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Swelling characteristics of bentonite after long-term dissolution in alkaline solution

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Abstract

In a high-level radioactive waste repository, bentonite may react with the alkaline solution produced by cement degradation. In this study, bentonite was mixed with alkaline solution in a closed system and reacted for 3–24 months. Furthermore, swelling tests were conducted on the alkaline-dissolved bentonite immersed in distilled water. The swelling deformation decreased significantly with increases in the concentration of NaOH solution and reaction time, and this was mainly due to montmorillonite dissolution. The fractal $e-p$ relationship (e is the void ratio and p is the vertical pressure) with two calculation coefficients (the swelling coefficient and the fractal dimension) was employed to determine the swelling of alkaline-dissolved bentonite. The fractal dimension increased slightly with increasing reaction time or concentration of NaOH solution, as the dissolution traces caused by the alkaline solution favoured an increase in the irregularity and fractality of the bentonite surface. The swelling coefficient decreased linearly with decreasing montmorillonite content. In addition, the swelling coefficient and the fractal dimension were related exponentially to the reaction time in alkaline solution. A relationship between the swelling of alkaline-dissolved samples and the reaction time was proposed, which might be used to assess the swelling properties of bentonite barriers that would be affected by long-term dissolution of the alkaline solution in a closed repository.

Keywords: alkaline solution, bentonite, fractal dimension, long-term reaction, swelling deformation

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The safe disposal of high-level radioactive waste (HLRW) presents a significant challenge for many countries. Deep geological disposal, in many approaches, includes HLRW in sealed canisters, buffer/backfill materials and host rocks, and this is internationally recognized as an effective method for the disposal of HLRW (Komine, 2008; Chen *et al.*, 2011; Hadi *et al.*, 2019). Bentonite is generally recognized as an ideal buffer/backfill material between the canister containing HLRW and the host rock in deep geological repositories due to its low hydraulic conductivity, high swelling capacity and good self-sealing capacity (Dixon & Gray, 1985; Komine & Ogata, 2004; Ye *et al.*, 2010; Xiang *et al.*, 2019a). The high swelling capacity of bentonite may form an enclosed environment that restricts effectively the migration of nuclides released from the repository (Zhu *et al.*, 2013; Ye *et al.*, 2017; Liu *et al.*, 2018). High volumes of concrete are used in the construction of a repository, such as in the Swedish KBS3 repository, where the amount of cement is planned to be ~900,000 kg (Karnland *et al.*, 2007). However, concrete components decay during the long-term operation of a repository, producing a highly alkaline solution (Nakayama *et al.*, 2004; Fernández *et al.*, 2010; Chen *et al.*, 2016; Liu *et al.*, 2018). The alkaline solution may react with bentonite near the concrete in the enclosed environment for a long time, leading to significant

local degradation in the mechanical properties of the buffer. Therefore, prediction of the swelling properties of bentonite affected by the long-term dissolution of alkaline solution is of great importance in the performance assessment of the repository.

The volumetric swelling of bentonites in aqueous solution was shown to be due primarily to water adsorption by the montmorillonite interlayer cations (Madsen & Müller-VonMoos, 1989). The montmorillonite content in bentonite is an important factor influencing its swelling properties. However, previous work has indicated that alkaline solutions may dissolve montmorillonite, reacting to form secondary by-products such as zeolites and Mg-trioctahedral sheet silicates and reducing the swelling of bentonite (Lehikoinen *et al.*, 1996; Karnland *et al.*, 2007; Yamaguchi *et al.*, 2007; Cuisinier *et al.*, 2008). In addition, the greater the OH⁻ concentration, or the pH value, the more significant the dissolution of montmorillonite (Chen *et al.*, 2016). Furthermore, the cement pore solution enhances the dissolution of montmorillonite and reduces the swelling potential of bentonite (Savage *et al.*, 2007). The dissolution of montmorillonite is accompanied by the precipitation of zeolite, formed by the reaction between bentonite and an alkaline solution with a high pH (10.0–13.5) (Ramírez *et al.*, 2002). Herbert *et al.* (2008) studied the variation in the swelling properties of MX-80 bentonite within varying solutions, and they reported that the variation of montmorillonite or kaolinite in bentonite within varying solutions was the main reason for the variation in the swelling properties.

The duration of the reaction between the alkaline solution and bentonite is also an important influencing factor in

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montmorillonite dissolution. X-ray diffraction (XRD) is used widely to analyse the mineral composition of bentonite dissolved by alkaline solutions. Montmorillonite is dissolved continuously through long-term dissolution in alkaline solutions, resulting in a decrease in the swelling properties and an increase in the porosity of bentonite, thereby reducing the buffering and self-closing properties of bentonite (Lehikoinen *et al.*, 1996; Nakayama *et al.*, 2004; Sánchez *et al.*, 2006; Yamaguchi *et al.*, 2007).

