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PREDICTION OF COMPRESSIBILITY DATA FOR HIGHLY PLASTIC CLAYS USING DIFFUSE DOUBLE-LAYER THEORY

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Abstract—Montmorillonite-rich clays are important in many engineering applications. The compressibility of such plastic clays at high consolidation pressures is important for predicting routine settlement and for applications in nuclear-waste repositories. Laboratory measurement of compressibility data at high consolidation pressures is not only time consuming but very expensive also. Theoretical predictions can help to determine the compressibility of plastic clays at high consolidation pressures. A linear relationship between e/e_N *vs.* $1/\sqrt{P}$ (e_N is the normalization void ratio at normalization pressure N and P is the consolidation pressure) was derived using diffuse double-layer theory. The compressibility data of several plastic clays in published studies were found to support the derived relationship. A generalized theoretical equation was proposed to predict the compressibility data over a wide range of consolidation pressures using an experimentally measured void ratio at low consolidation pressure. The compressibility data for different plastic clays were predicted accurately up to maximum consolidation pressures that ranged from 0.7 to 30 MPa using an experimentally measured void ratio near the pre-consolidation pressure. The pre-consolidation pressures for different clays considered here ranged from 25 to 133 kPa. The proposed predictive model is supported by experimental data, is simple, and does not require knowledge of clay-surface and pore-fluid parameters.

Key Words—Compressibility Behavior, Diffuse Double-Layer Theory, High Consolidation Pressures, Plastic Clays, Predictive Model.

INTRODUCTION

Compacted plastic clays that contain high percentages of montmorillonite minerals (*e.g.* bentonitic clays) are of much interest as barrier material in waste-disposal facilities for isolation of toxic wastes (Pusch and Yong, 2006; Tripathy and Schanz, 2007; Zheng *et al.*, 2011; Sellin and Leupin, 2014). Compacted plastic clays are also used extensively as backfilling materials for sealing the excavated tunnels (Tripathy and Schanz, 2007). Typical initial void ratios of such highly plastic clays are in the range 10–30 (Marcial *et al.*, 2002). The void ratio is defined as the ratio between the volume of soil voids and the volume of soil solids.

The *in situ* settlement behavior of plastic clays under the anticipated pressures is of great interest for engineering applications. Compressibility data (*i.e.* interrelationship between void ratio (e) and consolidation pressure (P)) from laboratory consolidation tests on representative clay samples are useful for predicting the *in situ* settlement. The determination of compressibility data is, therefore, undertaken routinely in the laboratory and requires several days of careful measurements.

Toxic-waste repositories are usually constructed at depths of several hundred meters below the ground surface and are surrounded by saturated host rock. The performance assessment of such repositories, therefore, requires compressibility/consolidation characteristics at consolidation pressures beyond 600 kPa. Specialized experimental apparatus is required for measuring the compressibility data at such high consolidation pressures due to limitations associated with conventional oedometers (Baille *et al.*, 2010). Measurement of compressibility data at high consolidation pressures is, therefore, not only laborious, but time consuming and expensive. Simple theoretical models are useful for predicting the compressibility data of plastic clays over a wide range of consolidation pressures.

Physicochemical factors such as clay-mineral composition, cation exchange capacity (CEC), specific surface area (SSA), and pore-fluid chemistry control the compressibility behavior of plastic clays (Bolt, 1956; Mesri and Olsen, 1971; Sridharan and Rao, 1973; Sridharan *et al.*, 1986; Mitchell, 1993; Abdullah *et al.*, 1997; Robinson and Allam, 1998; Sridharan and Choudhury, 2002; Sridharan and Choudhury, 2008; Bharat *et al.*, 2013). The applicability of diffuse double layer (DDL) theory to understand compressibility behavior of highly plastic clays and the qualitative prediction of compressibility data using DDL theory on experimental data have been reported by several researchers (Bolt, 1956; Sridharan and Jayadeva, 1982; Tripathy *et al.*, 2004; Tripathy and Schanz, 2007). Relationships between non-dimensional, mid-plane electric potentials and the distances between the particles for different ranges of 'averaged' valences using the DDL

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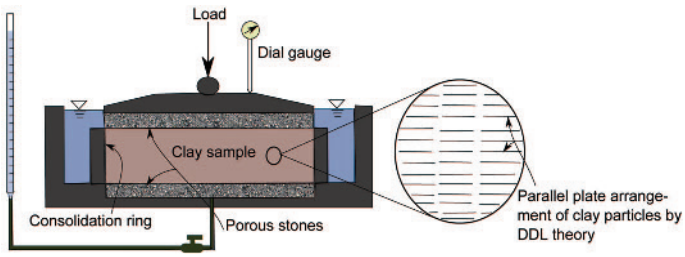


Figure 1. Laboratory oedometer set-up for the measurement of consolidation data.

theory were established by Tripathy *et al.* (2004). The established theoretical relationships were modified by adjusting the equation coefficients to match the experimental compressibility data of commonly used bentonitic clays, *i.e.* MX80, Febex, and Montigel. The established empirical relations were, however, derived from specific experimental data and were valid for limited ranges of averaged ion valences *i.e.* 1.66−1.73 and 1.97. The applicability of these equations, therefore, is not guaranteed for a wide range of clays. This method, further, requires the determination of clay-surface and pore-fluid parameters for prediction. The quantitative prediction of compressibility data of plastic clays for a wide range of such clays remains to be completed, however.

The objective of the present study was to predict quantitatively the compressibility data using theoretical models. A linearization model for compressibility data using DDL theory was developed which was supported by the experimental data. A predictive model was then developed using the proposed linearization procedure for predicting the compressibility data using three normalization void ratios. A generalized theoretical equation relating the normalized void ratio with $\sqrt{P}$ was derived to predict the compressibility data over a wide range of consolidation pressures using a single experimentally measured void ratio at pre-consolidation pressure. The performance of the predictive model was validated by comparison with several measured compressibility data from the literature. The proposed method offered several advantages over the existing methods as it did not require knowledge of clay-surface and pore-fluid parameters, unlike previous methods. The new method will reduce the cost of routine laboratory experimentation and the cost of experimentation on specialized and sophisticated equipment for measuring the compressibility data at high consolidation pressures. The method requires only a single experimentally measured void ratio value, just beyond the pre-consolidation pressure.

MATERIALS AND METHODS

Diffuse double-layer theory

The volume of a laterally confined clay specimen decreases with increase in the external pressure. The compressibility behavior of plastic clays is studied using laboratory consolidation tests (Figure 1). The void ratios of saturated clay specimens are measured under applied consolidation pressure to establish the constitutive relationship between e and P. The compressibility behavior of clays containing large percentages of montmorillonite (*e.g.* bentonitic clays) under externally applied pressures depends on physicochemical interaction between the mineral surfaces and the chemical composition of the surrounding liquid phase, *i.e.* pore-fluid. The compressibility behavior of clays can, therefore, be studied theoretically if the clay-water-electrolyte system is assumed to consist of a parallel plate system (Figure 1). The interaction between clay particles and the surrounding diffuse double layer containing exchangeable ions leads to the existence of repulsive pressures between the particles (Figure 2). The osmotic pressure (ϕ_d) thus developed between the interacting diffuse double layers at equilibrium is the measure of

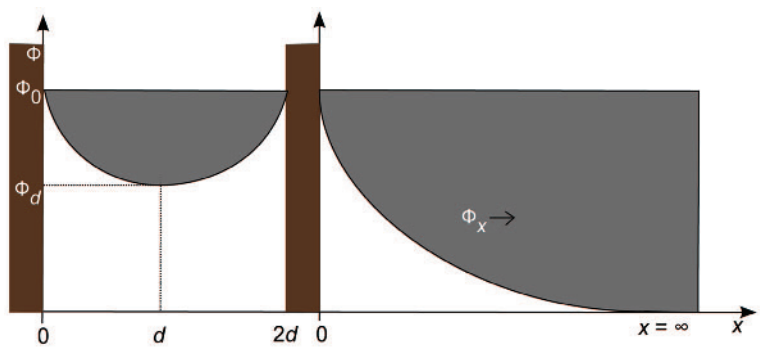


Figure 2. Electrostatic potential distribution around parallel and single clay platelets according to GC theory.

