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A new method to predict swelling pressure of compacted bentonites based on diffuse double layer theory

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Abstract. Compacted bentonites were chosen as the backfill material and buffer in high level nuclear waste disposal due to its high swelling pressure, high ion adsorption capacity and low permeability. It is essential to estimate the swelling pressure in design and considering the safety of the nuclear repositories. The swelling pressure model of expansive clay colloids was developed based on Gouy-Chapman diffuse double layer theory. However, the diffuse double layer model is effective in predicting low compaction dry density (low swelling pressure) for certain bentonites, and invalidation in simulating high compaction dry density (high swelling pressure). In this paper, the new relationship between nondimensional midplane potential function, u, and nondimensional distance function, Kd, were established based on the Gouy-Chapman theory by considering the variation of void ratio. The new developed model was constructed based on the published literature data of compacted Na-bentonite (MX80) and Ca-bentonite (FoCa) for sodium and calcium bentonite respectively. The proposed models were applied to re-compute swelling pressure of other compacted Na-bentonites (Kunigel-V1, Voclay, Neokunibond and GMZ) and Ca-bentonites (FEBEX, Bavaria bentonite, Bentonite S-2, Montigel bentonite) based on the reported experimental data. Results show that the predicted swelling pressure has a good agreement with the experimental swelling pressure in all cases.

Keywords: bentonite, swelling pressure, expansive soils, nuclear waste disposal

1. Introduction

Deep geological repository is popularly being considered as storage for high level nuclear waste in many countries such as Sweden, France, Japan, China, Czech Republic, Germany, Belgium, Spain and so on (Dixon and Grey 1985, Pusch 1992, Delage 2007, Sun *et al.* 2018). The high level nuclear wastes are mainly from the 'burning' of uranium fuel in a nuclear reactor. The used uncycled nuclear fuels are firstly storage in ponds for 40 or 50 years. After that the used fuel assemblies are ready for encapsulation or loading into casks ready for indefinite storage or permanent disposal underground (WNA 2016). Spent nuclear fuel will be stored in isolated copper canisters inside underground bedrock with a depth of several hundred meters. The canisters will be placed in the tunnel galleries surrounded by multiple release barriers. Generally, the compacted bentonite blocks or other forms of bentonite like bricks and pellets are often planned as buffer and backfill material thanks to its low hydraulic conductivity, high-ion adsorption capacity and high swelling pressure (Bucher and Muller Vonmoos 1989, ENRESA 2000, Schanz and Tripathy 2009, Gueddouda 2011, Sun 2015). The function of compacted bentonite barrier is that the material should hold the canister tightly when subjected to water, simultaneously limit the access of water to waste container and prevent the migration of radionuclide into the nature, and should transfer the heat produced by nuclear radiation of nuclear fuels to the host rocks.

Once emplaced, the tunnel galleries are closed and the around environment are rebalanced. The unsaturated compacted bentonite will be hydrated by adsorbing the water or electrolytes from surrounding bedrocks. The space between canisters and bedrocks are always assumed to be constant full of compacted bentonite. The initially unsaturated compacted bentonite will exhibit swelling pressure when meet with water. To make sure the safety of the repository, the swelling pressure of the compacted bentonite must be no more than the in situ strength of the surrounding rocks. Thus, the estimation and determination of swelling pressure is essential and crucial for safety design and long terms functionality of backfill materials and buffer in nuclear repositories. Swelling pressure determined by laboratory tests have been studied by many researchers (Pusch 1982, Dixon and Gray 1985, ENRSEA 2000 and Komine 2004) who performed the tests at confined conditions without volume change of the compacted bentonite specimen.

Compacted bentonites are clays with high content of montmorillonite. Bentonite swelling is because of the clay-water-electrolyte interaction (Bolt 1956). It is known that the physicochemical forces are responsible for the swelling behavior of colloid bentonite substance from diffuse double layer theory (Bolt 1956, van Olphen, 1977, Mitchell 1993). According to Mašín (2015) summarized, the basic forms of diffuse double layer theory are only available for montmorillonite clay particles suspended in monovalent liquid solutions at low concentrations (Warkentin and

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Schofield 1958, Sridharan and Jayadeva 1982, Zhang *et al.* 1993, Yong 1999a, b, Komine and Ogata 1996, Mitchell and Soga 2005, Phillips and Tripathy 2011, Liu 2013). Some researchers attempted to study the swelling pressure of compacted bentonite based on diffuse double layer theory related to the physicochemical forces (Komine and Ogata 1996, Sridharan and Choudhury 2002, Tripathy *et al.* 2004, Bharat *et al.* 2013, 2017, Liu 2015, Sun 2017). The reason for difference between theoretical values from DDL theory and experimental data have been studied by many researchers (Barcey *et al.* 1972, Pusch 1982, Young and Mohamed 1992, Sridharan and Choudhury 2002, Tripathy *et al.* 2004 and Schanz and Tripahty 2009).

2. Literature review

Three mechanisms for clay mineral formation (inheritance, neoformation, and transformation) operating in three geological environments (weathering, sedimentary, and diagenetic-hydrothermal) yield nine possibilities for the origin of clay minerals in nature (Eberl 1984). The main group of clay minerals are kaolinite, illite and montmorillonite. Montmorillonite is the main content of bentonites. Generally, many types of exchangeable cations exist in bentonites material. The amount and type of cations of the bentonite depends on the mechanisms and geological environments of formation as mentioned before.

The unit layer of the montmorillonite consists of an aluminium octahedral sheet sandwiched between two silica tetrahedral sheets, as shown schematically in Fig. 1(a) (1-3). The thickness of unit layer is about 9.6 Ångström (Å) shown in Fig. 1(a) (3). Generally, cations and water molecules exist in the interlayer. The unit layers are bonded by weak van der Waal forces which occurred between molecules. The silica tetrahedral - aluminium octahedral - silica tetrahedral forms the structure of unit layer. The strong chemical bond is formed by atoms (Si-O, Al-O), which is much higher than van der Waal forces. The extensive isomorphous substitution occurred for silicon and aluminum which were replaced by other cations such as magnesium, iron, zinc, nickel, lithium etc. (Grim 1968). The charge deficiencies due to isomorphous substitution are balanced by exchangeable cations located within clay particles as shown in Fig. 1(b). The ideal arrangement of unit layers is parallel structure as shown in Fig. 1(c). In general, several unit layers comprise clay particles. The thickness of clay particles varies from 10 to 50 Å. The space between clay particles are called inter-particle pores where the diffuse double layer formed. At very low water content condition, the exchangeable cations are strongly attracted to the negative surface of unit layer. Two parallel unit layers are separated by cations which shows a very low level of hydration. This would be correspond to water in region A in Fig. 1(d). With ingress of water, the cations are partial hydrated by combining with the water molecules. The distance between unit layers will expand when the potential energy of repulsion is higher than the potential energy of attraction which comes from van der Waals forces and electrostatic forces between negative charged surface and positive charged interlayer cations. Probably, at this situation the water exist in region B as shown in Fig. 1(d). The balance of attraction and repulsion is denoted as crystalline swelling (Norrish 1954, Kittrick 1969). Crystalline swelling is a process whereby 0 to 4 discrete layers of water molecules are intercalated between unit layers shown in Fig. 1(b) (3). Layer hydrates with 0, 1, 2, 3, and 4 layers of water molecules are distinguished by basal spacings of approximately 10.0, 12.5, 15.0, 17.5 and 20.0 Å, respectively (Laird 2006). Upon further ingress of water, it will reach region C as shown in Fig. 1(d), the cations were fully hydrated and produced a higher concentration compared with the bulk fluid ion concentration far away from the clay surface. At the same time, the cations near surface try to diffuse to equilibrium the whole concentration due to the difference in ion concentration in bulk fluid solution and high ion concentration near clay surfaces. The swelling of the clay particles due to the osmotic phenomena are termed as osmotic swelling or diffuse double layer swelling (Van Olphen 1977). The negative charged clay surface, the distributed charged cation and the adsorbed water are together termed as the diffuse double layer (Bolt 1956, Van Olphen 1963). Usually, the thickness of diffuse double layer is around 200 Å which depends on the exchange cations, electronical surface charge, pore fluid, density, temperature etc.

