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Determination of Interplate Distance between Two Clays and Initial Bulk Concentrations: An Inverse Problem

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***Abstract* – The space between two clay particles consists of two electric double layers, one adjacent to each clay surface and in-between effective pore space. While solving the Poisson-Boltzman equation, one boundary condition is that, at infinite distance from the clay surface potential is zero. Practically midpoint of the interplate distance is considered to be at infinite distance from the clay surface. Without knowing the interplate distance, the Poisson-Boltzman equation can not be solved for computing the electric double layer. In this paper, a method is suggested for computation of effective pore space as well as electric double layer and consequently the interplate distance. The existing methods of bulk concentration measurement suit the evaluation of electric double layer and effective pore space in saturated soil. However, applying mass balance for individual ion, the bulk concentration and the distribution of ions in unsaturated state are predicted accurately treating it as an inverse problem.**

***Keywords* – Clay Surface, Electric Double Layer, Interplate Distance, Pore.**

I. INTRODUCTION

Sodic soils are more difficult to reclaim than the saline soil because the sodic soils require chemical amendments for replacing the exchangeable sodium with calcium for improving soil permeability as well as leaching [1]-[4]. Before applying the required quantity of gypsum to a sodic soil for reclamation, it is necessary to know the concentration of sodium and calcium ions present in the soil water. The general practice is to prepare a saturation extract to ascertain the concentrations of ions in the bulk solution [5]. The *in situ* soil moisture will be mostly at unsaturated state. By saturating the soil for getting the saturation extract, the bulk concentrations of sodium and calcium ions in the soil water in the unsaturated soil get modified. Hardly any direct method exists to evaluate the *in situ* concentrations of ions in the bulk solution when the soil is in an unsaturated state. To ascertain the initial bulk concentrations when the soil is in unsaturated state, the requirement is prior knowledge of the distance between the two clay plates.

In case of the clay minerals, the lower valence cations isomorphically substitute the higher valence cations that take place during the formation of clay minerals [6]-[8]. The deficit charges thus formed are compensated by adsorption of the cations (e.g., Ca^{2+}, Mg^{2+}, K^{+}, Na^{+} etc.) on the exterior surface [9],[10]. The electrostatic forces that exist between the charges on the clay surface and the cations present in the soil water form the electric double layer (EDL) at the clay surface [11]. The thickness of electrical double layer is an important controlling factor for the structural development, hydraulic conductivity, and other physico-chemical and mechanical properties of soil [12]-[17]. The electric double layer thickness is governed by the concentrations of salts and types of cations in the soil water [18]-[21]. From the measured bulk concentrations of the saturation extract, the electric double layer (EDL) thickness is obtained [22]-[26]. The effective pore space between two clay particles in dispersed state is determined from the measured hydraulic conductivity of a saturated sodic soil sample in a laboratory [27]. In case of strongly dispersed sediments as in sodic soil, the particles are oriented in a parallel way [28]-[31] reducing the infiltration rate as well as hydraulic conductivity. Figure 1 illustrates two clay plates in a dispersed sediment structures. Thus, the two electric double layers adjacent to the clay surfaces and the effective pore space constitute the interplate distance [32]. In this paper, a method is proposed to compute the interplate distance taking into account the electric double layers and evaluate the bulk concentrations of sodium and calcium ions when the soil is in an unsaturated state.

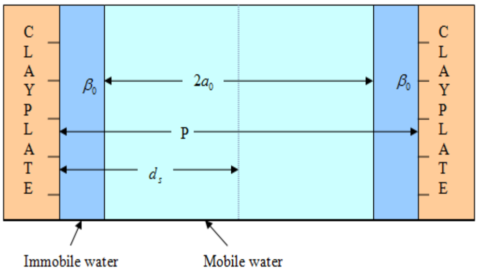

Fig.1. Mobile and immobile soil moisture zone between two clay plates in a dispersed system

II. MATERIALS AND METHODS

Determination of interplate distance (P)