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consolidation pressure (Bolt, 1956). The mid-plane distance ($x = d$) between the clay particles at equilibrium is a measure of void ratio and is a measurable parameter in the laboratory. The decrease in particle distance results in increase in the osmotic pressure (or decrease in void ratio) with increase in the consolidation pressure in a macro-scale representation. The DDL theory is, therefore, useful in relating the microscopic parameters (ϕ_d, d) with the macroscopic compressibility parameters (P, e) of clay by assuming a parallel plate configuration of DDL plates (Figure 2). Changes in the thickness of DDLs due to changes in the pore-fluid composition can be related directly to the changes in void ratio at a given consolidation pressure using the theory.

The Gouy–Chapman (GC) model, proposed independently by Gouy (1910) and Chapman (1913), assumes Boltzmann distribution of ions in the double layer (Verwey and Overbeek, 1948). The GC model is the most commonly used DDL theory to understand qualitatively the compressibility behavior of plastic clays due to variations in the clay–water–electrolyte interactions (Sridharan and Jayadeva, 1982; Yong and Mohamed, 1992; Tripathy *et al.*, 2004; Bharat *et al.*, 2013; Sridharan, 2014). The assumptions associated with the GC model (McBride, 1997), however, limit the application for quantitative understanding of the engineering behavior (Sridharan and Jayadeva, 1982). Some of the limitations of GC theory are: (1) The clay-water-electrolyte system is assumed to consist of parallel DDL plates. The fabric of the clay, however, changes due to several factors. The effect of particle orientation (Anandarajah and Lu, 1991) is too complex to incorporate in the analysis. (2) The cations are assumed to be point charges and distributed around the particles (van Olphen, 1963). The size of the cations is not taken into consideration in the theory, therefore. Cation size was shown by Sridharan *et al.* (1986) to influence the compressibility of clays. (3) The theory is not applicable to analysis of the consolidation behavior of clays when the pore-fluid concentration is >0.001 M. The anion concentration dominates in the DDL when the pore-fluid concentration is >0.001 M (Yong *et al.*, 1962). (4) A precise estimate of the surface properties of the clay particles is required for the theory. Precise measurement of clay-surface propertiesis difficult, however (Hang and Brindley, 1970; Chan *et al.*, 1984; Santamarina *et al.*, 2002).

All the aforementioned limitations affect the other models also. The Stern model distinguishes the presence of different cations of the same valence in the Stern layer, but the application of such theories is limited to a system of constant surface potential (van Olphen, 1963; Chan *et al.*, 1984; Tripathy *et al.*, 2014). The Stern model cannot, therefore, be used to study the compressibility behavior of clays, where interacting DDL clay platelets are encountered (Tripathy *et al.*, 2014). The GC theory is, therefore, found to be the only alternative for qualitative understanding of the engineering behavior of clays, where interacting DDL plates are encountered. Theoretical predictions for the compressibility behavior are scarce due to the aforementioned limitations in the theory. A novel approach was introduced here for quantitative prediction of compressibility behavior of plastic clays using a GC model.

Experimental results

The compressibility data of several plastic clays with different clay-surface properties and pore-fluid chemistries were considered in the literature. Initial void ratios for all of the selected clays were in the range of 13 (Oikawa, 1987) to 41 (Low, 1980). Compressibility data of these clays were linearized for developing a predictive model. The experimental data were, further, used to validate the proposed predictive model. The relevant properties of the soils as reported in the literature are summarized in Table 1. The experimental compressibility data of all of the selected plastic clays showed (Figure 3) that the relationship between void ratio and consolidation pressure was very non-linear and was dependent on the clay-water-electrolyte properties. The compressibility data were generally convex beyond a consolidation pressure of 30 kPa. The pre-consolidation pressure for most of the selected plastic clays was, therefore, <50 kPa. The differences in the compressibility data for different clays were, however, significant up to a consolidation pressure of ~1000 kPa.

Table 1. Properties of plastic clays.

Clay type	Properties						Reference
	Specific gravity	Liquid limit (%)	Plastic limit (%)	CEC (meq/100 g)	SSA (m^2/g)	% Clay	
Na-Ca MX80	2.65	520	62	68.0	700	85	Marcial *et al.* (2002)
Na-Kunigel	2.79	474	27	73.2	687	64	Marcial *et al.* (2002)
Upton #1	2.80[#]	1531*	–	90.0	800	100	Low (1980)
Yugoslav	2.80[#]	948.7*	–	89.5	559	100	Low (1980)

* Moisture content corresponding to 2.5 kPa consolidation pressure
[#] Assumed

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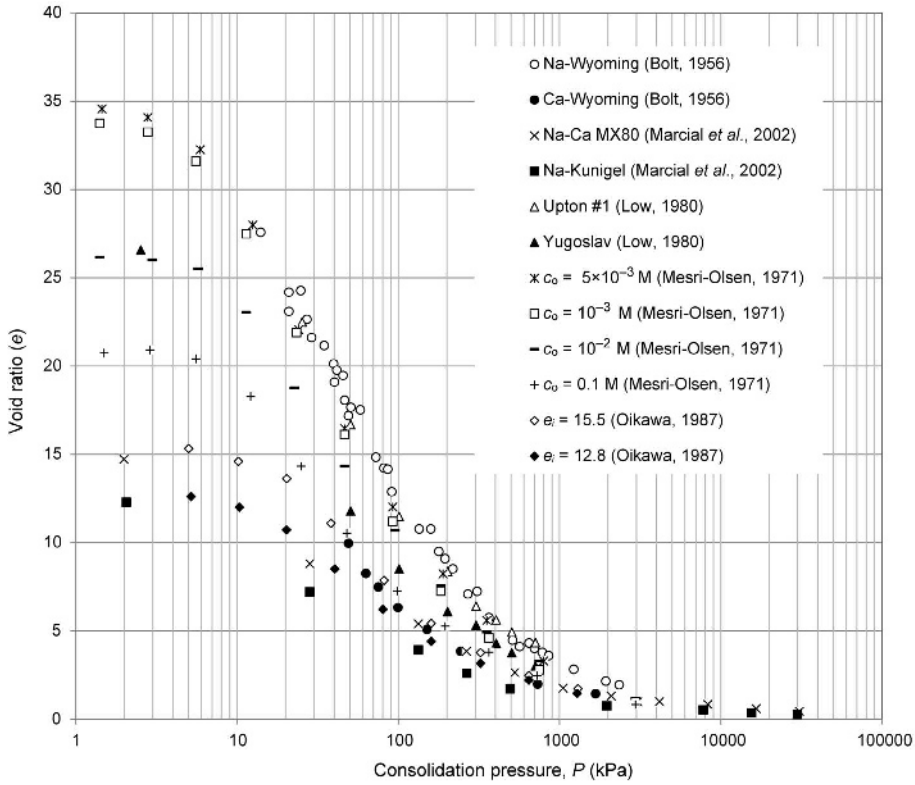


Figure 3. Experimental compressibility data for selected clays.