The montmorillonite clay usually contains some monovalent or divalent cations during its formation under different geological environment. The different types of bentonite are each named after the respective dominant element, such as sodium bentonite, calcium bentonite etc. The types of exchange cations deeply influence the behavior of montmorillonite. Lambe and Whitman (1969) give the radius of un-hydrated and hydrated sodium ions as 0.98 Å and 7.8 Å, respectively. The thickness of DDL can expand to 120 Å for sodium bentonite under certain conditions (Grim 1968, Van Olphen 1977). If the monovalent sodium cations are replaced by divalent calcium or magnesium cations, there would need to be only half as many cations present to balance the negative charges on the clay particles, and the whole size of the clay particle would shrink (Nelson *et al.* 2015). Benson and Meer (2009) showed that clays having an abundance of monovalent cations had a much higher swell index than those with an abundance of divalent cations. Tamura *et al.* (2000) shown that the water uptake capacities of calcium or magnesium montmorillonites are nearly similar. The osmotic repulsive pressure is much less for divalent cations (Norrish 1954). However, the repulsive energy due to hydration of divalent exchangeable cations is significantly greater than that of monovalent ions (MacEwan 1954). These may contribute to the different swelling behavior of Na-bentonite and Ca-bentonite.

The basic form of DDL theory was proposed by Gouy-Chapman (Gouy 1910, Chapman 1913). This theory can predict the swelling behavior of bentonites by consideration of variations in clay-water-electrolyte systems. According to DDL theory, the interaction forces between double layers depends on the mindplane potential and ion concentration at mindplane between the two parallel clay platelets and its value equals to the osmotic pressure in that plane (Bolt 1956).

It is shown that the DDL theory can predict good value of swelling pressure with dry density lower than 1.55Mg/m^3

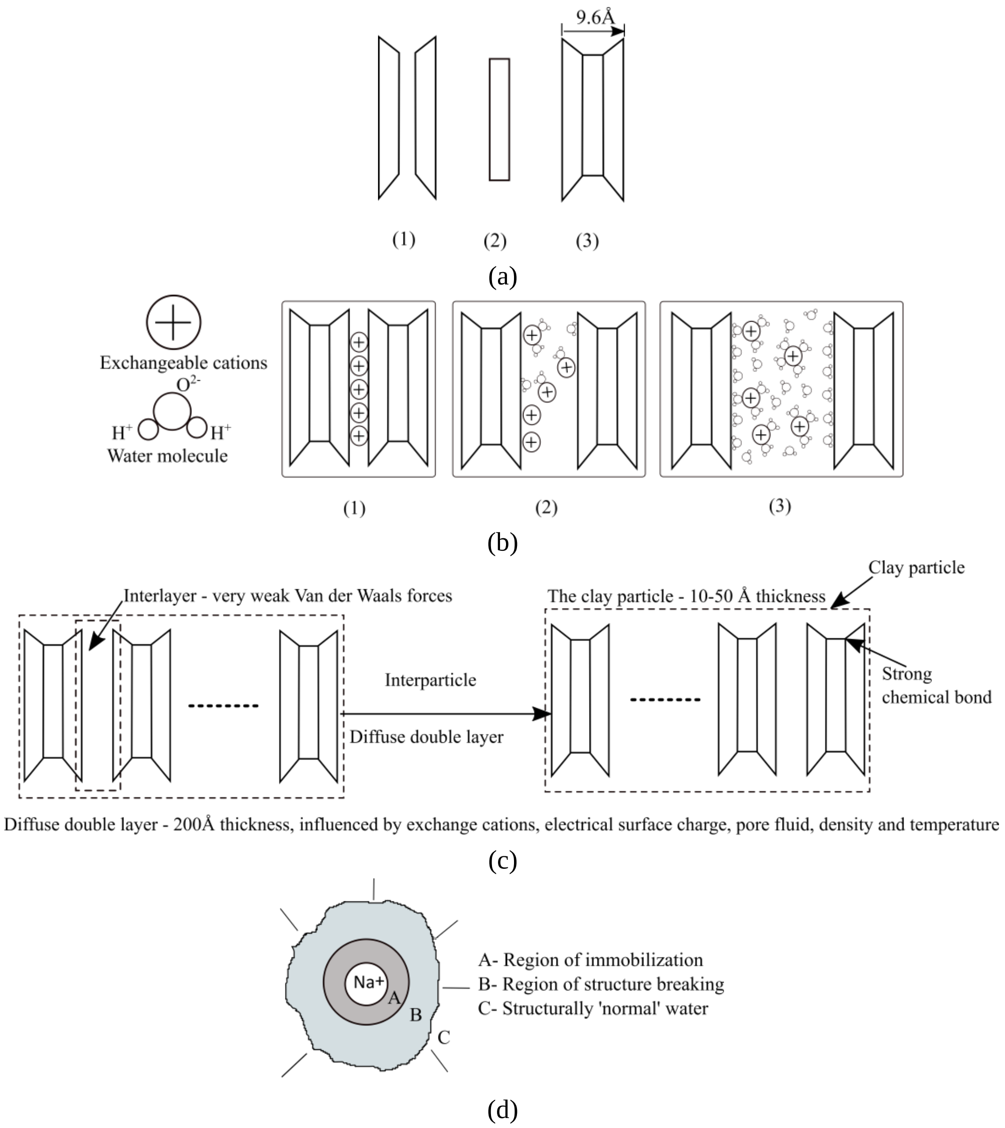


Fig. 1 Schematic diagram of structure of montmorillonite, (a) structure of unite clay layer, (1) Silica tetrahedral sheet, (2) Aluminium octahedral sheet, (3) unit clay layer; (b) Clay-water interactions, (1) dry condition, (2) water molecules wedge into the interlayer, (3) fully hydrated cations which results in repulsive forces and expanding caly layers (due to hydration energy); (c) interaction of clay particles, interpaticle, interlayer, diffuse double layer (thickness data from Lambe (1958) and Grim (1968)) and (d) Iron hydration (modified from Frank and Wen (1957))

for bentonite S-2 (Schanz and Tripathy 2009). Komine and Ogata (1996) pointed out that the DDL can predict adequately the swelling characteristics of compacted bentonite when the swelling pressure is less than about 3MPa and the type is sodium bentonite (Kunigel). However, there exists big difference between the theoretical value and experimental data at high dry density. The difference between theoretical values from DDL theory and experimental data have been studied by many researchers (Barcey *et al.* 1972, Pusch 1982, Young 1984, Young and Mohamed 1992, Sridaran and Choudhury 2002, Tripathy *et al.* 2004, Schanz and Tripahty 2009, Puppala *et al.* 2017). Some researchers (Verwey and Overbeek 1948, Van Olphen 1963 and Yong 1999) pointed out that the hydration energy due to surface and ion hydration are the major contributing factors to swelling pressure at close separation of clay platelets (high dry density). The difference between theoretical and experimental data was attributed to many factors (Bolt 1956, Sridharan and Jayadeva 1982, Mitchell 1993, Tripathy *et al.* 2004): (1) poorly developed or partially developed diffuse double layers, (2) surface and ion hydration near clay particles, (3) nonuniform size of clay particles, (4) deviation of clay fabric structure, (5) presence of non-swelling minerals in the clay.