Referring to Fig. (1), the interplate distance is given by,

$$P = 2a_0 + 2\beta_0 = 2d_s \tag{1}$$

a) Determination of a_0

The soil becomes saturated when the whole pore space (P) is filled with water as shown in Fig. 1. At that time, the distance between the two electric double layers of clay plates is $2a_0$ and the thickness of each double layer is β_0. The saturated hydraulic conductivity $\left(\tilde{K}\right)$ of the soil can be measured by a falling head permeameter in laboratory. Knowing $\tilde{K}$, a_0 is estimated from the relation[33],[34]:

$$a_0 = \sqrt{\frac{3\nu\tilde{K}}{g}} \tag{2}$$

where $\nu = \dfrac{\mu}{\rho_b}$ = kinematic viscosity of water ($m^2 s^{-1}$); g = acceleration due to gravity (ms^{-2}); μ = viscosity ($Ns\ m^{-2}$). The kinematic viscosity of water $\nu = 0.9025 \times 24 \times 60 \times 60 \times 10^{-6}$ $m^2\ day^{-1}$ at 25 ^{0}C.

b) Determination of β_0

The thickness of the double layer when both sodium and calcium ions are present in soil water in the saturation extract is given by [35]:

$$\beta_0 = \sqrt{\frac{\varepsilon RT}{2F^2\left\{Z_1{}^2 C_{bNa^+} + Z_2{}^2 C_{bCa^{2+}}\right\}}}$$

Thus, knowing a_0 and β_0, P is known from Eq. (1).

c) Estimation of d_s

The distance d_s is the half of the interplate distance. Hence, $d_s = \dfrac{P}{2}$ (3)

This space is filled with soil water when the soil is saturated.

d) Determination of d_f

Let, the thickness of each clay plate is d_c. Consider a parallelepiped of one unit cross section area (1 nanometer square) from middle of one clay plate to the other as shown in the Fig. 2.

Volumetric moisture content at saturation,

$$\theta_s = \frac{\text{volume of water in saturated soil}}{\text{total volume of soil}}$$

$$= \frac{\text{volume of water in saturated soil}}{\text{volume of soil + volume of void}}$$

$$= \frac{2d_s\ nm \times 1\ nm^2}{2 \times \frac{1}{2} d_c\ nm \times 1\ nm^2 + 2d_s\ nm \times 1\ nm^2}$$

$$= \frac{2d_s}{2d_s + d_c} \qquad (4)$$

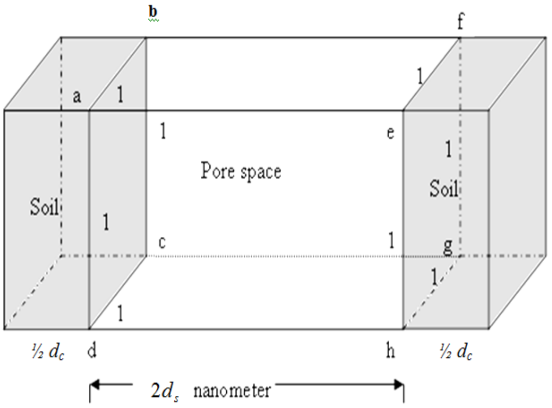

Fig.2. A parallelepiped with unit cross section area ranging from middle of one clay plate to another

When θ_s is known, d_c can be determined from Eq. (4) by, $d_c = \dfrac{2d_s}{\theta_s} - 2d_s$

Volumetric moisture content at less than saturation (θ_f) is given by:

$$\theta_f = \frac{\text{volume of water when soil moisture is at field capacity}}{\text{total volume of soil}}$$

$$= \frac{2d_f\ nm \times 1\ nm^2}{2 \times \frac{1}{2} d_c\ nm \times 1\ nm^2 + 2d_s\ nm \times 1\ nm^2}$$

$$= \frac{2d_f}{2d_s + d_c} \qquad (5)$$

Dividing Eq. (5) by Eq. (4),

$$\frac{\theta_f}{\theta_s} = \frac{\dfrac{2d_f}{2d_s + d_c}}{\dfrac{2d_s}{2d_s + d_c}} = \frac{d_f}{d_s} \qquad (6)$$