The swelling of bentonite within alkaline solutions was also investigated through microstructure studies. Several studies reported the dissolution of montmorillonite and an increase in the total porosity in a synthetic alkaline groundwater environment (Karnland *et al.*, 2007; Cuisinier *et al.*, 2008; Chen *et al.*, 2016; Liu *et al.*, 2018). Karnland *et al.* (2007) reported that the microstructures of bentonite samples before and after contact with 1.0 mol L⁻¹ NaOH solution for several months were significantly different. After the reaction of MX-80 bentonite with solutions of various ionic concentrations and pH values for 3 years, significant alterations to the montmorillonite microstructure were identified by transmission electron microscopy (TEM). After reaction in various solutions, all smectite particles were transformed from an initial film-like matrix of very thin, platy shape to a matrix of discrete, well-oriented platy shape. Chen *et al.* (2016) carried out permeability and dissolution experiments on Gaomiaozhi bentonite within NaOH solutions, and they observed cracks in the montmorillonite aggregates using scanning electron microscopy. The formation of small montmorillonite aggregates was also observed, suggesting that the aggregates were broken down into small ensembles due to dissolution via alkaline solution.

At present, a scientific estimation method for the swelling characteristics of bentonite after long-term reaction with alkaline solution has not been determined. Based on the fractal microstructures of bentonite, Xu *et al.* (2014) observed that there is a fractal relationship between the void ratio (e) of bentonite at saturation and the vertical pressure (p) in distilled water, represented as:

$$e = \kappa p^{D_s-3} \quad (1)$$

where κ is the swelling coefficient and D_s is the surface fractal dimension of bentonite. The equation offers a simple method for predicting the swelling characteristics of the clay, but it does not take into consideration the reaction between the alkaline solution and bentonite. This study investigates the variation in montmorillonite content and the microstructure of bentonite affected by long-

term dissolution in alkaline solution in a closed environment, and it employs the calculation coefficients κ and D_s in the fractal e - p relationship to characterize the effects of montmorillonite content and microstructure on the swelling of bentonite, respectively. Then, a method for predicting the swelling characteristics of bentonite after long-term reaction with alkaline solution is proposed based on the relationship between the calculation coefficients and the reaction time.

Materials and methods

Materials

Natural sodium bentonite from Henan Province, China, was used for the reaction with alkaline solution. The bentonite has a liquid limit of 303.5% and a plastic limit of 28.6%. X-ray diffraction was employed for semi-quantitative evaluation of the mineral content using *X'Pert HighScore Plus* software based on the relative intensity of Bragg reflections, including weight-based reference intensity ratio correction (Fig. 1). *Powder Diffraction File-4 (PDF-4)* was used for phase identification (Faber & Fawcett, 2002). The XRD tests and mineral composition analyses were carried out by specialized personnel from the Instrumental Analysis Center of Shanghai Jiao Tong University. The original bentonite contained 77.3% montmorillonite, 16.3% quartz and 6.1% feldspar. The physicochemical properties of the bentonite tested were similar to those of GMZ01 bentonite (Ye *et al.*, 2017), which has been selected as the preferred buffer/backfill material for the planned Chinese repository for HLRW. Therefore, the bentonite tested can be considered as the backup material for the GMZ01 bentonite.

Reaction of alkaline solution with bentonite

Because the cement leachates have complex compositions (Berner, 1992), the reactions between bentonite and cement leachates are also complex (Savage *et al.*, 1992). As the tested bentonite was dominated by Na, only the effects of OH⁻ on montmorillonite content and swelling deformation will be considered. Therefore, the NaOH solution was employed to simulate hyper-alkaline porewater.

The bentonite was dried at 105°C for 24 h, and 500 g of dried bentonite was mixed with 500 mL of 0.5 and 1.0 mol L⁻¹ NaOH solution. The wet bentonite was sealed in various high-density polyethylene Nalgene bottles and stored at room temperature

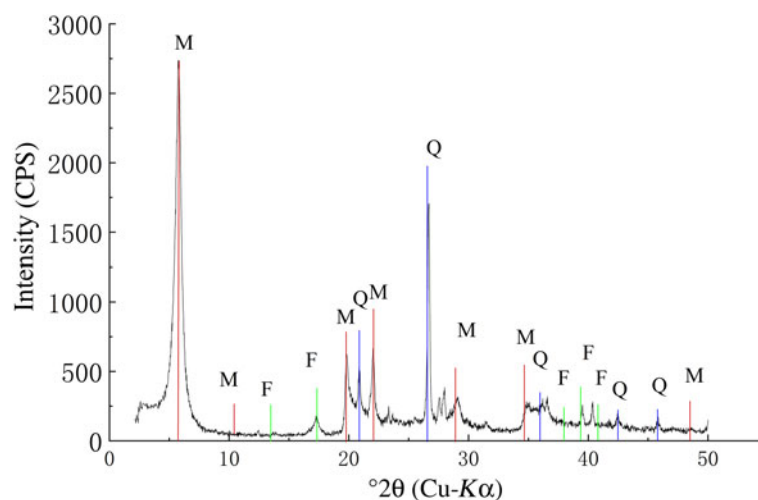


Fig. 1. XRD traces of the original bentonite. CPS = counts per second; Q = quartz; M = montmorillonite; F = feldspar.