From the literature review, it can be seen that the DDL was effective in predicting swelling pressure with low dry density for some certain bentonites (Komine and Ogata 1996) and has been successfully to explain the compressibility behavior of colloid clays (Mitchell 1993). Tripathy *et al.* (2004) proposed three new equations for prediction of swelling pressure by considering multiple valent montmorillonite, while it doesn’t work well on some divalent rich bentonites (Schanz and Tripathy 2009).

In this paper the Gouy-Chapman diffuse double layer theory are used to calculate the theoretical swelling pressure. The difference of swelling pressure between theoretical and experimental data are shown for all compacted Na-bentonites and Ca-bentonites from literature. Two new proposed equations are derived from Na-bentonites (MX80) and Ca-bentonites (FoCa), later the new equations are proposed to predict the swelling pressure of other compacted bentonites.

3. Theoretical relationship between swelling pressure and dry density

The swelling pressure is the difference between the osmotic pressure in the central plane between two clay platelets and the osmotic pressure in the equilibrium solution (Bolt 1956). The osmotic pressure at the central plane between clay platelets can be calculated from Gouy-Chapman diffuse double layer theory which original presented by Bolt (1956) and Van Olphen (1963) and improved by Sridharan and Jayadeva (1982). The following equations are used to establish the theoretical relationship between swelling pressure and dry density

$$\mathrm{e} = \mathrm{G}\gamma_w S d \times 10^6 \tag{1}$$

$$\gamma_d = \frac{G\gamma_w}{1+e} \tag{2}$$

$$\int_z^u \frac{1}{\sqrt{(2\cosh y - 2\cosh u)}} dy = \int_0^d d\xi = -Kd \tag{3}$$

$$\begin{aligned} -(\frac{dy}{d\xi})_{x=0} &= \sqrt{(2\cosh z - 2\cosh u)} \\ &= (\frac{B}{S})\sqrt{\frac{1}{2\varepsilon_0 D n_0 kT}} \qquad \text{at x=0, y=z} \end{aligned} \tag{4}$$

$$p = 2n_0 kT(\cosh u - 1) \tag{5}$$

$$K = (\frac{2n_0 e'^2 v^2}{\varepsilon_0 DkT})^{\frac{1}{2}} \tag{6}$$

where e is the void ratio of compacted bentonite, G is the specific gravity, γ_w is the density of water, γ_d is the dry density, S is the specific surface area (m^2/g), d is the half of the distance of clay platelets (m), u is the nondimensional midplane potential, y is the nondimensional potential at a distance x from the clay surface, z is the nondimensional potential function at the surface (x = 0), ξ is the distance function(ξ = Kx), B is the base cation exchange capacity (meq/100g), ε_0 is the permittivity of vacuum (8.8542×10^{-23} $C^2J^{-1}m^{-1}$), D is the dielectric constant of bulk fluid (80.4 for water), n_0 is the ionic concentration of the bulk fluid in ions/m^3, k is the Boltzmann's constant (1.38×10^{-23} J/K), K is the diffuse double layer parameter (1/m), T is the absolute temperature in Kelvin, p is the swelling pressure (Pa), e' is the elementary electric charge (1.602×10^{-23} C), v is the weighted averaged of valency of exchangeable cations (Tripathy *et al.* 2004).

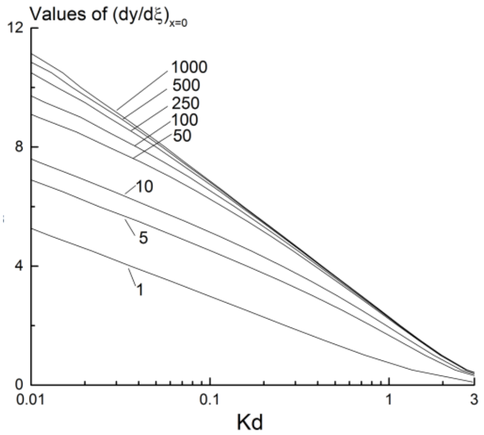


Fig. 2 Theoretical u-Kd relationship (Sridharan and Jayadeva 1982)

Sridharan and Jayadeva (1982) summarized the u and kd relationships for different values of (dy/dξ)x=0 shown in Fig. 2. A linear relationship between u and log(Kd) was suggested to compute u value for known Kd. Sridharan and Choudhury (2002) proposed u-log (Kd) for saturated slurries of Na-montmorillonites with valence of 1. Three different u-log Kd relationships were proposed as following

$$\mathrm{u} = 2.35 - 4.375\log Kd \tag{a}$$

$$\mathrm{u} = 2.10 - 4.583\log Kd \tag{b}$$

$$\mathrm{u} = 2.81 - 3.375\log Kd \tag{c}$$

Eq. (a) was constructed based on all the clays (illite, kaolinite, motmorillonite) in pressure of 10-1000KPa. Eq. (b) was the theoretical u-log(Kd) only related to montmorillonite in pressure of 50-400KPa. Eq. (c) was recalculated based on experimental Na montmorillonite data.

Tripathy *et al.* (2004) used the procedures similar to Sridharan and Choudhury (2002) to propose u-log(Kd) relations. Tripathy *et al.* (2004) proposed three methods to establish the u-log(Kd) relations. The method 1 considered all u and kd values in pressure of 50-40000 KPa and with n_0 from 10^{-5} - 10^{-3}M. The method 2 was used by consideration of all the u and Kd values from 50 to 40000KPa, with n_0 equals to 10^{-4}M. The method 3 considered u-log(Kd) in the pressure range of 50-400kpa, similar to Eq. (b). The method 3 was used in his article to establish new relationship between the nondimensional midplane potential function, u and the distance function, Kd, for MX80, FEBEX, Montigel bentonite, based on comparing the experimental results and swelling pressure derived from diffuse double layer theory.

Because the swelling pressure tests were performed under the infiltration of distilled water with a concentration of approximately 10^{-4}M, thus the method 2 were adopted as theoretical DDL values in this paper. The swelling pressure can be predicted by relating the value of u obtained from Eq. (5) and Kd for any given properties of bentonite and the known bulk fluid properties (Sridharan and Jayadeva 1982, Tripathy *et al.* 2004). For a range of assumed swelling pressure, the value of u can be obtained

Table 1 Physical properties, montmorillonite content and cation exchange capacity for all compacted bentonites

Type	Name	MMT (%)	CEC (meq/100g)				BEC (meq/100g)	G	S (m^2/g)	v	w_L(%)	w_p (%)	References
			Na	Ca	Mg	K							
Na - bentonites	MX80	75	62.4	7.4	3	0.2	73	2.76	562	1.14	411±10	47	Muller-Vonmous and Kahr (1982), (1983)
	Kunigel-V1						104.4	2.7	525	1.46	416	21	Japan nuclear Cycle Development Insitute (1999)
	Voclay	69	56.6	29.3	13.2	1.6	100.7	2.84	558.9*	1.42	628.2	44.8	Komine (2004
	Neokunibond	76	62	33	6.3	1.9	103.5	2.68	615.6*	1.38	607.5	50.69	Komine 2004
	GMZ	75.4	43.4	29.1	12.3	2.5	78.3	2.71	570	1.67	276	38	Wen (2006)
Ca - bentonites	FoCa	80	3.6	73.1	6.5	0.8	84	2.67	300	1.94	112	50	Volckaert *et al.* (2000) Marcial *et al.* (2002)
	FEBEX	92±3	25	42	32	3	102	2.7	752±47	1.73	102±4	53±3	ENRESA 2000, Lajudie et al.1996
	Montigel	66	1.8	37.6	22.4	0.2	62	2.85	493	1.97	50	50	Muller-Vonmous and Kahr (1982), (1983), Lajudie *et al.* (1996)
	Bentonite S-2	92±4	22±3	41±7	31±7	2.5±1	97	2.78	614±74	1.66	105±10		ENRESA 2000
	Bavaria		9	33	32		74	2.8	650	1.88	178	56.1	Schanz *et al.* (2009)