THEORETICAL COMPRESSIBILITY CURVES

Theoretical compressibility data for plastic clays were obtained using the GC model for developing a predictive model. Theoretical equations describing the inter-relationships between void ratio and consolidation pressure for different clay–water–electrolyte interactions were arranged as shown below (Sridharan and Jayadeva, 1982):

$$P = 2c_0 RT(\cosh y_d - 1) \quad (1)$$

$$\left(\frac{dy}{d\xi}\right)_{x=0} = -\frac{CEC}{SSA}\left(\frac{2513.6}{\sqrt{\varepsilon c_0 T}}\right) \quad (2)$$

$$\left(\frac{dy}{d\xi}\right)_{x=0} = -(2\cosh(y_0) - 2\cosh(y_d))^{1/2} \quad (3)$$

$$\kappa d = -\int_{y_0}^{y_d} (2\cosh(y) - 2\cosh(y_d))^{-1/2} dy \quad (4)$$

$$e = G\rho_w SSA d \quad (5)$$

where:
CEC = cation exchange capacity (meq/100 g)
SSA = specific surface area of the clay (m^2/g)
P = consolidation pressure (kPa)
R = gas constant
T = absolute temperature
y_d = scaled midway potential ($\nu e' \phi_d / RT$)
ξ = scaled distance (κx)
y = scaled electrostatic potential at any distance ($\nu e' \phi / RT$), x, from the surface of a single clay platelet
ε = dielectric constant,
c_0 = molar concentration of the ions in bulk solution (M)
$k_B T$ = thermal energy per ion (joules)
y_0 = scaled potential at the clay surface ($\nu e' \phi_0 / RT$)
G = specific gravity of soil particles
e = void ratio
ρ_w = density of water (kg/m^3)
d = midway distance between the particles
ϕ values are the electrostatic potentials.

The quantity $1/\kappa$ (units of length) is the characteristic length or 'Debye length' which is expressed as $\sqrt{\frac{\varepsilon k_B T}{8\pi e'^2 \nu^2 c_0}}$, where v is the cation valence in the DDL. Equations 1–5 are valid in the case of symmetrical electrolytes. The determination of the void ratio for any given clay–water–electrolyte system using the aforementioned equations involved computation of the elliptical integral (equation 4). The relationship between e and P was, therefore, established by solving equation 4 numerically and solving the simultaneous equations for P for a given void ratio. A model calculation for obtaining the void ratio from an assumed consolidation pressure using equations 1–5 is given in the Appendix below.

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Theoretical data showing void ratio *vs*. consolidation pressure relationship with the variation in clay-water-electrolyte properties obtained using the aforementioned procedure were analyzed (Figures 4a–4d). Theoretical data for different electrolyte concentrations revealed (Figure 4a) that the void ratio decreased with increase in the electrolyte concentration at a given consolidation pressure due to reduction in thickness of double-layer with increase in the concentration. The particles thus come close to each other which encourages flocculation. Decrease in the void ratio with increasing electrolyte concentration was negligible beyond consolidation pressures of 400 kPa for concentrations in the range 0.00001–0.001 M. Application of Boltzmann and Poisson's equations (equations 2–3) in DDL theory is invalid beyond an electrolyte concentration of 0.001 M for consolidation behavior (Yong *et al.*, 1962; Bharat, 2004; Tripathy *et al.*, 2004) as the influence of anion domination in the double layers is ignored in the theory (van Olphen, 1963; Frahm and Diekmann, 1979). The theoretical compressibility data for different exchangeable cation valence showed (Figure 4b) decreases in the void ratio with increasing ion valence which encourages the tendency towards flocculation. Theoretical results were consistent qualitatively with the experimental results of Sridharan *et al.* (1986).

The influence of clay-surface parameters, CEC and SSA, on the compression curve was studied (Figure 4c) by considering common ranges of CEC and SSA values from the published data for different clays (Bolt, 1956; Mesri and Olsen, 1971; Low, 1980; Marcial *et al.*, 2002). The increase in the void ratio was directly proportional to the surface area for a given consolidation pressure as the particles can accommodate greater numbers of exchangeable cations on the surface. The increase in SSA, therefore, encourages the formation of thick double layers and favors dispersion of the particles. A very minor influence of CEC was noticed, however, on theoretical compressibility data.

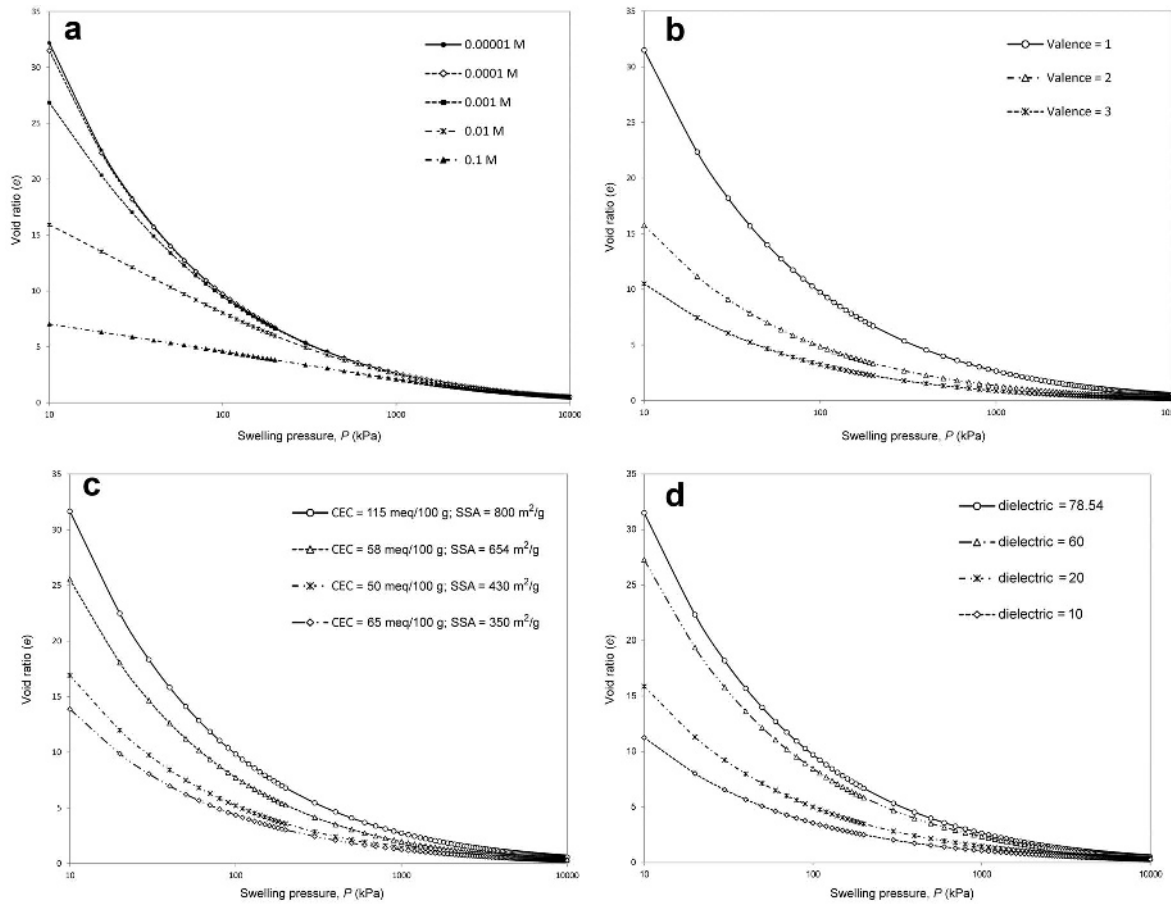


Figure 4. (a) Effect of pore-fluid electrolyte concentrations on void-ratio variation based on GC theory with pressure for assumed values of ε = 78.54; CEC = 100 meq/100 g; SSA = 800 m^2/g; v = 1; T = 298 K. (b) Effect of cation valence on void-ratio variation with pressure based on GC theory for assumed values of c_0 = 0.0001 M; ε = 78.54; CEC = 100 meq/100 g; SSA = 800 m^2/g; T = 298 K. (c) Void-ratio variation with pressure based on GC theory for different clay-surface properties for assumed values of c_0 = 0.0001 M; ε = 78.54; v = 1; T = 298 K. (d) Void-ratio variation with pressure based on GC theory for different dielectric constants of pore fluids for assumed values of c_0 = 0.0001 M; v = 1; CEC = 100 meq/100 g; SSA = 800 m^2/g; T = 298 K.