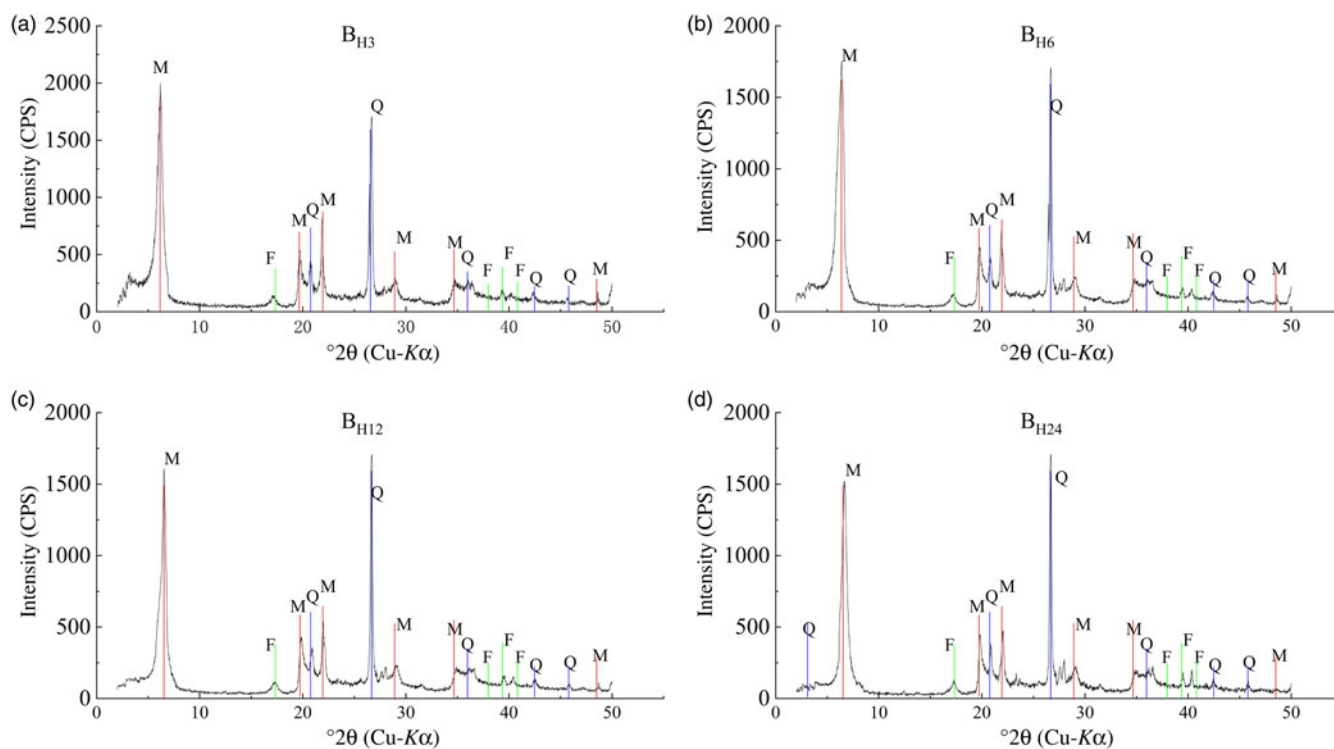


Fig. 2. XRD traces of the bentonite samples dissolved in 1.0 mol L^{-1} NaOH solution. CPS = counts per second; Q = quartz; M = montmorillonite; F = feldspar.

for a scheduled period of time (3, 6, 12 and 24 months). After these time intervals had elapsed, the alkaline-dissolved bentonite was removed. The samples were washed carefully with distilled water five times to remove the residual alkaline solution, dried at 105°C for 24 h and ground to a powder.

Characterization of alkaline-dissolved bentonite

The mineral compositions of the original and alkaline-dissolved bentonite samples were analysed using a D8 ADVANCE X-ray powder diffractometer (Bruker, Germany). The fractal dimension of bentonite was obtained by N_2 adsorption tests using an ASAP 2010 M + C surface area and porosimetry analyser (Micromeritics, USA). The surface morphology of the smectite particles was observed by TEM (H-9000NAR, Hitachi, Japan). The XRD analysis was performed with Cu- $K\alpha$ radiation ($K = 0.15$, 418 nm). The measurement range was from 2 to $50^\circ 2\theta$, with a scanning rate $8^\circ 2\theta \text{ min}^{-1}$.

Swelling deformation tests

The original and pre-treated bentonite powder was mixed with water to reach a water content of $\sim 18\%$. The wet soil was statically compacted into cylindrical samples of 60 mm in diameter and 10 mm in height. The initial dry density of the samples was controlled at $1.70 \pm 0.02 \text{ g cm}^{-2}$.

The compacted samples were placed into a sample ring and covered with two porous stones on the upper and lower surfaces. After vertical pressure was applied to the samples, distilled water was circulated through the porous plate at the bottom of the samples. The swelling deformation of the samples was recorded using a strain gauge with a precision of 0.001 mm. The samples were

Table 1. Mass percentages of the mineral compositions of the bentonite samples.

Sample	Mass percentage		
	Montmorillonite	Quartz	Feldspar
B ₀	77.3	16.3	6.1
B _{L3}	73.6	17.7	8.5
B _{L6}	70.5	19.2	10.1
B _{L12}	68.1	19.8	11.9
B _{L24}	66.8	20.1	12.9
B _{H3}	71.6	18.7	9.4
B _{H6}	68.4	19.5	11.7
B _{H12}	66.1	20.4	13.2
B _{H24}	64.2	20.9	14.5

considered to be stable and fully saturated when the reading on the dial gauge remained constant for 24 h.

The original bentonite powder was labelled as B₀, and the series of bentonite samples that were dissolved in the lower concentration of the NaOH solution for 3, 6, 12 and 24 months were labelled as B_{L3}, B_{L6}, B_{L12} and B_{L24}, respectively. Similarly, the series of bentonite samples dissolved in 1.0 mol L^{-1} (*i.e.* a more concentrated NaOH solution) were labelled as B_{H3}, B_{H6}, B_{H12} and B_{H24}, respectively.

