorillonites was proven to be most suitable for because of its high swelling potential and ability to sorb the radionuclides.

The requirements and the major characteristics of a buffer material in a high-level radioactive waste repository were analyzed, and then its quantitative functional criteria has been recommended to be supplied as a guideline for the development of the refernce disposal concept and the related buffer material in Korea. The criteria are composed of seven major items, such as hydraulic conductivity, retardation capacity, swelling potential and swelling pressure, thermal conductivity, longevity, organic content, and mechanical properties. As the functional criteria are, however, closely related to the design concept of the repository, this criteria may be reviewed and revised when the repository design concept is fixed.

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