*Note: MMT- montmorillonite, BEC – base exchange capacity, CEC- cation exchange capacity, G – specific gravity, S – specific surface, *-value estimated form the specific surface of MMT (810 m^2/g) (Komine and Ogata 1996)

Table 2 Initial water content, compaction pressure, dimensions, laboratory temperature and reference of compacted bentonite used in the paper

Bentonite	Initial WC (%)	Compaction pressure(KN)	Dimensions		T (℃)	Reference
			Dia/mm	H/mm		
MX80		100-300	56.4	20-25	20	Bucher and Müller-Vonmoos (1989)
Kunigel-V1	6.5~10.0	8.5~145	60	5		Japan Nuclear Cycle Development Institute (1999) and Komine (2004)
Voclay	11.1~15.1	8.5~145	60	5	22±1	Komine (2004)
Neokunibond	11.3~12.3	8.5~145	60	5	22±1	Komine (2004)
GMZ	11.14		50	20		Schanz and Al-Badran (2014)
FoCa	12		38~57	12-20	24±1	Imbert and Villar (2006)
FEBEX	12.5~15.5		53	7-8	20~25	ENRESA (2000)
Montigel		100~300	56.4	20~25	20	Bucher and Müller-Vonmoos (1989)
Ben. S-2	9~11		53	7~8	20~25	ENRESA (2000)
Bavaria	9.9		50	15	20	Schanz and Tripathy (2009)

from Eq. (5) and the value of z can be calculated from Eq. (4). The value of Kd also can be calculated from Eq. (3) by knowing u and z. The value of K can be determined by Eq. (6), so the value of d can be obtained from Eq. (3). Knowing d, Eq. (1) can be used to calculate e. The dry density can be calculated from Eq. (2). The integration of Eq. (3) is evaluated numerically using the “quad” MATLAB method. Thus, the theoretic relationship between swelling pressure and dry density can be obtained.

4. Materials

Various bentonites including Na-bentonites and Ca-bentonites were studied as buffer materials for nuclear waste disposal all over the world. Such as MX80, Na-bentonites original from Wyoming, USA, and FoCa bentonite, Ca- bentonites original from France and so on, which were planned as buffer and backfill materials for high level nuclear waste disposal. In this paper, it contents five Na-bentonites and five Ca-bentonites. The detailed physical properties, montmorillonite content, cation exchange capacity, liquid limit and plastic limit were shown in Table 1. Generally, the value of swelling pressure depends on the amount of montmorillonite content. It can be seen the MX80 bentonite with the montmorillonite content is around 75%, which is the average content among all the compacted Na-bentonites in the publish papers. The FoCa bentonites with the montmorillonite content is around 80% which is also the average content among all the compacted Ca-bentonites in the publish papers. In addition, the MX80 and FoCa bentonite contains the highest sodium and calcium cation content respectively. Based on this priority, the two bentonites were selected as representative of Na- bentonites and Ca-bentonites respectively. The valencies of the bentonites were obtained from the weighted average of the valencies of the cations exist in the bentonites, which suggested by Tripathy *et al.* (2004).

The swelling pressure of compacted bentonites was usually determined in the laboratory. For preparation of each dry density of specimen, the bentonite powders were statically compacted in a special compaction mould corresponding to different compaction forces. The general condition was oedometric condition which fixing the top part of the oedometric cell. And the water was infiltrated from the bottom of the specimen, the swelling pressure is measured using the load cell placed between the restrain and the top of the compacted bentonites. The volume of the compacted bentonites keeps constant during the saturation process. The tests were conducted in normal soil mechanic laboratories which has a temperature around 20 ℃. After swelling pressure test, the water content was determined by oven drying, the degree of saturation with a value of 90-120% indicted that the specimens were almost saturated. The detailed information about the specimens such as initial state, preparation procedures, specimen dimensions, testing equipment and laboratory testing temperature were listed in Table 2.

5. Difference between theoretical and experimental swelling pressure

To calculate the theoretical swelling pressure, the u-log(Kd) relationship must be established as presented before. According to Sridharan and Jayadeva (1982) and Tripathy *et al.* (2004), the ionic concentration of the bulk fluid nearly has a molal concentration of approximately 10^{-4} M, so the concentration n_0 is assumed to 10^{-4} M in this paper. The swelling pressure varies from 0 kPa to 40 MPa for all the studied compacted bentonites. The following procedures were suggested to establishe the u-Kd relationship for all compacted bentonites: (i) assumed the pressure range which is from 50 to 40 000 KPa increased by step, u values can be obtained by Eq. (5); (ii) the value of z can be calculated from equation (4) by given the value of B, S; (iii) The value of Kd also can be calculated from equation (3) by given the value of u and z. In order to save the space, here only MX80 Na-bentonite and FoCa Ca-bentonite were listed, the calculated u and Kd values are shown in Table 2.

From Table 3, the value of $(dy/d\xi)_{x=0}$ is 211.17 and 455.208 for MX80 and FoCa bentonite respectively. According to Sridharan and Jayadeva (1982), it was noted that u plotted against log(Kd) is linear for larger values of $(dy/d\xi)_{x=0}$ and for u>1.0. The linear fitting curve of u-log(Kd) of MX80 and FoCa bentonites were shown in Fig.3. Once the relationship was obtained, the theoretical u can be calculated from Kd, in which d was calculated from Eq. (1) by assuming a series of dry density, K was calculated from Eq. (6) by the known parameters listed in Table 1. Once the value of theoretical u was got, the theoretical swelling pressure can be calculated from Eq. (5). Thus, the swelling pressure of theoretic DDL prediction of each compacted bentonite were presented in the following Eqs. (7)-(16).

Figs. 4 and 5 show the theoretical and experimental swelling pressure of sodium and calcium compacted bentonites versus dry density respectively. It can be observed that the theoretical swelling pressure of Na-bentonite corresponds to the experimental data well at low dry density for MX80 bentonite and Kunigel-V1 bentonite (<1.5 Mg/m^3).

$$p = 2n_0kT[\cosh(-3.462\log(Kd) + 3.292) - 1] \quad \text{MX80} \tag{7}$$

$$p = 2n_0kT[\cosh(-3.749\log(Kd) + 3.052) - 1] \quad \text{Kunigel–V1} \tag{8}$$

$$p = 2n_0kT[\cosh(-3.684\log(Kd) + 3.108) - 1] \quad \text{(Voclay)} \tag{9}$$

$$p = 2n_0kT[\cosh(-3.638\log(Kd) + 3.148) - 1] \quad \text{(Neokunibond)} \tag{10}$$

$$p = 2n_0kT[\cosh(-3.500\log(Kd) + 3.262) - 1] \quad \text{(GMZ)} \tag{11}$$

Table 3 Values of u, z, $(dy/d\xi)_{x=0}$ and Kd for MX80 bentonite and FoCa bentonite