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The influence of the dielectric properties of the pore-fluids on theoretical compressibility data showed (Figure 4d) a decrease in double layer thickness with increase in dielectric constant. The range of dielectric constant values considered in the study represents different pore fluids such as water, ethyl acetate, methyl alcohol, *etc*. The theoretical results of the influence of dielectric constant were qualitatively similar to the experimental results of Sridharan and Rao (1973). The theoretical compressibility data were, therefore, qualitatively accurate and satisfactory.

LINEARIZATION OF THE COMPRESSIBILITY CURVES

The predictive model was developed in two stages. Theoretical compressibility data were normalized in the first stage. The normalized compressibility data were linearized to develop the predictive model in the second stage.

Void ratios of the compressible layer were normalized using different normalization factors, e_N (void ratios corresponding to different consolidation pressures, N). Different normalization factors, *e.g.* e_{50}, e_{100}, and e_{200}, were considered to determine the effective normalization factor. Theoretical compressibility curves are presented (Figures 5a(i)–5d) as normalized void ratio *vs*. consolidation pressure (on a logarithmic scale) using different normalization factors and for different clay-water-electrolyte parameters.

Normalized compressibility data using e_{50} as a normalizing factor for different electrolyte concentrations are presented in Figure 5a(i). All of the data converged to a smooth and unique curve beyond the normalization pressure (50 kPa) when electrolyte concentrations were $\leqslant$0.001 M. The aforementioned trend was not observed for electrolyte concentrations >0.001 M and the scatter is suggested to be due to the applicability of the Boltzmann and van't Hoff equations for electrolyte concentrations >0.001 M (Yong *et al.*, 1962; Tripathy *et al.*, 2004) where the potential distribution follows an exponential behavior (Bharat 2004; Bharat *et al.*, 2013). Plastic clays, moreover, do not display high liquid-limit void ratios and high compressibilities at such concentrations (Castellanos *et al.*, 2008) due to the suppression of DDLs. The theoretical data considered were for $c_0 \leqslant 0.001$ M, therefore. The scatter displayed before the normalization pressure is not expected to affect the prediction, however. Normalized compressibility data using e_{100} showed (Figure 5a(ii)) that the observations were similar to Figure 5a(i) except that the data converged beyond 100 kPa, the normalization pressure in this case. The data for electrolyte concentrations >0.001 M were ignored for the same reasons as mentioned above. Similar results were also observed for e_{200} (not shown here). Normalized compressibility data as e/e_{50} *vs*. P, e/e_{100} *vs*. P, and e/e_{200} *vs*. P for different clay-surface and pore-fluid parameters are presented (Figure 5b–d). The data with all the normalization factors were consistent and were observed to follow a unique relationship.

The normalized compressibility data for different clay-surface and pore-fluid parameters were presented together using three different normalization factors (Figure 6). From the fitted results, all the normalization factors, *e.g.* e_{50}, e_{100}, and e_{200}, were clearly very effective in normalizing the theoretical compressibility data. A simple power relationship was found to exist between e/e_N and consolidation pressure with an exponent of -0.5 and was valid for any clay-water-electrolyte parameters. The results were re-plotted as normalized void ratio *vs*. $1/\sqrt{P}$ (Figure 7) using different clay-surface and pore-fluid parameters as presented above (Figure 4). Linear relationships were observed with high correlation coefficients (R^2), *i.e.* 0.987, 0.988, and 0.981, using the normalization factors of e_{50}, e_{100}, and e_{200}, respectively. Only the compressibility data beyond consolidation pressure of 25 kPa ($1/\sqrt{P} < 0.2$) were considered here as the void ratios under low consolidation pressures are usually influenced by the pre-consolidation effects. The predicted correlation was, therefore, extremely good. The normalized theoretical equations can be written as:

$$e/e_{50} = K_{50}P^{-0.5} \quad (6)$$

$$e/e_{100} = K_{100}P^{-0.5} \quad (7)$$

and

$$e/e_{200} = K_{200}P^{-0.5} \quad (8)$$

where K_{50}, K_{100}, and K_{200}, expressed in $\sqrt{\text{kPa}}$ were proportionality constants equal to 6.7, 9.6, and 13.9, respectively. Equations 6–8 serve as predictive equations because the entire theoretical compressibility curve can be predicted using experimentally measured void ratios corresponding to any consolidation pressure, *e.g.* 50, 100, or 200 kPa. Furthermore, the predictive equations do not require knowledge of the clay-water-electrolyte parameters. The measured normalization void ratio (either e_{50}, e_{100}, or e_{200}) and corresponding proportionality constant (K_{50}, K_{100}, or K_{200}) contained the information about compressibility behavior of clay-water-electrolyte system implicitly. Only values of e_N and K_N were required, therefore, to predict the entire compressibility data. Comparison between equations 6, 7, and 8 revealed that the theoretical relationship using e_{50} appeared to be the most appropriate as the experiments might be conducted only up to lower consolidation pressures when the pre-consolidation pressure of the clay is <50 kPa (*i.e.* normalization pressure). The pre-consolidation pressures of highly plastic clays are, often, very small (<50 kPa).

Experimental data were analyzed to investigate the existence of similar relationships and to compare them with theoretical predictions. Empirical equations were