Results and discussion

Mineralogical composition

In the XRD traces (Fig. 2), the diffraction maximum in the diffraction angle of $5\text{--}7^\circ$ corresponds to montmorillonite, and its intensity decreased with increasing reaction time, suggesting that the montmorillonite dissolved gradually.

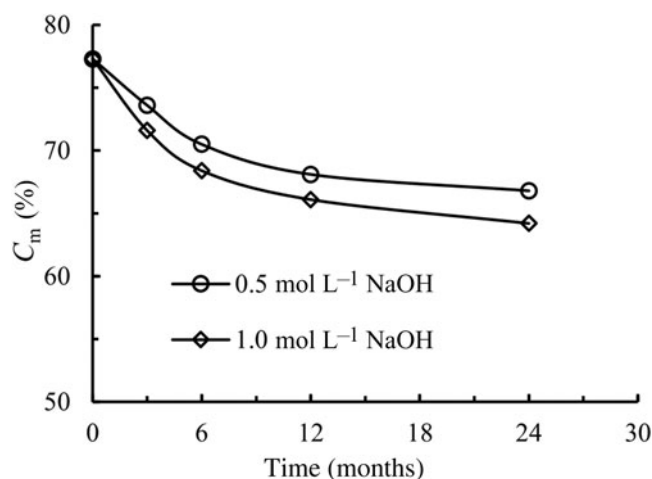


Fig. 3. Montmorillonite content (C_m) vs reaction time for samples dissolved in alkaline solution. CPS = counts per second.

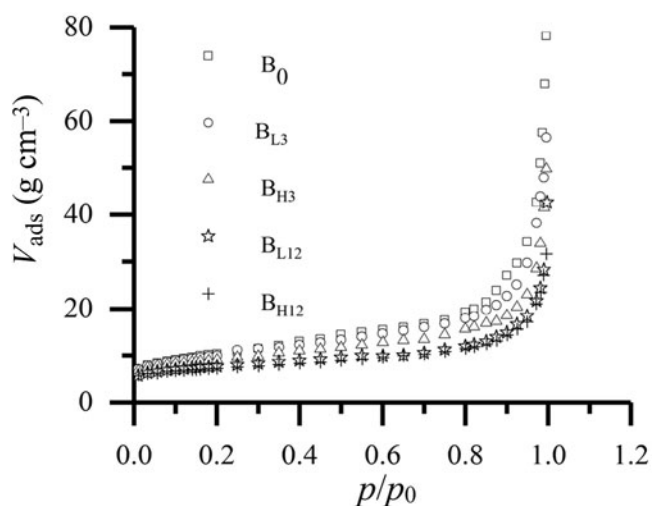


Fig. 4. N_2 adsorption isotherms of the bentonite samples.

The mineralogical compositions of alkaline-dissolved samples were obtained using *X'Pert HighScore* software by taking the average of three samples in an identical solution over the same reaction time (Table 1). The montmorillonite content exceeded 60% in all of the specimens, but it decreased with increasing dissolution time (Fig. 3). The montmorillonite content of the samples dissolved in 0.5 mol L^{-1} NaOH solution was greater than that of samples dissolved in 1.0 mol L^{-1} NaOH solution over the same reaction time. The montmorillonite content decreased quickly over short reaction times due to dissolution by the two NaOH solutions. The rate of decrease decelerated gradually because the OH^- concentration in the bentonite samples decreased gradually under the tested conditions.

Fractal dimension

The N_2 adsorption isotherms of the bentonite samples (Fig. 4) show that the original bentonite (B_0) has the greatest adsorption amount (V_{ads}) at full saturation, and the adsorption amount decreased gradually with increasing reaction time and

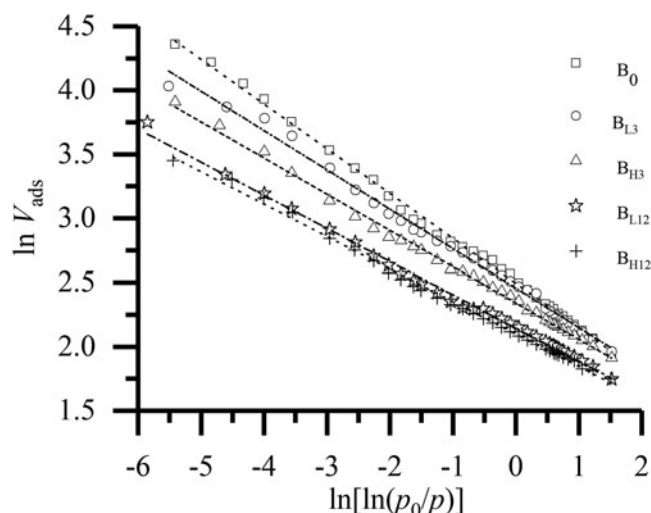


Fig. 5. Fractal dimensions of the bentonite samples according to N_2 adsorption.

Table 2. Surface fractal dimensions of the bentonite samples.