Hence, as shown in Fig. (3),

$$d_f = \frac{\theta_f}{\theta_s} \times d_s \qquad (7)$$

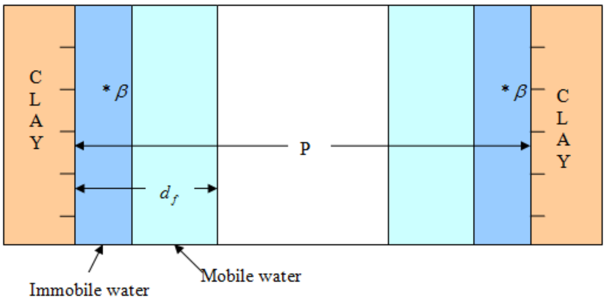

Fig.3. Schematic representation of the mobile and immobile water when soil is unsaturated

Solution of the Inverse Problem

The inverse problem is solved through mass balance. As distilled water is added to the unsaturated soil sample while preparing the saturation extract, there is no change in total concentrations of ions. The mass balance equations for Na^+ and Ca^{2+} ions can be written as:

$$\int_0^{d_s} C_{bNa^+} e^{\frac{-FZ_1\varphi(x)}{RT}} dx - \int_0^{d_f} C^*_{bNa^+} e^{\frac{-FZ_1\varphi^*(x)}{RT}} dx = 0 \qquad (8)$$

$$\int_0^{d_s} C_{bCa^{2+}} e^{\frac{-FZ_2\varphi(x)}{RT}} dx - \int_0^{d_f} C^*_{bCa^{2+}} e^{\frac{-FZ_2\varphi^*(x)}{RT}} dx = 0 \qquad (9)$$

where $\int_0^{d_s} C_{bNa^+} e^{\frac{-FZ_1\varphi(x)}{RT}} dx$ = total number of Na$^+$ ions in the saturation extract, C_{bNa^+} = bulk concentration of

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Na^+ ions in saturation extract which is measured, hence, known; $\int_0^{d_f} C^*_{bNa^+} e^{\frac{-FZ_1\varphi^*(x)}{RT}} dx =$ total number of Na$^+$ ions and $C^*_{bNa^+}$ = bulk concentration of Na$^+$ ions when the soil is unsaturated, both unknown a priori; $\int_0^{d_s} C_{bCa^{2+}} e^{\frac{-FZ_2\varphi(x)}{RT}} dx =$ total number of Ca^{2+} ions in the saturation extract, $C_{bCa^{2+}} =$ bulk concentration of Ca^{2+} ions in the saturation extract which is measured, hence, known; $\int_0^{d_f} C^*_{bCa^{2+}} e^{\frac{-FZ_2\varphi^*(x)}{RT}} dx =$ total number of Ca^{2+} ions , $C^*_{bCa^{2+}}$ = bulk concentration of Ca^{2+} ions when the soil moisture is at field capacity both unknown a priori; $\varphi(x)$ has been obtained using the linearized solution of Poisson-Boltzman equation, $\varphi(x)=\varphi(0)e^{-\lambda x}$;

$$\kappa = F\sqrt{\frac{2}{\varepsilon RT}\left\{Z_1^{\,2} C_{bNa^+} + Z_2^{\,2} C_{bCa^{2+}}\right\}}\,;\varphi(0)=\frac{\sigma}{\varepsilon\kappa}\,;$$

$\sigma =$ surface charge density; $\sigma = -\frac{CEC}{S}$; CEC = cation exchange capacity which measured, S = specific surface which is also known.