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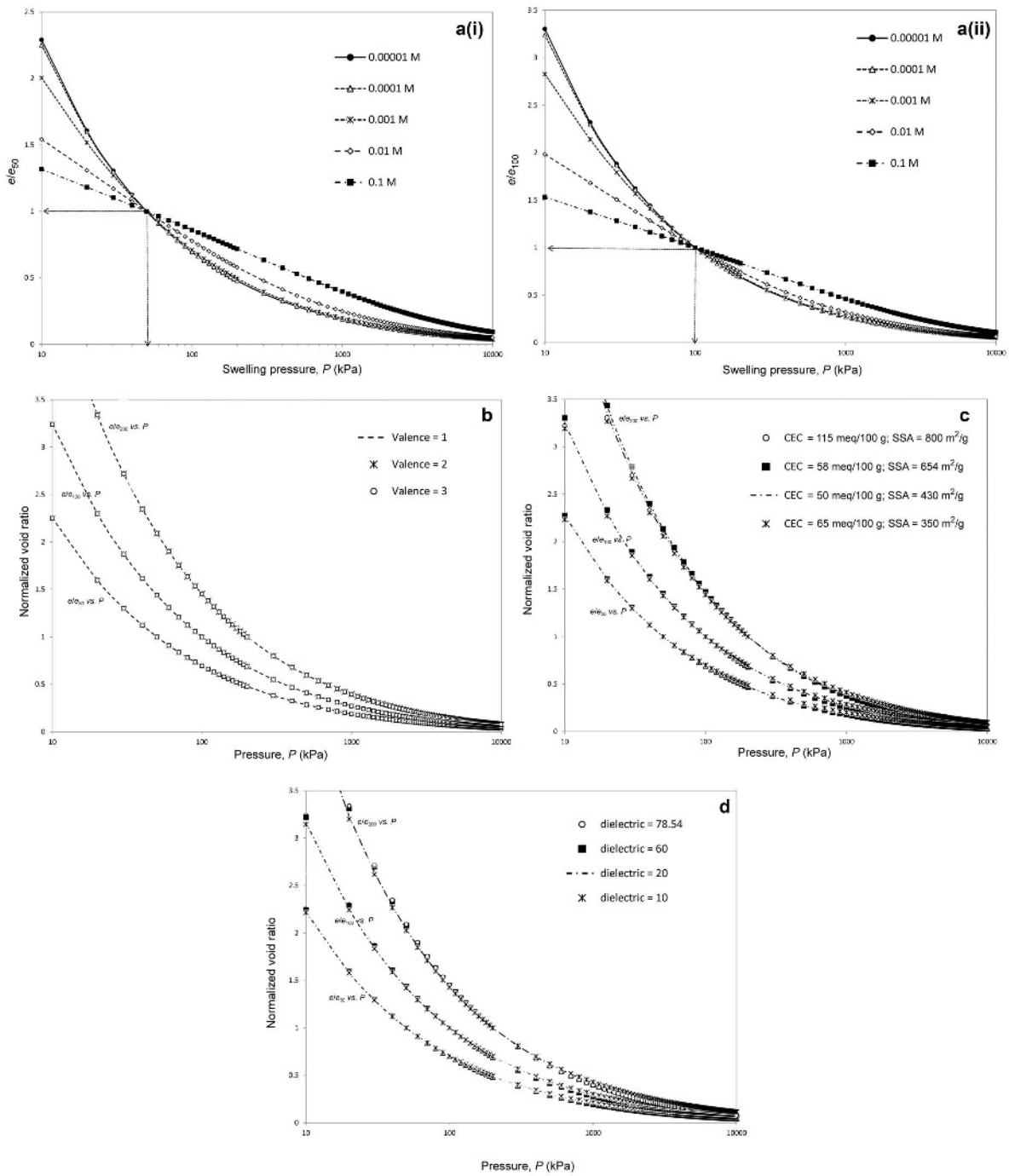


Figure 5. (ai) Normalized compression curves using e_{50} as the normalizing factor for different electrolyte concentrations for the assumed values of $v = 1$; $\varepsilon = 78.54$; CEC = 100 meq/100 g; SSA = 800 m^2/g; $T = 298$ K. (aii) Normalized compression curves using e_{100} as the normalizing factor for different electrolyte concentrations for the assumed values of $v = 1$ and for the same assumed values of ε, c_0, CEC, SSA, and T as above. (b) Normalized compression curves for different cation valence and for the same assumed values of ε, c_0, CEC, SSA, and T as above. (c) Normalized compression curves for different clay properties and for other assumed values of v, ε, c_0, CEC, SSA, and T as above. (d) Normalized compression curves for different dielectric pore fluids and for the same assumed values of v, ε, c_0, CEC, SSA, and T as above.

developed by following the aforementioned normalization and linearization procedure using the experimental data. Experimental compressibility data (Figure 3) as e/e_{50} *vs.* P are shown in Figure 8. The normalized data followed a unique relationship, in similar fashion to the theoretical data. The experimental data (Figure 3) were, therefore,

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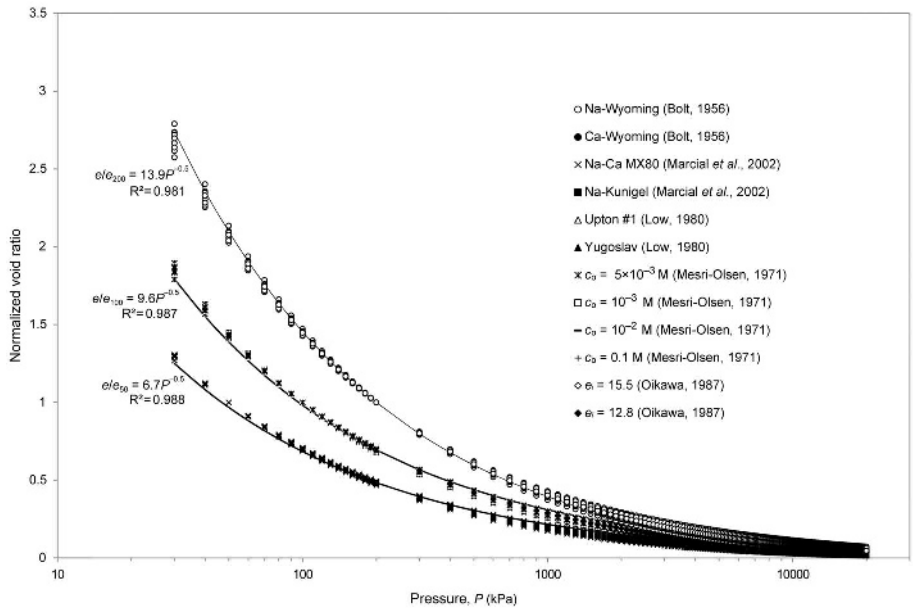


Figure 6. Normalized compression curves for all of the theoretical data presented in Figure 3.

re-plotted as normalized void ratio *vs.* $1/\sqrt{P}$ (Figure 9) using different normalization factors. The linear equations along with the regression coefficients are also presented in Figure 9. The theoretical equations and empirical relationships are compared in Table 2. The fitted empirical relationship was consistent with the predicted theoretical equations. The theoretical models based on GC theory were, therefore, accurate and reliable for predicting the compressibility data. As the pre-consolidation pressure is history dependent, a generalized predictive equation was developed in the following section to predict the compressibility data using any experimentally measured void ratio.

GENERALIZED PREDICTIVE EQUATION

The form of the predictive equation can be established from equations 6–8 as:

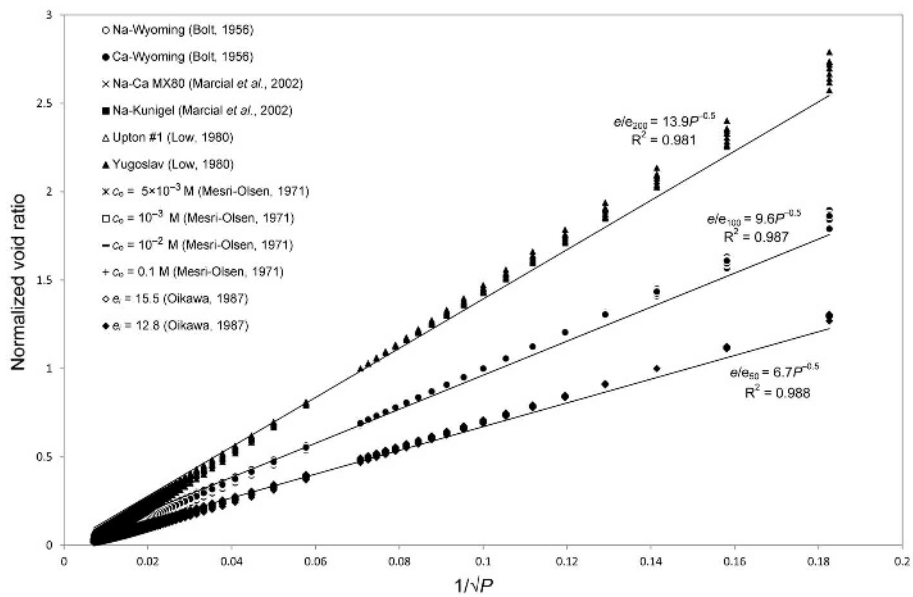


Figure 7. Normalized void ratio *vs.* $1/\sqrt{P}$ for all of the theoretical data presented in Figure 3.