Sample	B_0	B_{L3}	B_{L6}	B_{L12}	B_{L24}	B_{H3}	B_{H6}	B_{H12}	B_{H24}
D_s	2.666	2.673	2.684	2.698	2.706	2.681	2.694	2.707	2.721

concentration of NaOH solution, mainly due to the decrease in montmorillonite content. The fractal dimension of the bentonite samples was calculated using the Frenkel–Halsey–Hill equation (Yin, 1991):

$$V_{\text{ads}} \approx \left[RT \ln \left(\frac{p_0}{p} \right) \right]^{-(3-D_s)} \quad (2)$$

where V_{ads} is the gas volume adsorbed at equilibrium pressure p , p_0 is the saturation pressure of the adsorbate, R is the molar gas constant and T is the temperature (in Kelvin). Then, the D_s value was estimated using Eq. (2) (Fig. 5), the results of which are listed in Table 2. The D_s values increase with increasing reaction time and concentration of NaOH solution. This could imply that greater dissolution by the alkaline solution led to a larger surface fractal dimension of bentonite. D_s was introduced for quantification of the complexity of the surface topographies based on the self-similarity of surfaces at various scales (Mandelbrot, 1990; Xu, 2003; Kolay & Kayabali, 2006; Xiang *et al.*, 2019b). The range for D_s is 2–3, where greater surface roughness indicates a larger D_s value and a smooth surface has a value of $D_s = 2$ (Xu, 2003; Kolay *et al.*, 2006; Xiang *et al.*, 2017). Transmission electron microscopy images at 20 nm resolution were used to observe the montmorillonite particles in the samples dissolved in 1.0 mol L^{-1} NaOH solution (Fig. 6). The TEM images indicate that the more significant the dissolution via alkaline solution, the more obvious the dissolution traces on the surface of montmorillonite. The dissolution traces increased the irregularity and the fractality of the bentonite surface.

Swelling deformation

The maximum swelling strain was influenced by the vertical pressure, the reaction time and the concentration of alkaline solution (Fig. 7). The maximum swelling strain always

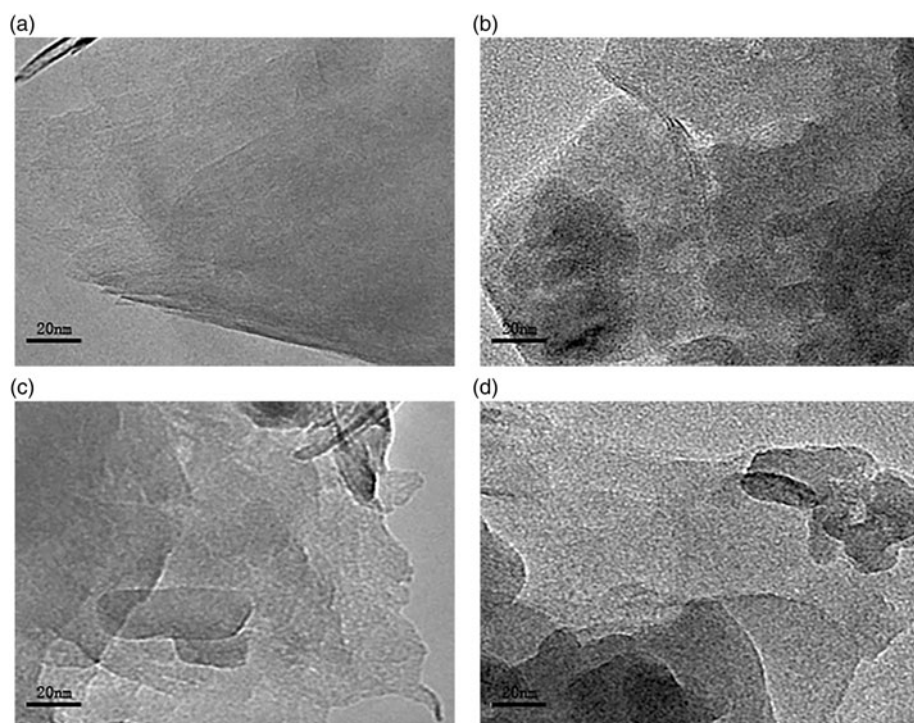


Fig. 6. TEM images of (a) the original bentonite and the samples reacted with 1.0 mol L^{-1} NaOH solution at (b) 3 months, (c) 12 months and (d) 24 months.