$\varphi^*(x)$ = potential when the soil is unsaturated (V) which can be expressed in terms of unknown bulk concentrations, $C^*_{bNa^+}$ and $C^*_{bCa^{2+}}$ as,

$$\varphi^*(x)=\varphi^*(0)e^{-\kappa^* x};$$

$$\kappa^* = F\sqrt{\frac{2}{\varepsilon RT}\left\{Z_1^{\,2} C^*_{bNa^+} + Z_2^{\,2} C^*_{bCa^{2+}}\right\}};$$

$$\varphi^*(0)=\frac{\sigma}{\varepsilon\kappa^*}.$$

The two unknowns $C^*_{bNa^+}$ and $C^*_{bCa^{2+}}$ are solved by Taylor series expansion. Let,

$$f_1 = \int_0^{d_s} C_{bNa^+} e^{\frac{-FZ_1\varphi(x)}{RT}} dx - \int_0^{d_f} C^*_{bNa^+} e^{\frac{-FZ_1\varphi^*(x)}{RT}} dx \tag{10}$$

$$f_2 = \int_0^{d_s} C_{bCa^{2+}} e^{\frac{-FZ_2\varphi(x)}{RT}} dx - \int_0^{d_f} C^*_{bCa^{2+}} e^{\frac{-FZ_2\varphi^*(x)}{RT}} dx \tag{11}$$

Let, $*C_{bNa^+}$ and $*C_{bCa^{2+}}$ be in the vicinity of zeroes of f_1 and f_2. Applying Taylor series expansion about the zeroes

$$f_1\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} + \frac{\partial f_1}{\partial C^\gamma_{Na^+}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Na^+} + \frac{\partial f_1}{\partial C^\gamma_{Ca^{2+}}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Ca^{2+}} = 0 \tag{12}$$

$$f_2\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} + \frac{\partial f_2}{\partial C^\gamma_{Na^+}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Na^+} + \frac{\partial f_2}{\partial C^\gamma_{Ca^{2+}}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Ca^{2+}} = 0 \tag{13}$$

$$\frac{\partial f_1}{\partial C^\gamma_{Na^+}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Na^+} + \frac{\partial f_1}{\partial C^\gamma_{Ca^{2+}}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Ca^{2+}} = -f_1\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \tag{14}$$

$$\frac{\partial f_2}{\partial C^\gamma_{Na^+}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Na^+} + \frac{\partial f_2}{\partial C^\gamma_{Ca^{2+}}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \Delta C_{Ca^{2+}} = -f_2\Big|_{*C_{bNa^+},*C_{bCa^{2+}}} \tag{15}$$

The Eq. (14) and (15) in matrix notation can be expressed as:

$$\begin{bmatrix} A(1,1) & A(1,2) \\ A(2,1) & A(2,2) \end{bmatrix}\begin{bmatrix} B(1) \\ B(2) \end{bmatrix} = \begin{bmatrix} C(1) \\ C(2) \end{bmatrix} \tag{16}$$

where the matrix elements are:

A (1, 1) = $\frac{\partial f_1}{\partial C^\gamma_{Na^+}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}}$;

A (1, 2) = $\frac{\partial f_1}{\partial C^\gamma_{Ca^{2+}}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}}$;

A (2, 1) = $\frac{\partial f_2}{\partial C^\gamma_{Na^+}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}}$;

A (2, 2) = $\frac{\partial f_2}{\partial C^\gamma_{Ca^{2+}}}\Big|_{*C_{bNa^+},*C_{bCa^{2+}}}$;

B (1) = ΔC_{Na^+} ; B (2) = $\Delta C_{Ca^{2+}}$;

C (1) = $-f_1\Big|_{*C_{bNa^+},*C_{bCa^{2+}}}$; and C (2) = $-f_2\Big|_{*C_{bNa^+},*C_{bCa^{2+}}}$.

Applying matrix inversion

$$\begin{bmatrix} B(1) \\ B(2) \end{bmatrix} = \begin{bmatrix} A(1,1) & A(1,2) \\ A(2,1) & A(2,2) \end{bmatrix}^{-1}\begin{bmatrix} C(1) \\ C(2) \end{bmatrix}$$

The improved values of $*C_{bNa^+}$ and $*C_{bCa^{2+}}$ are $*C_{bNa^+} + \Delta C_{Na^+}$ and $*C_{bCa^{2+}} + \Delta C_{Ca^{2+}}$ respectively.