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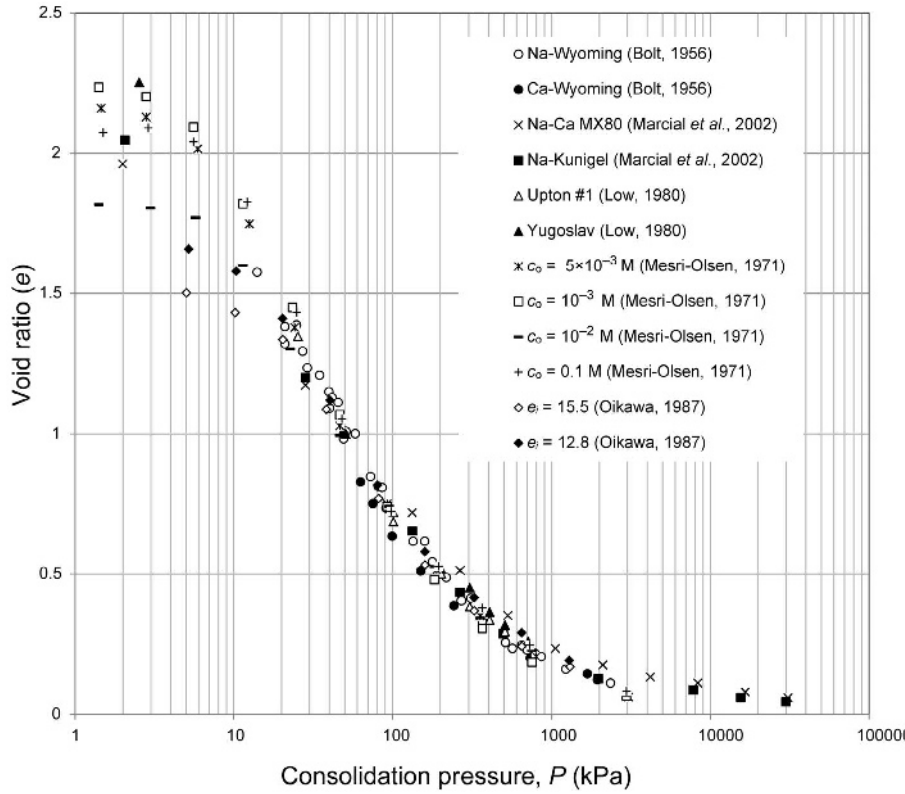


Figure 8. Normalized experimental compressibility data (presented in Figure 3) using e_{50}.

$$e\sqrt{P} = e_{Pi}K_i \quad (9)$$

where $e_{Pi=1,2,3...}$ are the normalizing factors that are void ratios under given consolidation pressures and $P_{i=1,2,3...}$

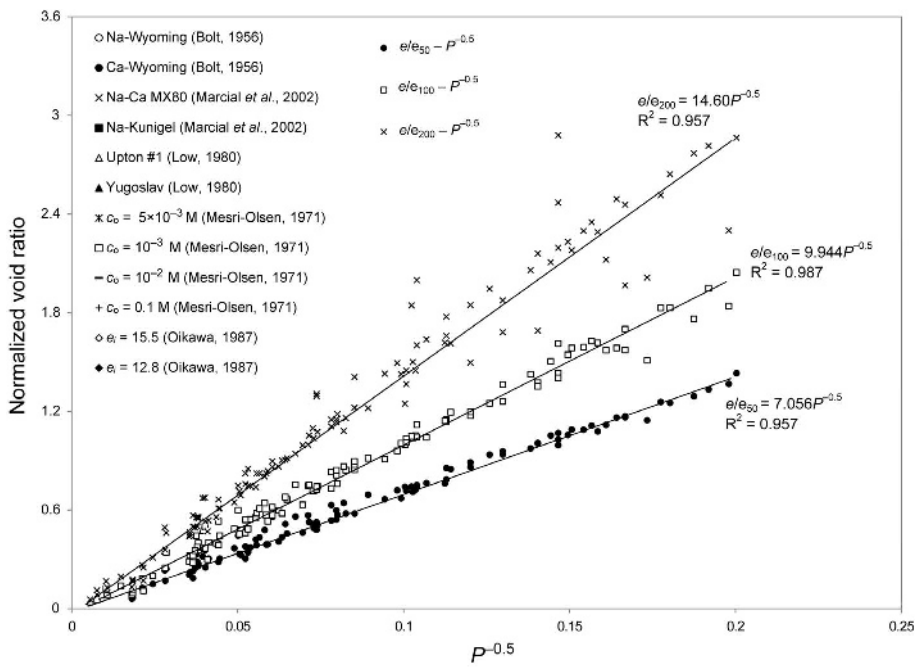


Figure 9. Linearized curves of experimental compressibility data given in Figure 3.

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Table 2. Proportionality coefficients ($\sqrt{}$kPa) of the predictive equations.

Normalizing factor	Empirical (Figure 8)	Theoretical (equations 6–8)	Generalized formula (equation 9)
e_{10}	–	–	3.2
e_{25}	–	–	5.0
e_{50}	7.1	6.7	7.1
e_{100}	9.9	9.6	10.0
e_{200}	14.6	13.9	14.1

and $K_{i=1,2,3...}$ are the proportionality constants. Equation 9 must satisfy all of the data points, *i.e.* ($e_{Pi=1,2,3...}$, $P_{i=1,2,3...}$) on the $e-P$ curve. The proportionality constants should, therefore, be

$$K_{i=1,2,3...} = \sqrt{P_{i=1,2,3...}} \tag{10}$$

The generalized equation can be written as:

$$e = \frac{e_{P_i}\sqrt{P_i}}{\sqrt{P}} \tag{11}$$

which represents a complete compressibility curve. The complete compressibility curve can, therefore, be predicted using an experimentally measured void ratio under any given consolidation pressure. Substitution of 50 kPa and 100 kPa for P in equation 10 results in

$$e = \frac{7.1e_{50}}{\sqrt{P}} \tag{12}$$

and

$$e = \frac{10e_{100}}{\sqrt{P}} \tag{13}$$

which are the same equations derived above in equations 6 and 7, respectively. The proportionality coefficients of the predictive equations based on empirical and theoretical analyses are listed in Table 2. Comparison of all the available equations revealed that the equations derived here by empirical and theoretical analysis were particular solutions of the generalized equation given in equation 11.