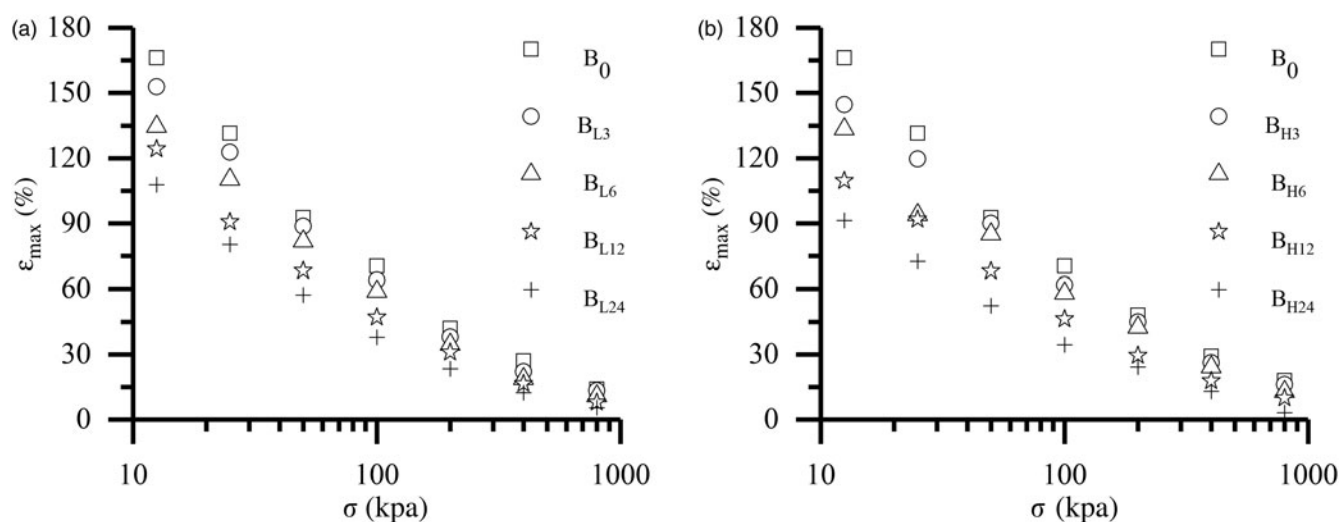


Fig. 7. Maximum swelling strain vs vertical pressure on bentonite samples dissolved in (a) 0.5 mol L^{-1} NaOH solution and (b) 1.0 mol L^{-1} NaOH solution.

decreased with increasing vertical overburden pressure. When all other conditions were held constant, the original bentonite samples showed the greatest swelling strains compared to the samples dissolved in the alkaline solution. With increasing reaction time, the maximum swelling strain of the alkaline-dissolved samples decreased gradually. The maximum swelling strain, ϵ_{max} , values of the samples dissolved in 0.5 mol L^{-1} NaOH solution were greater than those of samples dissolved in 1.0 mol L^{-1} NaOH solution under the same reaction conditions.

Correlation of the void ratio to vertical pressure

The void ratio of the bentonite samples decreased with increasing vertical pressure, reaction time and concentration of the NaOH

solution (Fig. 8). In addition, for samples dissolved in the same NaOH solution for the same reaction time, the plots of the void ratio vs vertical pressure may be represented by a uniform fractal $e-p$ relationship using the measured fractal dimension. The $e-p$ relationships for the original bentonite and the alkaline-dissolved samples are listed in Table 3. The swelling coefficient, κ , decreased with increasing concentration of the NaOH solution. The κ values of samples dissolved in 0.5 mol L^{-1} NaOH solution were greater than those of samples dissolved in 1.0 mol L^{-1} NaOH solution. Similarly, the κ values decreased gradually with increasing reaction time, where the κ values were minimal for samples dissolved in the alkaline solution over 24 months. The trend regarding the swelling deformation, which was mainly caused by the decrease in montmorillonite content. According to the relationship between the

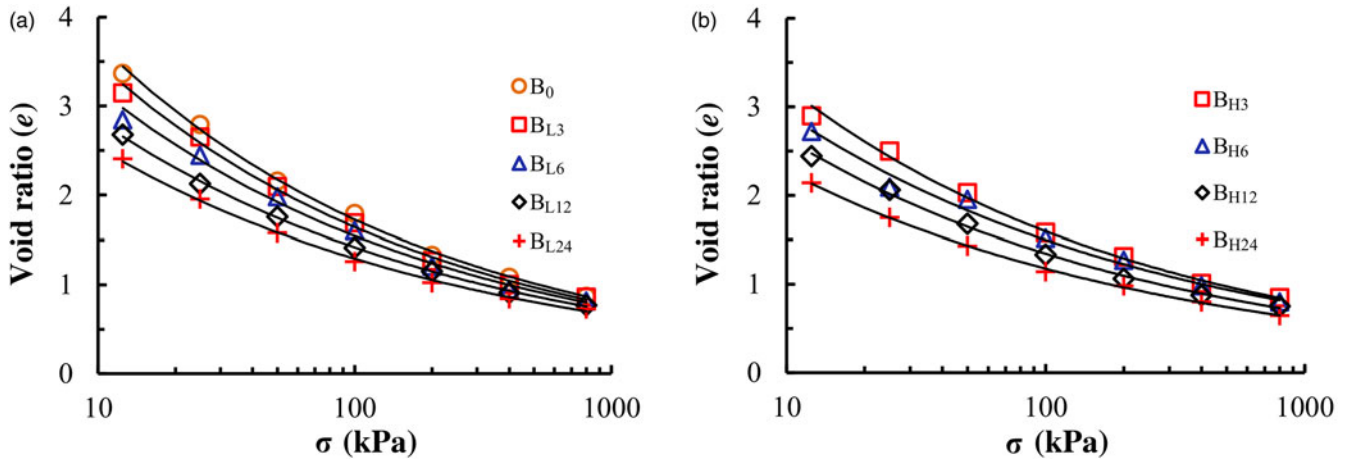


Fig. 8. Void ratio at saturation vs vertical pressure on bentonite samples dissolved in (a) 0.5 mol L⁻¹ NaOH solution and (b) 1.0 mol L⁻¹ NaOH solution.

Table 3. *e*-*p* relationship for the original and alkaline-dissolved bentonite samples.

<i>c</i> (mol L ⁻¹)	<i>t</i> (months)	<i>e</i> - <i>p</i> relationship	κ	R ²
Original bentonite		$e = 7.954p^{-0.334}$	7.954	0.994
0.5	3	$e = 7.413p^{-0.327}$	7.413	0.993
	6	$e = 6.610p^{-0.316}$	6.610	0.995
	12	$e = 5.705p^{-0.302}$	5.705	0.994
	24	$e = 4.987p^{-0.294}$	4.987	0.992
1.0	3	$e = 6.845p^{-0.319}$	6.845	0.989
	6	$e = 5.996p^{-0.306}$	5.996	0.994
	12	$e = 5.181p^{-0.296}$	5.181	0.991
	24	$e = 4.215p^{-0.279}$	4.215	0.994

swelling coefficient, κ , and the montmorillonite content, c_m , for samples dissolved in both NaOH solutions at various reaction times (Fig. 9), the κ values decreased linearly with decreasing montmorillonite content.