The procedure is repeated until f_1 and f_2 attains desired accuracy.

III. RESULTS AND DISCUSSION

The bulk concentrations in the soil water when the soil is in an unsaturated state is determined making use of the bulk concentrations of the saturation extract for fine loamy and sandy loam soils. For fine loamy soil the adopted data [36] are, $\theta_s = 0.47$ and $\theta_f = 0.129$ and $\widetilde{K} = 0.0184$ m day^{-1}. The computed value of surface charge density for fine loamy soil is, $\sigma = -0.058$ Cm^{-2} [25]. Bulk concentrations of ions in the saturation extract (assumed) are, $C_{bNa^+} = 20$ mol m^{-3} and $C_{bCa^{2+}} = 10$ mol m^{-3}, and corresponding $\kappa = 8.05\times10^{8}$ m^{-1}, hence, $\beta_0 = 1.24$ nm. Corresponding to $\widetilde{K} = 0.0184$ m day^{-1}, $a_0 = 242.6$ nm. Therefore, $d_s = 243.84$ nm and $d_f = 66.8$ nm. When the soil is saturated for the saturation extract, in that saturated state, $\varphi(0) = -0.103$ V. The bulk concentrations of Na$^+$ and Ca^{2+} ions when the soil is at unsaturated state, corresponding to the assumed bulk concentration in the saturation extract are found to be $C_{bNa^+} = 27.17$ mol m^{-3} and $C_{bCa^{2+}} = 0.427$ mol m^{-3}, which have been obtained from Eq. (12) and Eq. (13), starting from an initial guess for $*C_{bNa^+}$ and $*C_{bCa^{2+}}$. The history of convergence is presented in Table 1.

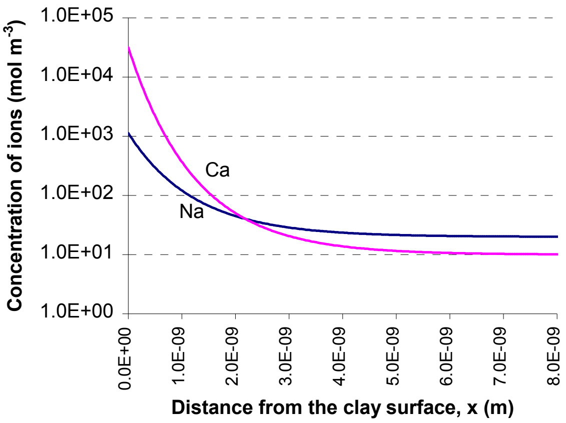
Table 1: History of convergence of $f_1|_{*C_{bNa^+},*C_{bCa^{2+}}}$; $f_2|_{*C_{bNa^+},*C_{bCa^{2+}}}$; ΔC_{Na^+} and $\Delta C_{Ca^{2+}}$ corresponding to $C_{bNa^+} = 20$ mol m^{-3} and $C_{bCa^{2+}} = 10$ mol m^{-3} and $\sigma = -0.058$ Cm^{-2} initial guess for $*C_{bNa^+} = 2$ mol m^{-3}, and for $*C_{bCa^{2+}} = 1$ mol m^{-3}