Pressure /KPa	u	MX80 bentonite			FoCa bentonite		
		z	$(dy/d\xi)_{x=0}$	Kd	z	$(dy/d\xi)_{x=0}$	Kd
$n_0=10^{-4}$ M							
50	5.32	10.710	211.17	0.211	12.242	455.208	0.216
100	6.01	10.714	211.17	0.146	12.243	455.208	0.152
200	6.70	10.723	211.17	0.101	12.245	455.208	0.106
400	7.39	10.741	211.17	0.069	12.249	455.208	0.074
800	8.08	10.775	211.17	0.046	12.257	455.208	0.051
1000	8.30	10.792	211.17	0.040	12.261	455.208	0.045
2000	9.00	10.872	211.17	0.026	12.280	455.208	0.031
4000	9.69	11.015	211.17	0.016	12.317	455.208	0.020
8000	10.38	11.250	211.17	0.010	12.386	455.208	0.013
10000	10.61	11.350	211.17	0.008	12.420	455.208	0.011
20000	11.30	11.739	211.17	0.005	12.571	455.208	0.007
40000	11.99	12.236	211.17	0.002	12.818	455.208	0.004

$$p = 2n_0kT[\cosh(-3.957\log(Kd) + 2.869) - 1] \quad \text{(FoCa)} \tag{12}$$

$$p = 2n_0kT[\cosh(-3.494\log(Kd) + 3.266) - 1] \quad \text{(FEBEX)} \tag{13}$$

$$p = 2n_0kT[\cosh(-3.494\log(Kd) + 3.267) - 1] \quad \text{Bentonite Montigel} \tag{14}$$

$$p = 2n_0kT[\cosh(-3.555\log(Kd) + 3.217) - 1] \quad \text{(Bentonite S-2)} \tag{15}$$

$$p = 2n_0kT[\cosh(-3.371\log(Kd) + 3.363) - 1] \quad \text{Bavaria bentonite} \tag{16}$$

This phenomenon was observed by Komine and Ogata (1996) of sodium Kunigel bentonite. According to them, the DDL theory was only effective in prediction swelling pressure with low dry density for certain bentonites. As for other Na-bentonites, the theoretical value was higher than experimental data at low dry density, while they consist with experimental data well only among some certain ranges. In Fig. 5 for compacted Ca-bentonites, the same phenomenon was found that the theoretical value corresponds well with the experimental data among low dry density range. Meanwhile it occurred bigger difference than Na-bentonites during high dry density range. In order to see the accuracy of the DDL prediction clearly, redraw the dry density against with the difference of swelling pressure which equals to the experimental data minus the DDL calculated values. Fig. 6(a) and 6(b) shows the difference of swelling pressure with increasing dry density for Na-bentonite and Ca-bentonite respectively. Assumed the difference is within ±0.5MPa, the model can be accepted. The dash line included the area between -0.5MPa and 0.5 MPa, it is clearly that the DDL model is only suitable for certain Na-bentonites shown in Fig. 6(a), while the DDL model is unavailable for Ca-bentonite with dry density higher than 1.6 Mg/m^3 shown in Fig. 6(b). It can be concluded that the DDL theory is unavailable for compacted bentonites and it should need some revises.

It is known that swelling of bentonite (montmorillonite) has two mechanisms. One is the crystalline swelling and the other is the diffuse double layer swelling (Van Olphen 1963). Swelling of montmorillonite takes place in two distinct ways. At closed clay platelet distance (<2.2 nm) (high dry density), the expansion was primary dependent on the exchangeable cation and the hydration energy of the cations. Upon further ingress of water, when the clay platelet distance is greater than 3.5 nm, the hydrated cations will move into the solution to form the electrical diffuse double layer. The montmorillonite was thought to develop the rest of the micelle fluid and the swelling was essentially osmotic (Norrish 1954). At lower dry density, a possible dissociation of ions from the clay particle surface contributed to the diffuse double layer repulsion (Schanz *et al.* 2009). At higher dry density, the adsorptive forces due to surface and ion hydration dominated the swelling pressure. The hydration energy due to surface and ion hydration are the major contributing factors to swelling pressure at close separation of clay platelets (Verwey and Overbeek 1948, Van Olphen 1963 and Yong 1999). The swelling pressure can be regard as equivalent to the double layer repulsive pressure only when double layer repulsion is the dominated force. It is clearly to see that there are some limited conditions when using the DDL theory. The effects of ion and surface hydration energy may have effects on the swelling pressure of compacted bentonite at very close separation distance. However, the DDL theory doesn't consider this influence.

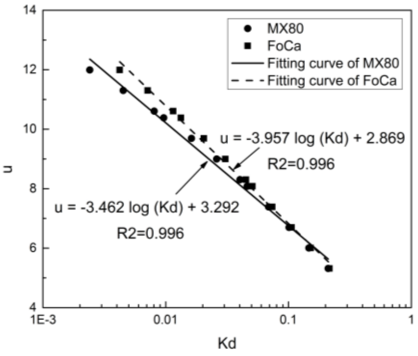


Fig. 3 Theoretic u-Kd relationships of MX80 and FoCa bentonite

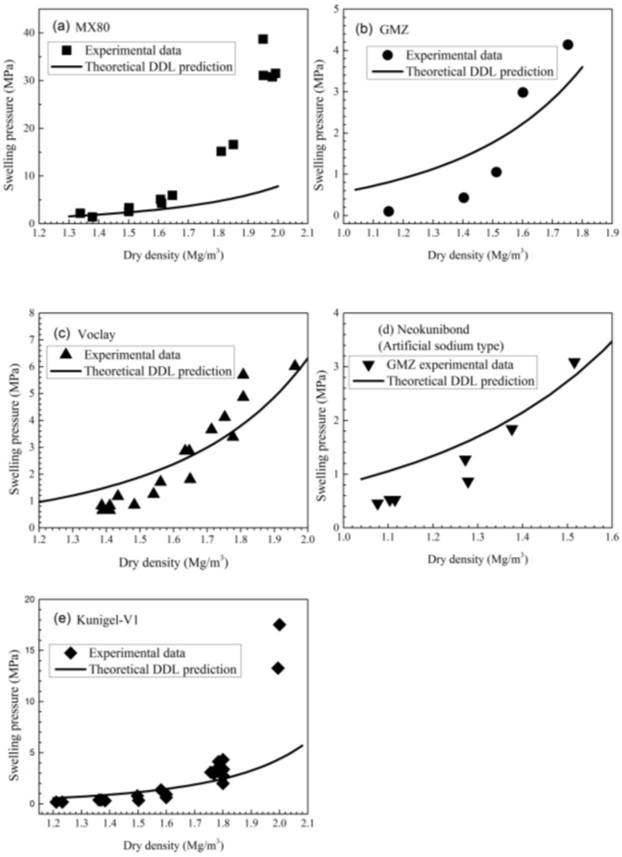


Fig. 4 Theoretical and experimental swelling pressure versus dry density for Na-bentonites, (a) MX80 bentonite, (b) GMZ, (c) Voclay, (d) Neokunibond and (e) Kunigel-V1