Estimation of swelling deformation

The swelling coefficient, κ , and the surface fractal dimension, D_s , can be represented as exponential functions of reaction time for samples dissolved in the alkaline solution in a closed environment:

$$0.5 \text{ mol L}^{-1}: \kappa = \kappa_0 e^{-0.248t} \quad \text{and} \quad D_s = D_0 e^{0.009t} \quad (3)$$

$$1.0 \text{ mol L}^{-1}: \kappa = \kappa_0 e^{-0.395t} \quad \text{and} \quad D_s = D_0 e^{0.013t} \quad (4)$$

where κ_0 and D_0 are the swelling coefficient and surface fractal dimension of the original bentonite, respectively, and t is the reaction time in years. Thus, the relationships between the void ratio at saturation and the reaction time for samples dissolved in 0.5 and 1.0 mol L⁻¹ NaOH solutions were expressed respectively as:

$$0.5 \text{ mol L}^{-1}: e = \kappa_0 e^{-0.248t} p^{D_0 e^{0.009t} - 3} \quad (5)$$

$$1.0 \text{ mol L}^{-1}: e = \kappa_0 e^{-0.395t} p^{D_0 e^{0.013t} - 3} \quad (6)$$

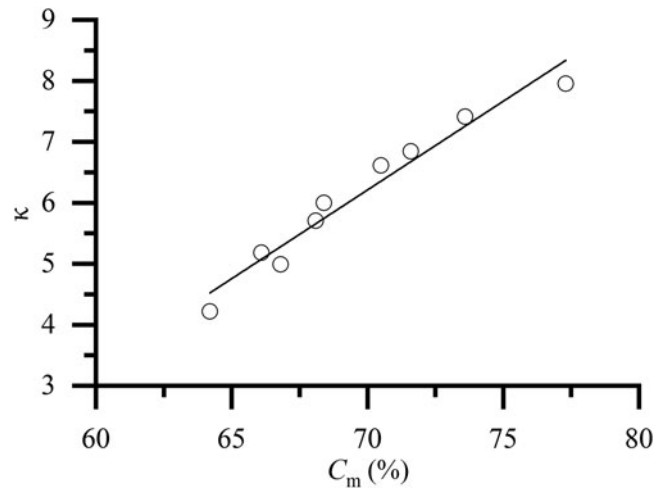


Fig. 9. Swelling coefficient vs montmorillonite content for alkaline-dissolved bentonite samples.

According to the relationship between the void ratio and the reaction time (Fig. 10), the void ratio of bentonite decreased with increasing reaction time. The curves were estimated using Eqs (5) and (6) for the samples dissolved in 0.5 and 1.0 mol L⁻¹ NaOH solutions, respectively. There was good agreement between the predicted void ratio and the experimental data.

The relationship between the maximum swelling strain, ϵ_{max} , and the void ratio, e , at full saturation can be written as (Xu *et al.*, 2003):

$$\epsilon_{max} = \left(\frac{G_b/G_m e + 1}{G_b} \rho_d - 1 \right) \times 100\% \quad (7)$$

where G_b is the specific gravity of bentonite, G_m is the specific gravity of montmorillonite and ρ_d is the initial dry density.

Thus, the relationship between the maximum swelling strain and reaction time can be obtained by combining Eqs (5) or (6) with Eq. (7). The experimental data of samples dissolved in 0.5 and 1.0 mol L⁻¹ NaOH solution were plotted for comparison (Fig. 11), and the predicted values of ϵ_{max} agreed well with the