Iteration number	$f_1\|_{*C_{bNa^+},*C_{bCa^{2+}}}$	$f_2\|_{*C_{bNa^+},*C_{bCa^{2+}}}$	ΔC_{Na^+}	$\Delta C_{Ca^{2+}}$	$*C_{bNa^+}$	$*C_{bCa^{2+}}$
1	-2.29E-04	-1.94E+01	-0.64708	0.31697	1.35292	1.31697
2	-8.11E-05	-7.51E+00	-0.45647	0.27974	0.89645	1.59671
3	-2.74E-05	-2.95E+00	-0.24003	0.24147	0.65642	1.83818
4	-8.80E-06	-1.16E+00	-0.04695	0.21477	0.60947	2.05296
5	-2.53E-06	-4.59E-01	0.13227	0.19731	0.74174	2.25027
6	-4.80E-07	-1.82E-01	0.33119	0.18082	1.07293	2.43109
7	1.71E-07	-7.23E-02	0.59070	0.15526	1.66364	2.58636
8	3.56E-07	-2.88E-02	0.95372	0.10985	2.61736	2.69621
9	3.82E-07	-1.15E-02	1.45816	0.03438	4.07553	2.73059
10	3.49E-07	-4.60E-03	2.11931	-0.07690	6.19484	2.65369
11	2.89E-07	-1.83E-03	2.89866	-0.21834	9.09351	2.43535
12	2.15E-07	-7.21E-04	3.66989	-0.36547	12.76340	2.06987
13	1.38E-07	-2.79E-04	4.20827	-0.47465	16.97167	1.59522
14	6.90E-08	-1.03E-04	4.22655	-0.49627	21.19823	1.09894
15	2.00E-08	-3.50E-05	3.46742	-0.40064	24.66566	0.69831
16	-2.58E-09	-9.79E-06	1.95920	-0.21481	26.62486	0.48349
17	-3.66E-09	-1.70E-06	0.515988	-0.053068	27.14085	0.43042
18	-3.73E-10	-8.84E-08	0.030136	-0.00294	27.17098	0.42748
19	1.29E-11	-1.54E-09	0.000097	-0.000009	27.17108[a]	0.42747[b]

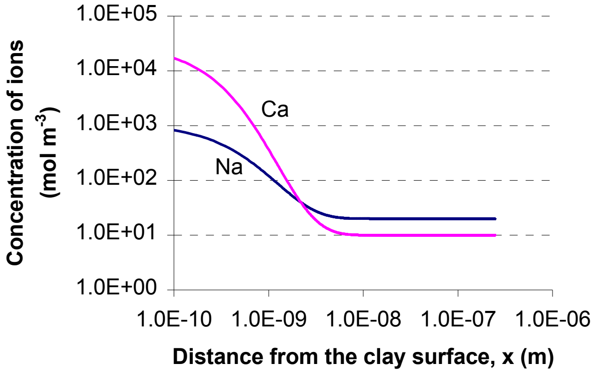
[a] *Final* $*C_{bNa^+} = 27.17108$ *mol* m^{-3}; [b] *Final* $*C_{bCa^{2+}} = 0.42747$ *mol* m^{-3}

The bulk concentration in the soil moisture when the soil is in unsaturated state can be obtained starting from any initial guess for $*C_{bNa^+}$ and $*C_{bCa^{2+}}$.

The variation of concentrations of Na$^+$ and Ca^{2+} ions with distance from the clay surface up to $x = d_s$ when the soil is saturated for sampling saturation extract are presented in Fig. 4 (a) and (b). In Fig. 4 (a), the variation near the clay surface is plotted in semi-log scale and in Fig. 4 (b), the variation of concentrations at large distance is plotted in log-log scale.

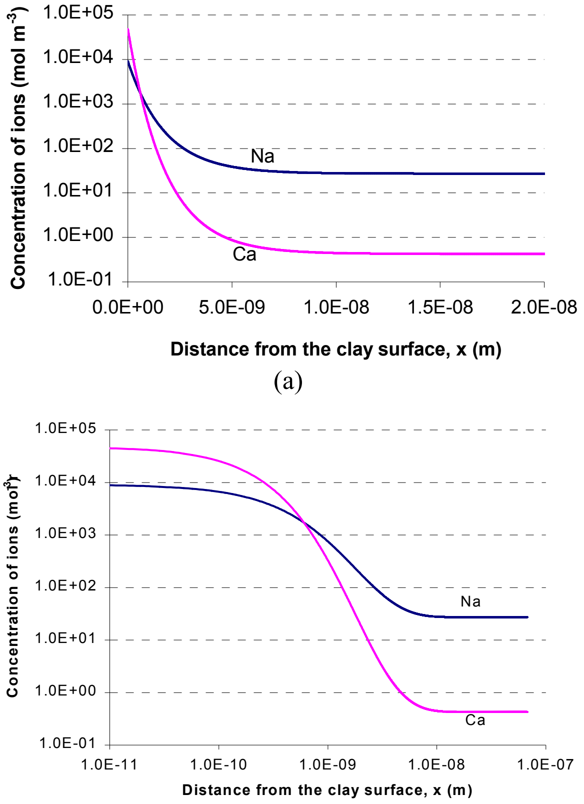


(a)