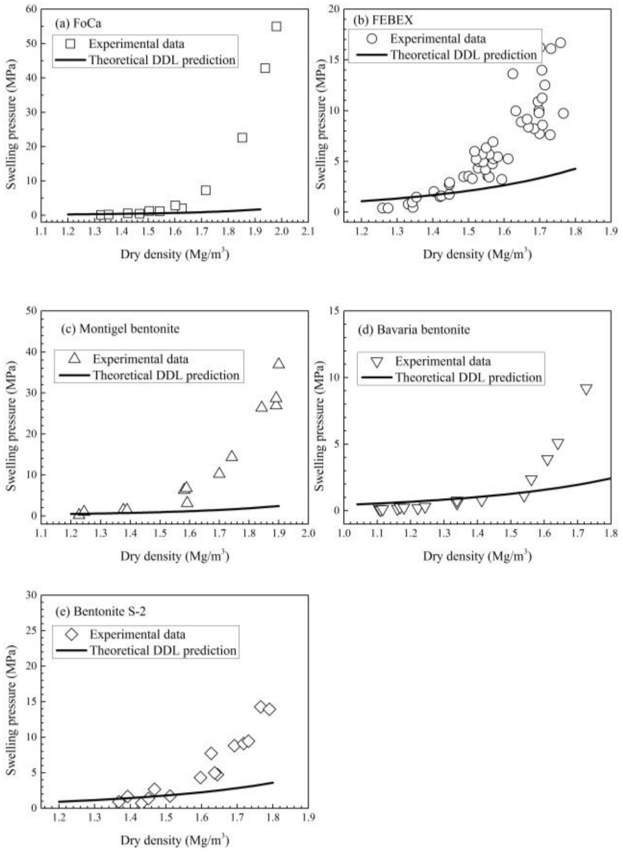


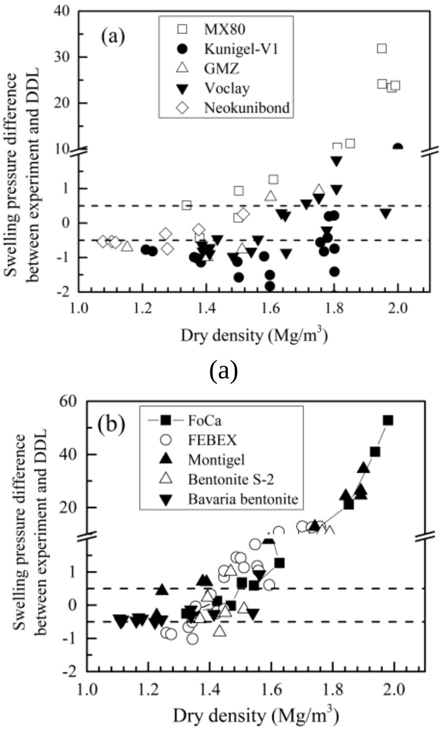
Fig. 5 Theoretical and experimental swelling pressure versus dry density for Ca-bentonites, (a) FoCa bentonite, (b) FEBEX, (c) Montigel bentonite, (d) Bavaria bentonite and (e) Bentonite S-2

It was noted that several factors may significantly influence the swelling pressure of compacted bentonite other than the one only predicted by diffuse double layer theory. There are different kinds of cations existing in compacted bentonite, which has different hydration radius and hydration energy, and they can contribute to swelling pressure. For higher dry density, the distance between clay platelets is much closer, even their respective force fields overlap and have a great influence on the behavior of the clay-electrolyte system. Therefore, it needs developed of the basic DDL theory used for predicting the swelling pressure of compacted bentonite.

6. Suggested new relationship of u-log(Kd)

The effects of ions and surface hydration energy can deeply influence the swelling pressure of compacted bentonite at very close separation distance as previous discussed. The swelling pressure is initially predicted by Eq. (5). The parameter n_0, k and T in Eq. (5) are assumed to be constant in this article, the theoretical swelling pressure only depends on the non-dimensional mid-plane potential u. The value of u is the reason for the difference between theoretical DDL prediction and experimental data. It is necessary to compare the theoretical and experimental u and study their difference. From experimental results, the u values can be calculated by knowing the swelling pressure by Eq. (5). K values can be obtained by Eq. (6). The values of d can be calculated by Eq. (1), thus Kd values can be obtained. The void ratio corresponding to each dry density also can be calculated by Eq. (1).

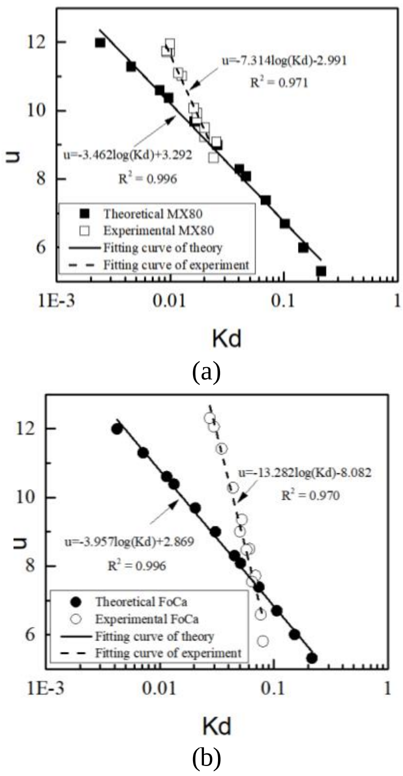
Considering the difference of preserve cations in different bentonite, the mineral components and the difference between theoretical and measured swelling pressure (Table 1, Fig. 6). Here, the MX80 and FoCa bentonite contained the highest sodium and calcium content respectively, also they have the average montmorillonite content among Na-bentonite and Ca-bentonite. Therefore, MX80 and FoCa bentonite can be regarded as the representative of Na-bentonites and Ca-bentonites respectively. Fig. 7 shows the theoretical and actual experimental relationship of u and log(Kd) for MX80 bentonite and FoCa bentonite. The slope of experimental data is much lower than theoretical value for both bentonites, which results the difference between theoretical and experimental results. Considering Fig. 6, it is obvious that the difference between the DDL predicted and experimental swelling pressure seems to have an exponential increase trend against the dry density for all bentonites.



(a)

(b)

Fig. 6 The swelling pressure difference between experimental data and DDL theoretical values versus dry density for (a) Na-bentonites and (b) Ca-bentonites



(a)

(b)

Fig. 7 The theoretical and experimental u-log(Kd) relationship of (a) MX80 bentonite and (b) FoCa bentonite

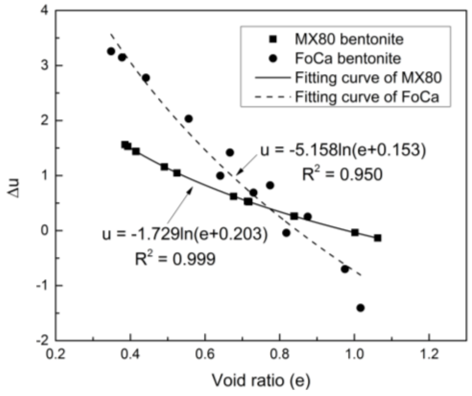


Fig. 8 Relationship between Δu and void ratio of (a) MX80 bentonite and (b) FoCa bentonite

Once the experimental u and log(Kd) relationship was obtained, the difference in u values, Δu, can be calculated using experimental u values minus theoretical DDL u values. Because of the limit of DDL, especially its inaccurately predicted at high dry density range, which the

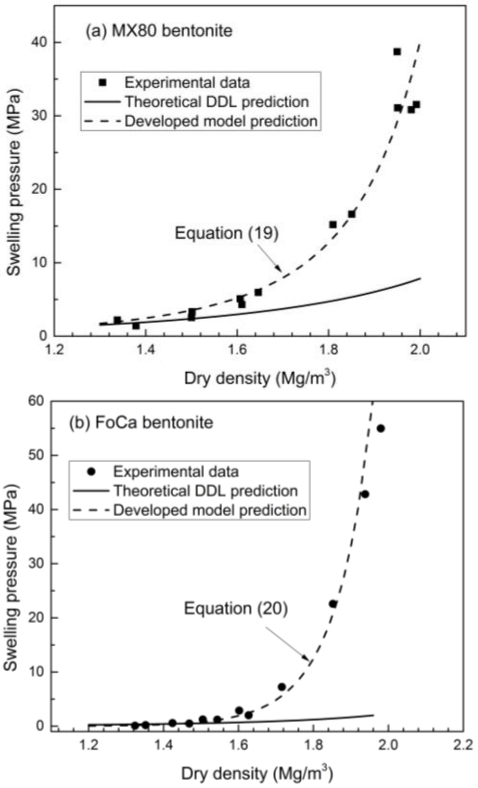


Fig. 9 Theoretical DDL prediction, experimental and developed model for swelling pressure of (a) MX80 bentonite and (b) FoCa bentonite

hydration energy of ions or repulsive forces mainly controls (Schanz and Tripathy 2009). It results in the difference of swelling pressure between theoretical and experimental data. The difference Δu may compensate the hydration energy, repulsive pressure etc. which are not considered in DDL theory. Because the swelling pressure has relations to u (Eq. (5)), and u has relations with clay platelets d (equation 3), d has relations with e (Eq. (1)), so the difference of u should follow some function of void ratio. From the expression of Eq. (5), an attempt was made to revise the relationship between u and log(Kd).