EXPERIMENTAL VALIDATION

Theoretical compressibility data for various clays were predicted from the measured void ratio under a low consolidation pressure. The prediction required normalization pressure and the corresponding measured void ratio. The normalization pressures for different clays were obtained using the observed variations in the nature of the compressibility curve from concave to convex. Whether the chosen normalization pressure was beyond the pre-consolidation pressure was easily verified by obtaining predicted compressibility data from equation 11 using a few experimental data points near the pre-consolidation pressure. The predicted compressibility curves using different experimental data would match exactly with each other if the selected normalization data were beyond the pre-consolidation pressure. The normalization pressures varied from 25 to 133 kPa for different clays selected in this study. Void ratios corresponding to the normalization pressures were read from the data and the proportionality constants were computed using equation 10 for different clays. Theoretical data were then predicted using equation 11 to a maximum consolidation pressure ranging between 0.7 and 30 MPa for different plastic clays

The normalization data used for the prediction are shown as open circles (○) in all of the validation plots. The predicted data are compared with the experimental data (Figure 10a–c). The compressibility data for two Na-montmorillonite clays (Low, 1980) and Akita soft clays (Oikawa, 1987) were predicted theoretically and presented (Figure 10a,b). The predicted compressibility data were based on the measured void ratios at consolidation pressures of 101 kPa and 51 kPa for the Upton #1 (Low, 1980) and Yugoslav (Low, 1980) clays, respectively. The measured void-ratio data at consolidation pressures of 38 and 40 kPa were used to obtain theoretical data for the Akita clay. The predictions were extremely good for all of the clays considered. The generalized model proposed will therefore be very useful for predicting the compressibility data over a wide range of consolidation pressures for plastic clays using data from tests carried out up to pre-consolidation pressures.

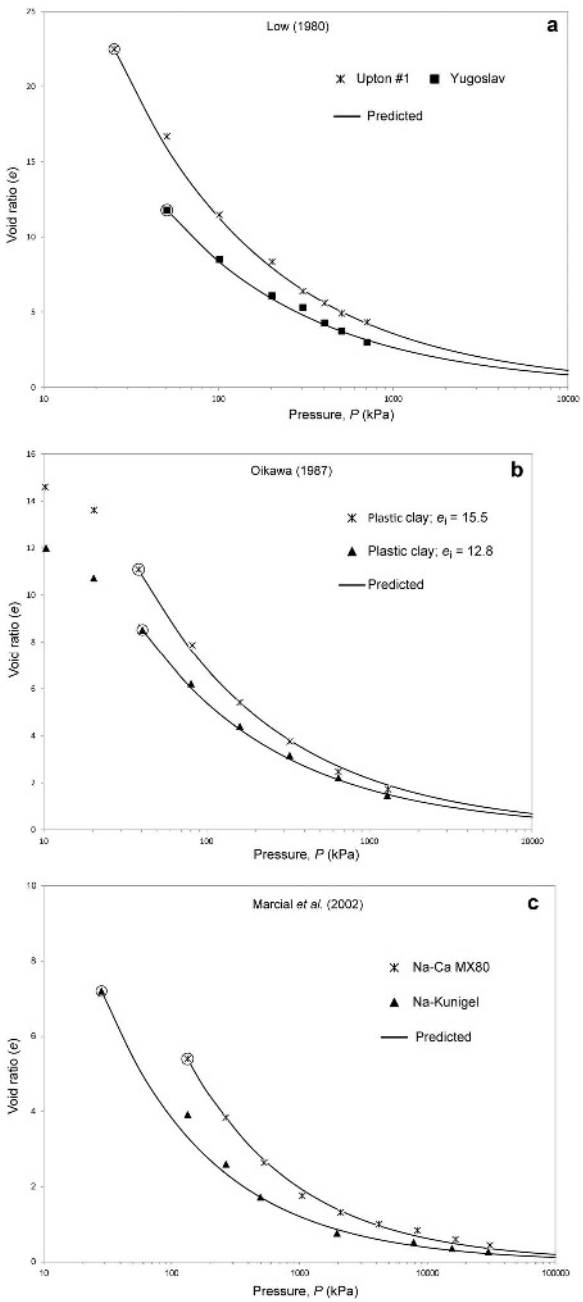
Similar validation experiments were conducted on Na-Ca MX80 and Na-Kunigel bentonites (Marcial *et al.*, 2002) using the measured compressibility data at consolidation pressures of 133 kPa and 28 kPa, respectively (Figure 10c). The theoretical prediction was consistent with the measured data. The theoretical predictions were extremely good considering the several factors involved in laboratory measurements at high consolidation pressures. The proposed model is very simple considering that the prediction requires only a single measured void ratio beyond pre-consolidation pressure.

A statistical regression analysis of theoretical predictions on the experimental data was considered. Values of the coefficient of determination, R^2, of the theoretical fit to the experimental data from validation experiments (Figure 10a–10d) are presented (Table 3). The computed R^2 values are in the range 0.993–0.999 for different experiments. The validation experiments on different clays showed that the predicted compressibility

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Table 3. Coefficient of determination values for the theoretical fit on the experimental data.

Source	Clay type	Figure number	R^2
Low (1980)	Upton #1	10a	0.995
	Yugoslav		0.994
Oikawa (1987)	Plastic clay, e_i = 15.5	10b	0.998
	Plastic clay, e_i = 12.8		0.999
Marcial *et al*. (2002)	Na-Ca MX80	10c	0.999
	Na-Kunigel		0.998



data up to a maximum pressure range (0.7 to 30 MPa) for different plastic clays were in very good agreement with the experimental data.

CONCLUSIONS

Theoretical and experimental compressibility data were linearized using the normalization factor. The normalization factor contained information about the clay-surface and pore-fluid characteristics. A unique relationship was, therefore, investigated using the normalization procedure to represent the compressibility data which was independent of clay-water-electrolyte characteristics. The proposed equation was valid for all the plastic clays tested in the study when the normalization factor was greater than the pre-consolidation pressure. Through validation experiments on several compacted plastic clays, the equations have been proven able to predict the compressibility data over a wide range of consolidation pressures with the help of measured void ratios at pre-consolidation pressure. The proposed method did not require knowledge of the clay-surface or pore-fluid characteristics, or the computation of midway potentials to relate with clay-plate distances, unlike previous predictive models. The proposed method is useful for predicting *in situ* settlement behavior of plastic clays and for special applications in nuclear-waste repositories. Application of the proposed method, therefore, will reduce the cost of routine laboratory experimentation and the need to use specialized equipment for measuring compressibility data at high consolidation pressures.

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Figure 10. (a) Predicted theoretical compression curves using equation 11 along with the experimental data for Na-saturated montmorillonites. (b) Predicted theoretical compression curves for the experimental data from the Akita clays. (c) Predicted theoretical compression curves using equation 11 along with the experimental data for Na-Ca MX80 and Na-Kunigel.

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REFERENCES

Abdullah, W.S., Al-Zou'bi, M.S., and Alshibli, K.L. (1997) On the physicochemical aspects of compacted clay compressibility. *Canadian Geotechnical Journal*, **34**, 551–559.

Anandarajah, A. and Lu, N. (1991) Numerical study of the electrical double-layer repulsion between non-parallel clay particles of finite length. *International Journal for Numerical and Analytical Methods in Geomechanics*, **15**, 683–703.

Baille, W., Tripathy, S., and Schanz, T. (2010) Swelling pressures and one-dimensional compressibility behaviour of bentonite at large pressures. *Applied Clay Science*, **48**, 324–333.

Bharat, T.V. (2004) Simplified methods of evaluation of diffuse double layer parameters employed in geotechnical engineering. MSc (Eng) thesis, Indian Institute of Science, Bangalore, India, 102 pp.

Bharat, T.V., Sivapullaiah, P.V., and Allam, M.M. (2013) Novel procedure for the estimation of swelling pressures of compacted bentonites based on diffuse double layer theory. *Environmental Earth Sciences*, **70**, 303–314.

Bolt, G.H. (1956) Physico-chemical analysis of compressibility of pure clays. *Geotechnique*, **46**, 291–311.

Castellanos, E., Villar, M.V., Romero, E., Lioret, A., and Gens, A. (2008) Chemical impact on the hydro-mechanical behaviour of high-density FEBEX bentonite. *Physics and Chemistry of the Earth, Parts A/B/C*, **33**, S516–S526.