(b)

Fig.4. (a) (b) The distributions of concentrations of cations between two clay plates when the soil get saturated while sampling saturation extract, C_{bNa^+} =20 mol m^{-3} and $C_{bCa^{2+}}$ =10 mol m^{-3} and σ = -0.058 Cm^{-2}; half of the distance between the clay plates = 243.84×10^{-9} m



(a)

(b)

Fig.5. (a) (b) The distributions of concentrations of cations from the clay surface when the soil is unsaturated, C_{bNa^+} solved as inverse problem 27.17 mol m^{-3} and $C_{bCa^{2+}}$ = 0.427 mol m^{-3} and σ = -0.058 Cm^{-2} ; width of the soil water zone= 6.68×10^{-8} m

As seen from the figure Fig. 5 (a) and (b), due to saturation for sampling the saturation extract, bulk concentration of Na$^+$ ions has decreased from 27.17 mol m^{-3} to 20 mol m^{-3}; the concentration of Na$^+$ ions near the clay surface has decreased from 9115.05 mol m^{-3} to 1130.6 mol m^{-3}. The total Na$^+$ ions in the parallelepiped of length $d_f = \int_0^{d_f} C^*_{bNa^+} e^{\frac{-FZ_1\varphi^*(x)}{RT}} dx = 5.324393 \times 10^{-6}$ mol, which has been obtained solving the inverse problem with accuracy 2.64×10^{-16}. The total Na$^+$ ions in the parallelepiped of length $d_s = \int_0^{d_s} C_{bNa^+} e^{\frac{-FZ_1\varphi(x)}{RT}} dx$ $= 5.324393 \times 10^{-6}$ mol. Thus, the mass balance has been satisfied with an accuracy of 2.64×10^{-16} in respect of total Na$^+$ ions.

In case of Ca^{2+} ions, due to saturation while sampling the saturation extract, bulk concentration of Ca^{2+} ions has increased from 0.427 mol m^{-3} to 10 mol m^{-3}, thus the bulk concentration has increased considerably on saturation. Therefore, the concentration of Ca^{2+} ions near the clay surface has decreased from 48107.9 mol m^{-3} to 31956.8 mol m^{-3}. The total Ca^{2+} ions in the parallelepiped of length $d_f = \int_0^{d_f} C^*_{bCa^{2+}} e^{\frac{-FZ_2\varphi^*(x)}{RT}} dx = 8.223899 \times 10^{-6}$ mol, which has been obtained solving the inverse problem with accuracy 1×10^{-14}. The total Ca^{2+} ions in the parallelepiped of length d_s = $\int_0^{d_s} C_{bCa^{2+}} e^{\frac{-FZ_2\varphi(x)}{RT}} dx = 8.223899 \times 10^{-6}$ mol. Thus, the mass balance has been satisfied with an accuracy of 1×10^{-14} in respect of total Ca^{2+} ions. Therefore, prediction of the bulk concentration when the soil was in unsaturated state is accurate.

IV. CONCLUSION

With the measured bulk concentrations, C_{bNa^+} = 20 mol m^{-3} and $C_{bCa^{2+}}$ = 10 mol m^{-3}, and hydraulic conductivity, the interplate distance of fine loamy soil was computed to be 487.68 nm. Subsequently, performing mass balance for individual ions, the bulk concentrations in the soil water when the soil is in unsaturated state are predicted accurately, treating it as an inverse problem. From the computed bulk concentrations, the spatial distributions of ions inside a soil pore can be illustrated. Thus, once the interplate distance between two clays is determined, the distribution of concentration of ions can be illustrated for both saturated and unsaturated conditions. The known interplate distance is also helpful to carry out simulation study of interaction among different ions inside the pore.

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