An idea came out that the difference of experimental and theoretical non-dimensional mid-plane potential, Δu, should follow the same trend with dry density. The growth form of hyperbolic function cosh(u) and exponential function follows the same style. Based on this, it is assumed that the Δu has an exponential increased with dry density. Considering the difference of swelling pressure is negative below dry density of 1.5 Mg/m^3 and positive in the following dry density, the Δu would have a natural logarithm decreased with increased of void ratio. Here we choose the void ratio instead of dry density considering the non-dimension of void ratio. Based on the experimental information of MX80 and FoCa bentonite, the relationship between Δu and e were established for Na-bentonites and Ca-bentonites. The Δu versus void ratio (e) of MX80 and FoCa bentonite was presented in Fig. 8.

The natural logarithm relationship between Δu and void ratio (e) can be obtained. Their relationships were shown in Eqs. (17) and (18). The values of Δu decrease with the increasing of void ratio. The new modified nondimension midplane potential can be got by added Δu to theoretical u. And the swelling pressure can be recomputed by Eqs. (19) and (20) for MX80 and FoCa bentonite respectively. Fig. 9 shows swelling pressure versus dry density of MX80 and FoCa bentonite by the experimental, DDL theory and proposed Eqs. (19) and (20). It can be seen the proposed equations predicted much better than the original DDL theory at high dry density range. The proposed equation has a good agreement with the experimental data. Eqs. (21) and (22) were the proposed equations for calculating swelling pressure of Na-bentonite and Ca-bentonite.

$$\Delta u_{MX80} = -1.729\ln(e+0.203) \quad \text{(Na - bentonite)} \tag{17}$$

$$\Delta u_{FoCa} = -5.158\ln(e+0.153) \quad \text{(Ca - bentonite)} \tag{18}$$

$$p = 2n_0kT[\cosh((-3.462\log(Kd) + 3.292)-1.729\ln(e+0.203)) - 1] \tag{19}$$

$$p = 2n_0kT[\cosh((-3.957\log(Kd) + 2.869)-5.158\ln(e+0.153)) - 1] \tag{20}$$

$$p=2n_0kT[\cosh(u_{\text{theory}} + \Delta u_{MX80}) - 1] \tag{21}$$

$$p=2n_0kT[\cosh(u_{\text{theory}} + \Delta u_{FoCa}) - 1] \tag{22}$$

7. Verification of proposed equations for swelling pressure

The modified swelling pressure equation (21) and (22) were derived based on experimental MX80 and FoCa bentonite data and DDL theory, which were proposed for Na- and Ca-bentonite respectively. And later the equations

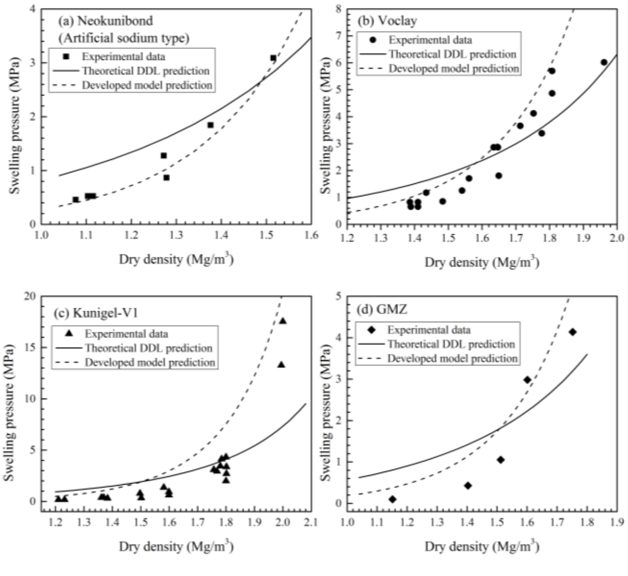


Fig. 10 Theoretical DDL prediction, experimental and developed model for swelling pressure of Na-bentonites (a) Nekunibond bentonite, (b) Voclay bentonite, (c) Kunigel-V1 bentonite and (d) GMZ bentonite

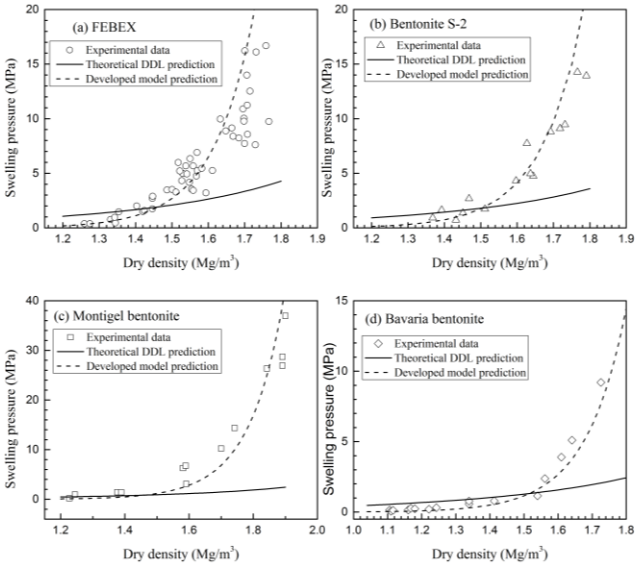


Fig. 11 Theoretical DDL prediction, experimental and developed model for swelling pressure of Ca-bentonites (a) FEBEX bentonite, (b) Bentonite S-2, (c) Montigel bentonite and (d) Bavaria bentonite

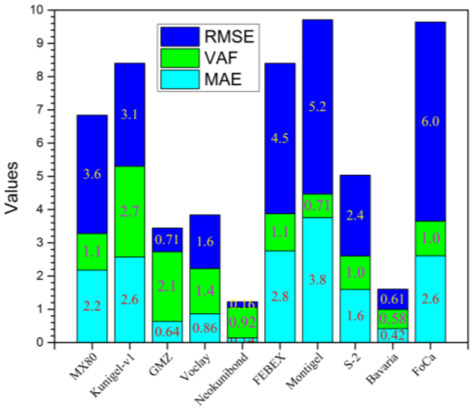


Fig. 12 Theoretical DDL prediction, experimental and developed model for swelling pressure of Ca-bentonites (a) FEBEX bentonite, (b) Bentonite S-2, (c) Montigel bentonite and (d) Bavaria bentonite