Chan, D.Y.C., Pashley, R.M., and Quirk, J.P. (1984) Surface potentials derived from co-ion exclusion. *Clays and Clay Minerals*, **32**, 131–138.

Chapman, L.A. (1913) Contribution to the theory of electrocapillarity. *Philosophical Magazine*, Series 6, **25**,475–481.

Frahm, J. and Diekmann, S. (1979) Numerical calculation of diffuse double layer properties for spherical colloidal particles by means of a modified non-linearized Poisson-Boltzmann equation. *Journal of Colloid and Interface Science*, **70**, 440–447.

Gouy, G. (1910) Sur la constitution de la charge électrique à la surface d'un électrolyte. *Journal de Physique*, *Théorique et Appliquée*, **4**, 457–468 (in French).

Hang, P.T. and Brindley, G.W. (1970) Methylene blue absorption by clay minerals. Determination of surface areas and cation exchange capacities. *Clays and Clay Minerals*, **18**, 203–212.

Low, P.F. (1980) The swelling of clay, II Montmorillonites. *Journal of the Soil Science Society of America*, **4**, 667–676.

Marcial, D., Delage, P., and Cui, Y.J. (2002) On the high stress compression of bentonites. *Canadian Geotechnical Journal*, **39**, 812–820.

McBride, M.B. (1997) A critique of diffuse double layer models applied to colloid and surface chemistry. *Clays and Clay Minerals*, **45**, 598–608.

Mesri, G. and Olsen, R.E. (1971) Consolidation characteristics of montmorillonite. *Géotechnique*, **21**, 341–352.

Mitchell, J.K. (1993) *Fundamentals of Soil Behavior*, 2nd edition. John Wiley and Sons, New York.

Oikawa, H. (1987) Compression curve of soft soils. *Soils and Foundations Journal*, **27**, 99–104.

Pusch, R. and Yong, R. (2006) *Microstructure of Smectite Clays and Engineering Performance*. Taylor and Francis, New York.

Robinson, G.R. and Allam, M.M. (1998) Effect of clay mineralogy on coefficient of consolidation. *Clays and Clay Minerals*, **46**, 596–600.

Santamarina, J.C., Klein K.A., Wang, Y.H., and Prencke, E. (2002) Specific surface: determination and relevance. *Canadian Geotechnical Journal*, **39**, 233–241.

Sellin, P. and Leupin, O.X. (2014) The use of clay as an engineered barrier in radioactive-waste management – a review. *Clays and Clay Minerals*, **61**, 477–498.

Sridharan, A. (2014) Soil clay mineralogy and physico-chemical mechanisms governing the fine-grained soil behaviour. *Indian Geotechnical Journal*, **44**, 371–399.

Sridharan, A. and Choudhury, D. (2002) Swelling pressure of sodium montmorillonites. *Géotechnique*, **52**, 459–462.

Sridharan, A. and Choudhury, D. (2008) Computation of hydraulic conductivity of montmorillonitic clays by diffuse double layer theory. *International Journal of Geotechnical Engineering*, **2**, 1–10.

Sridharan, A. and Jayadeva, M.S. (1982) Double layer theory and compressibility of clays. *Géotechnique*, **32**, 133–144.

Sridharan, A. and Rao, G.V. (1973) Mechanisms controlling volume change of saturated clays and the role of the effective stress concept. *Géotechnique*, **23**, 359–382.

Sridharan, A. and Satyamurty, P.V. (1996) Potential-distance relationships of clay-water systems considering the Stern theory. *Clays and Clay Minerals*, **44**, 479–484.

Sridharan, A., Rao, S.M., and Murthy, N.S. (1986) Compressibility behavior of homoionized bentonites. *Géotechnique*, **36**, 551–564.

Tripathy, S. and Schanz, T. (2007) Compressibility behaviour of clays at large pressures. *Canadian Geotechnical Journal*, **44**, 355–362.

Tripathy, S., Sridharan, A., and Schanz, T. (2004) Swelling pressures of compacted bentonites from diffuse double layer theory. *Canadian Geotechnical Journal*, **41**,437–450.

Tripathy, S., Bag, R., and Thomas, H.R. (2014) Effect of Stern-layer on the compressibility behaviour of bentonites. *Acta Geotechnica*, **9**, 1–13.

van Olphen, H. (1963) *An Introduction to Clay Colloid Chemistry*. Interscience, New York.

Verwey, E.J.W. and Overbeek, J.T.G. (1948) *Theory of the Stability of Lyophobic Colloids*. Elsevier, Amsterdam.

Yong, R., Taylor, L.O., and Warkentin, B.P. (1962) Swelling pressures of sodium montmorillonite at depressed temperatures. *Clays and Clay Minerals*, **11**, 268–281.

Yong, R.N. and Mohamed, A.M.O. (1992) A study of particle interaction energies in wetting of unsaturated expansive clays. *Canadian Geotechnical Journal*, **29**, 1060–1070.

Zheng, L., Samper, J., and Montenegro, L. (2011) A coupled THC model of the FEBEX in situ test with bentonite swelling and chemical and thermal osmosis. *Journal of Contaminant Hydrology*, **126**, 45–60.

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APPENDIX

Theoretical e vs. P relationship

The clay surface properties were assumed to be CEC = 100 meq/100 g and SSA = 800 m^2/g; pore-fluid parameters: $\nu = 1$, $c_0 = 10^{-4}$ M (*i.e.* the molar concentration which is equal to 10^{-4} N for monovalent electrolytes), $T = 298$ K, and $\varepsilon = 78.54$. The specific gravity of the clay was assumed to be 2.76. The procedure for computing the void ratio for a given consolidation pressure of 100 kPa was given in the following steps:

1. Scaled mid-plane potential (equation 1)

$$y_d = \text{arccosh}(1 + P/2c_0RT) = 6$$

2. Scaled surface potential, y_0, was computed by equating the value of $(dy/d\xi)_{x=0}$ in equations 1–2. Using equation 2,

$$\left(\frac{dy}{d\xi}\right)_{x=0} = -\frac{CEC}{SSA}\left(\frac{2513.6}{\sqrt{\varepsilon c_0 T}}\right) = -205.38.$$

The surface potential from equation 3 was

$$y_0 = \text{arccosh}(0.5\{[(dy/d\xi)_{x=0}]^2 + 2\cosh(y_d)\}) = 10.66$$

3. Scaled half-space distance was computed by the following integration

$$\kappa d = -\int_{y_0}^{y_d+\delta} (2\cosh(y) - 2\cosh(y_d))^{-1/2}$$

where δ is an infinitesimally small number (*i.e.* 10^{-5}) for numerical stability of the elliptic integral. The scaled distance

$$\kappa d = -\int_{10.66}^{6+\delta} (2\cosh(y) - 2\cosh(6))^{-1/2} dy = 0.146$$

4. The pore-fluid parameter (inverse of the 'Debye' length) was

$$\kappa = \sqrt{\frac{8\pi e'^2 \nu^2 c_0}{\varepsilon k_B T}} = 0.33 \times 10^6 \text{cm}^{-1} = 0.0033 \text{ Å}^{-1}$$

where c_0 = (normality $\times 10^{-3} \times$ Avogadro's number) ions/cm^3 (Van Olphen, 1963), $e' = 4.8 \times 10^{-10}$ esu, and $k_B T = 0.4 \times 10^{-13}$ ergs

5. The half-space distance was, therefore, $d = \kappa d/\kappa$ = 44.42 Å

6. Void ratio, therefore, was $e = d \times \rho_w \times G \times SSA = 9.81$

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