Buffer Retardation Experiment for Radionuclides Under the Elevated Temperature Conditions: In-situ Synchrotron X-ray Powder Diffraction Study for the Korean Bentonite Under the Dry Conditions

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1. Introduction

The radioactive wastes including the spent nuclear fuels generate decay heats for a very long time. Thus, to host such wastes, the thermal criteria for the hosting buffer materials for the engineered barrier system must be set for the disposal repository. In most countries, it is set to below 100°C, due to the possible transformation of the smectite to illite which will result in the loss of the properties requested for the buffer.

If the thermal criteria for the maximum temperature for the hosting buffer system increases, the disposal density for the high-level radioactive wastes will increase dramatically [1]. This will certainly help certain nations with a high population density and a small area available for the disposal. Thus, understanding of the performance of the buffer system under the elevated temperatures are of central interest. We have developed a strategy and methodology for the Korean bentonite retardation experiments to validate the performance of the buffer materials under the elevated temperatures, especially from a viewpoint of a buffer retardation of radionuclides [2]. This includes (i) set up a rational condition for the Korean bentonite as a buffer material with time (Fig. 1), (ii) the characterization of the buffer in the absence and/or presence of the groundwater under the elevated temperatures, and (iii) the testing of the buffer retardation for the radionuclides of interest.

Here, we have investigated the hydration and dehydration reactions of the Korean bentonite to provide the characteristics of the buffer under the the elevated temperature conditions. More specifically, we have conducted in-situ X-ray powder diffraction experiments as a function of increasing temperature for the Korean bentonite (e.g., the Gyeongju bentonite).

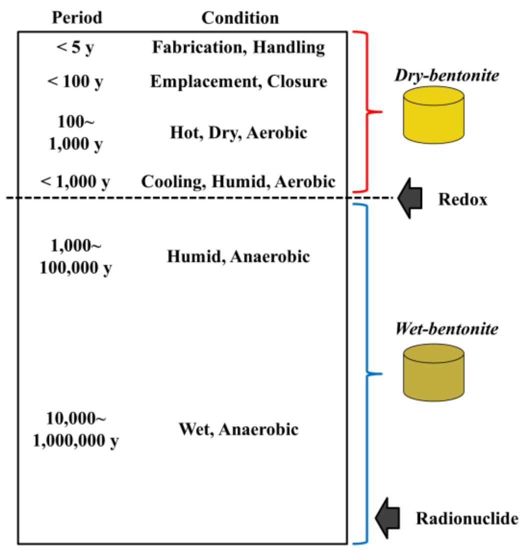


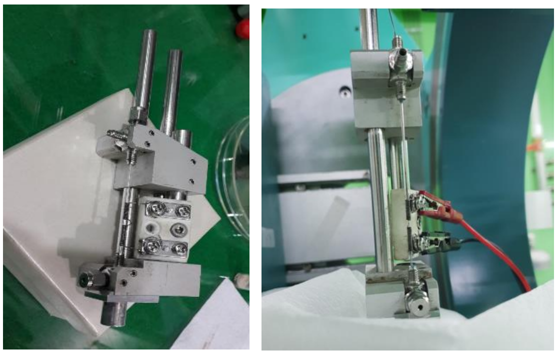
Fig. 1. Illustration of the conditions for the Korean buffer system with time (modified from Peter Keech's work) [2].

2. In-situ Synchrotron X-ray Powder Diffraction

2.1 Method

In-situ synchrotron X-ray powder diffraction on montmorillonite was performed at beamline 3D at Pohang light source (PLS-II) at Pohang accelerator laboratory (PAL). At beamline 3D, approximate 100 ㎛ beam of monochromatized X-rays with a wavelength of 0.6888 Å and MAR345 image plate detector was used to collect powder diffraction data.

For the in-situ sample heating environments, a sealed capillary with a diameter of 700 ㎛ surrounded by nickel-chrome resistance heating coils was heated from RT to 250℃ as a function of temperature (Fig. 2). Temperature was raised for 30 minutes to each targeted steps, and was stabilized for 30 minutes.



(a) (b)

Fig. 2. Setup for in-situ synchrotron XRD experiments under the elevated temperatures for the Korean bentonite (a) front view and (b) side view with heating linseed.

2.2 Montmorillonite under Dry Condition

Under dry condition, the Gyeongju bentonite shows thermal expansion after first dehydration sequence with 35.4% volume contraction at 120°C. Such hydration and/or dehydration reactions of montmorillonite are of significance in geological processes and also the waste disposal [3,4]. At cooling sequence, (001) reflection remains with 10 Å d-spacing without any hydration expiation. After air contact for a week, however, (001) d-spacing showed complete recovery (15.12 Å) to initial state (Fig. 3).

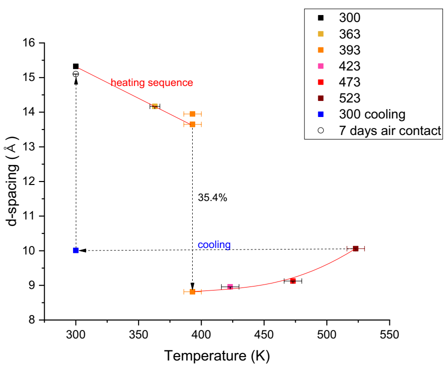


Fig. 3. Changes in the d-spacing (Å) of montmorillonite (001) reflections under dry condition as a function of temperatures.

3. Conclusions

We have experimentally established the characterization methodology for the Korean buffer under the elevated temperature employing a setup especially for the in-situ synchrotron XRD measurements. The montmorillonite (001) d-spacing of the Korean bentonite changes under the elevated temperature conditions show particular contraction at 120℃ with dehydration sequence. Gradual volume expansion of montmorillonite up to 250℃ implies its thermal expansion is anticipated after the dehydration of the buffer.

ACKNOWLEDGEMENT

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