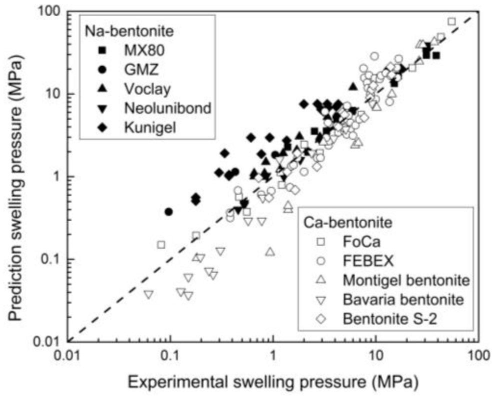


Fig. 13 Experimental swelling pressure versus prediction swelling pressure for all the compacted bentonite

will be applied in other nuclear waste barrier materials as listed before. Detailed as following: experimental swelling pressure data of FEBEX bentonite (ENRESA 2000) from Spain, Kunigel V1 (Japan Nuclear Cycle Development Institute 1999), Voclay and Neokunibond bentonite (Komine 2004) from Janpan, bentonite S-2 (ENRESA 2000) from Spain, Bavaria Bentonite (Schanz and Tripathy 2009) and Montigel bentonite (Bucher and Müller-Vonmoos 1989) from Germany, GMZ bentonite (Schanz and Al-Badran 2014) from Gaomiaozi County (Inner Mongolia, China) were selected for the verification of the new modified swelling-pressure equations. The theoretical u-log(Kd) relationship were presented from Eqs. (7)-(16). The experimental u-log(Kd) can be obtained by assumed the known swelling pressure as mentioned before.

The swelling pressure of Na-bentonites and Ca-bentonites, reported by experimental tests, calculated by theoretical DDL prediction and recalculated by proposed equations, versus dry density relationship are plotted in Figs. 10 and 11, respectively. It can be seen that the proposed equations can predict the swelling pressure more accuracy than the DDL prediction.

In order to discuss the accuracy of the developed model, the variance accounted for two values (VAF), mean absolute error (MAE) and the root mean square error (RMSE) which were presented by Eqs. (23), (24) and (25) respectively, were also computed to evaluate the performance of the proposed model. VAF, MAE and RMSE are frequently used to evaluate the difference between values predicted by a model and the measured values actually obtained in laboratory.

$$\mathrm{VAF} = \left[1-\frac{\mathrm{var}(X_{exp,i}-X_{model,i})}{\mathrm{var}(X_{exp,i})}\right]\times 100\% \tag{23}$$

$$\mathrm{MAE} = \frac{1}{n}\sum_{i=0}^{n}\left|x_{exp,i}-x_{model,i}\right| \tag{24}$$

$$\mathrm{RSEM} = \sqrt{\frac{\sum_{i=0}^{n}\left(X_{exp,i}-X_{model,i}\right)^{2}}{n}} \tag{25}$$

where $x_{exp,i}$ and $x_{model,i}$ is the experimental value and the model predicted value at i stage, respectively. n is the observation times.

If VAF equals to 100%, MAE and RMSE equals to zero, in which the experimental value equals to model predicted value, it means that the model is quite perfect. The higher the VAF value and the lower the MAE and RSEM values, the better the model works. The swelling pressure predicted by the proposed equations are compared with their experimental data. All the values of each compacted bentonite are shown in Fig. 12.

In Fig. 13, the abscissa represents the experimental swelling pressure and the ordinate represent the model prediction swelling pressure of all the bentonites. The model prediction values are very accurate when all the points located in diagonal line (dash line). In other words, the nearer the points closed to the line, the more accuracy the model is. The Kunigel-V1 values are above the line at 0.3-09 MPa range, which means that the model predicted a little higher than the experimental data. This phenomenon can be confirmed by the highest VAF value of Kunigel-V1 in Fig. 12. At lower swelling pressure range 0.06-0.3 MPa, the Bavaria bentonite data is below the line, which means that the model predicted much lower than the experimental data. Due to the logarithmic-logarithmic coordinates, a tiny difference would reflect a big difference in the Figures. At this time, the MAE values will help us to evaluation the accuracy of the proposed model. Results show that the prosed model work well for Bavaria bentonite. The agreement between experimental and model predicted value can be observed in Fig. 13. The results show that the proposed model prediction Eqs. (21) and (22) worked well for Na-bentonites and Ca-bentonites.

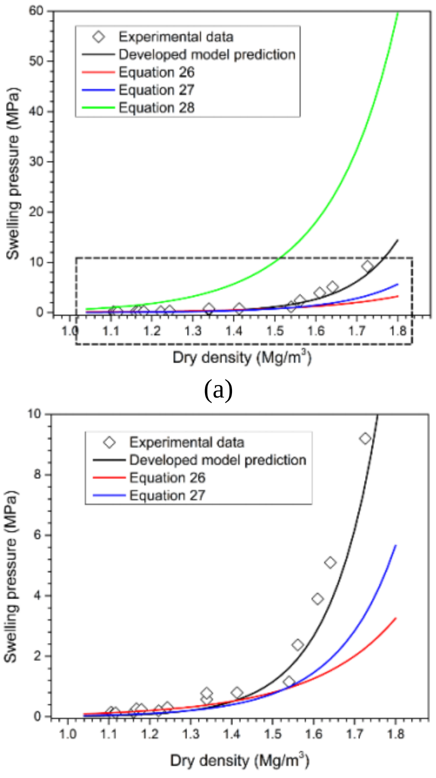
8. Comparison with other developed DDL model

In order to compare the proposed model with previous published models especially for divalent rich bentonite, the Bavaria bentonite was a good candidate. The modified swelling pressure equations proposed by Tripathy *et al.* (2004) considering the weight average valency of cations in clay are as following

$$p = 2nkT[\cosh(-7.277\ \log_{10} Kd - 2.91) - 1] \quad \text{(for v = 1.14 -1.50)} \qquad (26)$$

$$p = 2nkT[\cosh(-10.427\ \log_{10} Kd - 7.72) - 1] \quad \text{(for v = 1.66 −1.73)} \qquad (27)$$

$$p = 2nkT[\cosh(-9.190\ \log_{10} Kd - 3.26) - 1] \quad \text{(for v = 1.97)} \qquad (28)$$



(a)

(b)

Fig. 14 Experimental data and model prediction values of Bavaria bentonite (a) and Zoom in dash area (b)

Because the average valency is 1.88, so the equation 28 was adopted to calculate the swelling pressure. The calculated results were shown in Fig. 14 with green line, it was far higher than the experimental results. Also, the Eqs. (26) and (27) were also used to calculate the swelling pressure as shown in Fig. 14. Results show that both equations predicted well below dry density of 1.55 Mg/m^3. Because Eqs. (26) and (27) were mainly for certain valence range, both is not suitable for Bavaria bentonite. It can be seen that the proposed model in the paper predicted well for Bavaria bentonite.

9. Conclusions

The swelling pressure of several bentonites was calculated from Gouy-Chapman diffuse double layer theory and compared with the experimental data. Results show that the original theoretical DDL prediction can't predict swelling pressure well. It predicted much lower at high dry density for all the bentonites. Many factors contribute to this as discussed in the paper. Based on the MX80 representing Na-bentonite and FoCa representing Ca-bentonite data, the modified new relationship between nonodimension midplane potential, u, and distance function, Kd, were derived. The new relationships were suggested to calculate swelling pressure of other bentonites. The proposed equations were verified by other compacted bentonites such as Kunigel, FoCa, Bentonite S-2 and Montigel bentonite, Voclay, Neokunibond, GMZ bentonite, FEBEX and Bavaria bentonite. The swelling pressures calculated by proposed equations indicated a good agreement with the experimental swelling pressures. The proposed equations are valid in predicting of swelling pressure of compacted bentonite.

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