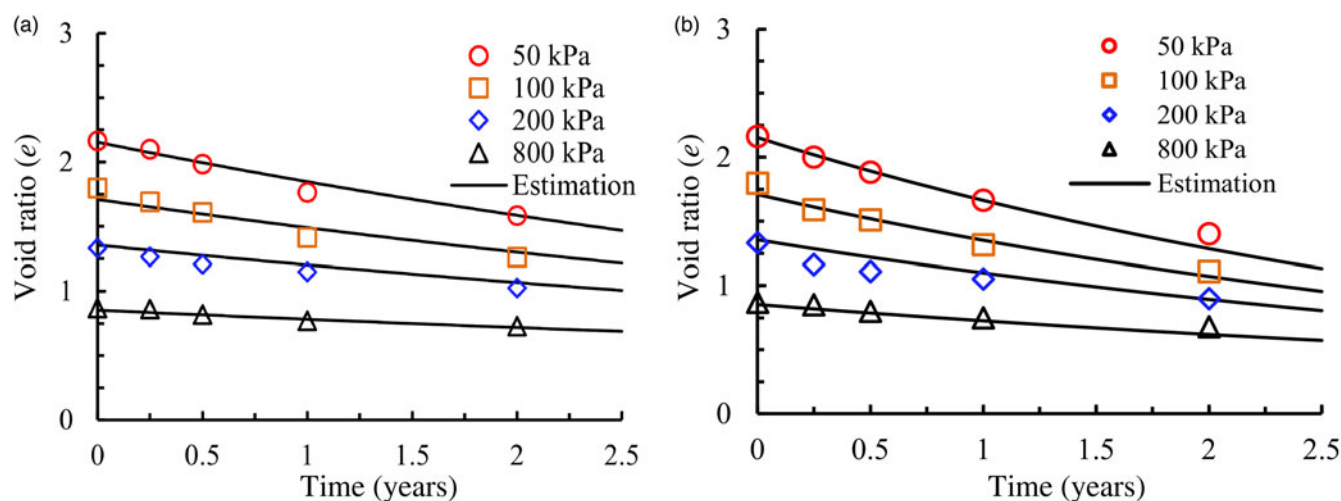


Fig. 10. Void ratio vs reaction time for bentonite dissolved in (a) 0.5 mol L^{-1} NaOH solution and (b) 1.0 mol L^{-1} NaOH solution.

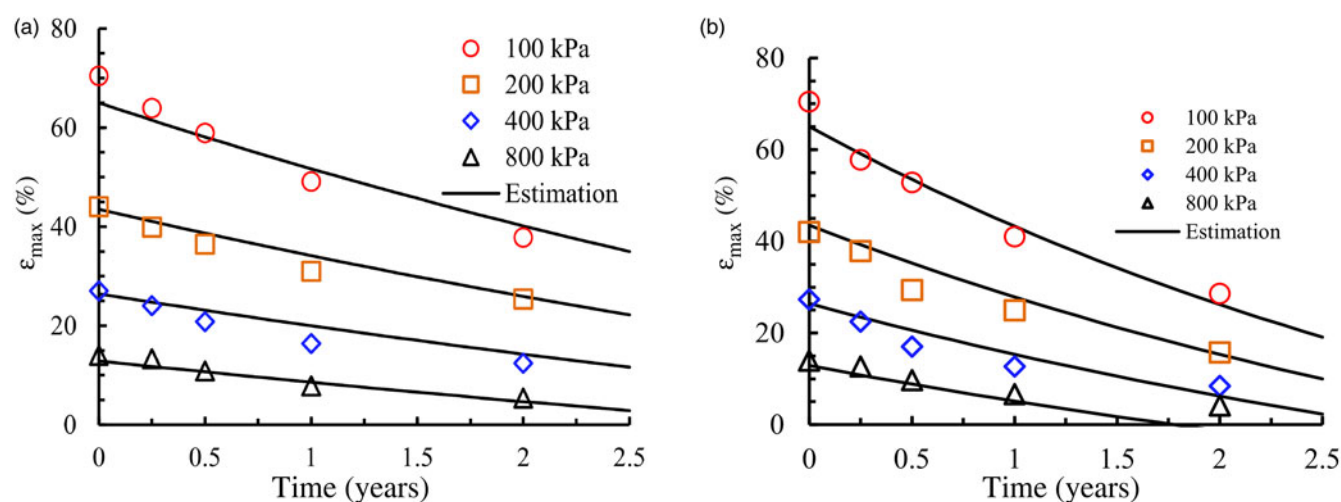


Fig. 11. Maximum swelling strain vs reaction time for samples dissolved in (a) 0.5 mol L^{-1} NaOH solution and (b) 1.0 mol L^{-1} NaOH solution.

experimental results. The e - p relationship may offer a new method for predicting the swelling deformation of bentonite after long-term reaction with alkaline solution in a closed repository, incorporating reaction duration.

Summary and conclusions

The alkaline solution affected significantly the montmorillonite mineral, as the montmorillonite content decreased with increasing concentration of the NaOH solution and reaction time in a closed environment. However, the dissolution rate of montmorillonite content slowed due to the decrease in the OH^- concentration in the bentonite samples.

The swelling deformation of bentonite also decreased with increasing NaOH concentration and reaction time. The void ratio at saturation of samples dissolved in the same solution for the same reaction time may be expressed by the fractal swelling model $e = \kappa p_s^{D_s - 3}$. The fractal dimension, D_s , slightly increased with increasing reaction time and concentration of NaOH solution due to the appearance of dissolution traces on the surface,

which increased the irregularity of the bentonite surface. The more significant the alkaline dissolution, the greater the surface fractal dimension of bentonite. However, the swelling coefficient, κ , decreased with increasing concentration of NaOH and reaction time. The swelling coefficient was linearly correlated with the montmorillonite content.

The swelling coefficient and the fractal dimension were expressed as exponential functions of the reaction time. The relationship between the swelling deformation of the samples and reaction time was established based on the fractal swelling model. Using this relationship, the void ratios of the samples dissolved in various NaOH solutions at various reaction times were estimated. Good agreement existed between the estimations and the measured values. The void ratio decreased with increasing reaction time, and the increase in the OH^- concentration of the NaOH solution promoted the reduction process. The variation of maximum swelling strain with reaction time was also estimated. The predicted value was consistent with the tested data, suggesting that the swelling strain decreased significantly with increasing reaction time.

This study proposed a relatively short-term reaction, and the longer-term effects of mineral growth on the swelling properties of bentonite were ignored. A long-term reaction will inevitably include mineral growth as well as mineral dissolution, as has been highlighted previously (Fernández *et al.*, 2006; Savage *et al.*, 2007; Yamaguchi *et al.*, 2007). Therefore, the longer-term effects of mineral growth and dissolution on the swelling properties of bentonite require further study.

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