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Review Of Two Electrokinetic Phenomena Named Electro-osmosis And Streaming Potential And Introduction Of Two Novel Electrokinetic Phenomena Named Field-Effect-Electroosmosis And Field-Effect-Streaming Potential.

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ABSTRACT

Two novel electrokinetic effects are proposed and studied theoretically. To understand these effects a review is done on the double layer at the insulator-electrolyte interface. These effects, Field-Effect-Electro-osmos is and Field-Effect-Streaming Potential are arising from manipulation of double layer charge at the interface of insulator-electrolyte. further details are coming in this manuscript and the references.

Keywords: Electrokinetic phenomena, Double layer, Field-Effect-Electro-osmosis, Field-Effect-Streaming potential, Zeta potential, Electro-osmosis, Streaming potential.

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INTRODUCTION

Background: Electrokinetic Phenomena and Double Layer

Electro kinetic phenomena were discovered quite early in the nineteenth century. Hence investigations in the field have been conducted for more than a century and a half. The discovery of electro osmosis and electrophoresis by Re us occurred soon after the first investigations on the electrolysis of water by Nicholson or Carlisle [1] and the electrolysis of salt solutions by Berzelius (1804) and Davy (1807). Reus [2] carried out two experiments, the first demonstrated the effect known as electro osmosis, and the second was actually the discovery of electrophoresis. Quincke [3] was the first person to show that there should exist a phenomenon opposite to electro osmosis. If electro osmosis is the movement of liquid in a porous medium arising under the action of an external electric filed, the opposite effect will be the appearance of a potential difference under the influence of the flow of liquid through a porous diaphragm, or under the influence of a pressure drop set up externally.

The universal character and diversity of electro kinetic phenomena, the formulation of more distinct notions of the electrical structure(double layer) of the interfacial layer, the discovery of electro kinetic phenomena in the simplest system (the single capillary) the growing understanding of the importance of discovering the mechanism of electro kinetic phenomena as a source of information about the double layer and surface chemistry all these created the necessary ground-work for the elaboration of the theory as early as the end of the nineteenth century. Such a theory of a formal nature, independent of the mechanism of forming the electric double layer, was first advanced by Helmhotz [4]. Considering the simplest case of electro osmosis in a single capillary, Helmholtz obtained a formula for the linear velocity of electro osmosis:

$$v_{eo} = -\frac{\varepsilon \xi}{4\pi\eta} E \qquad (1)$$

Where ξ is the interfacial electric potential difference, E is the electric field strength , and η is the viscosity of the liquid. Helmholtz also obtained a formula for the streaming potential, $\Delta V_{st\gamma}$, arising in the capillary under the effect of a pressure P, namely

$$\frac{\Delta V_{st\gamma}}{P} = \frac{\varepsilon \xi}{4\pi\eta\kappa} \qquad (2)$$

Where κ is the specific conductivity of the liquid. Although equations 1 and 2 were derived for a single capillary, Smoluchowski [5] was able to extend the theory of electroosmosis and the streaming potential to the case of a porous plug.

Smoluchowki's theory was restricted by the condition that the thickness of the double layer be small in comparison with the width of the capillaries of the porous plug. With a similar assumption that the double-layer thickness is small in comparison with the linear dimensions of the particle, Smoluchowski derived a formula for the velocity of electrophoresis of a particle of arbitrary shape :

$$v_{ef} = -\frac{\varepsilon \xi}{4\pi\eta} E \qquad (3)$$

It should be noted that equations 1 and 3 differ only in sign. If the interfacial electric potential difference is negative in both cases, the direction of electro omosis will be directed along the field , since it results from the action of the field on a positive (in the given case) charge of the liquid. The direction of electrophoresis will be the opposite, since the determining factor is the effect of the particle. As a result of the elaboration of the theory and, in particular, because of Saxen's experiments [6], entirely new premises appeared for experimental research. Not only did investigations of electro kinetic phenomena become possible at this stage, but studies of the double layer on the basis of the electro kinetic phenomena were also conducted.
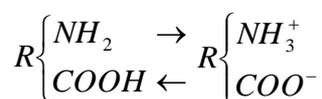
The effect of the chemical nature of the surface and the ionic composition of electrolyte on the sign and magnitude of the potential (determined by the electro kinetic measurement) was determined experimentally in the early parts of this century, and furnished the grounds for solving the problem of the mechanism of the formation of the double layer of colloid particles.

Freundlich [7] called attention to a possible connection between the appearance of the double layer and an adsorption phenomenon.

It was considered that, if the adsorption coefficients of the ions were different, the strongly adsorbed ions would be present in excess on the surface, and the weakly adsorbed ions would be present in excess in the liquid part of the double layer, together giving rise to the double layer.

Another possible mechanism for the formation of the double layer is linked with the dissociation of surface ionogenic groups under the influence of a polar dispersion medium. This mechanism was first studied in regard to proteins. The ionogenic groups in proteins are of different chemical natures (acidic carboxyl, basic amino groups, etc.), and proteins are classified as amphoteric electrolytes.

In a first approximation , the amphoteric nature of monomer units of the protein molecule may be characterized by the following model:

$$R\begin{cases} NH_2 \\ COOH \end{cases} \begin{array}{c} \rightarrow \\ \leftarrow \end{array} R\begin{cases} NH_3^+ \\ COO^- \end{cases}$$

At low pH, the protein carries a + charge. As ph is increased, the isoelectric point is first reached, and then there is a charge in sign of the charge on the protein.

Chemical groups on the insulator surface, at an interface between a liquid and an insulating solid, dissociate similarly to the above mechanism. Due to this surface ionization and specific adsorption , the interface is charged and ions of opposite polarity to the interfacial charge(counterions) are attracted to it, while ions of the same polarity are repelled.

An early theory due to Helmholtz [4] assumed that the counterions were attracted to the charged surface at a distance of about one molecular diameter. The potential then falls to zero rapidly as shown in Fig. 1(a), and the double layer can be treated as a paralled plate capacitor. This can be a first approximation , because thermal agitation must tend to diffuse the ions held in the liquid. This Helmholtz model was superseded by the Gouy-Chapman model [8,9], in which the counterions were assumed to form a diffuse layer, with the concertrations and the potential falling off rapidly with distance at first , and then more gradually as shown in Fig.1(b).

This model was useful for plane surfaces with a low charge density and for distances not too close to the surface, but was inadequate for small distances and high charges because it neglected the diameters of the ions which were treated as point charges. His theory was therefore modified by Stern[10], he assumed the liquid side of the double layer of couterions strongly held close to bound surface charge and a more diffuse layer of the some polarity. According to Stern, the potential falls off rapidly and linearly at first and then more gradually. The bound counterions in the Stern layer can even change the sign of the potential, as shown in Fig1(d).
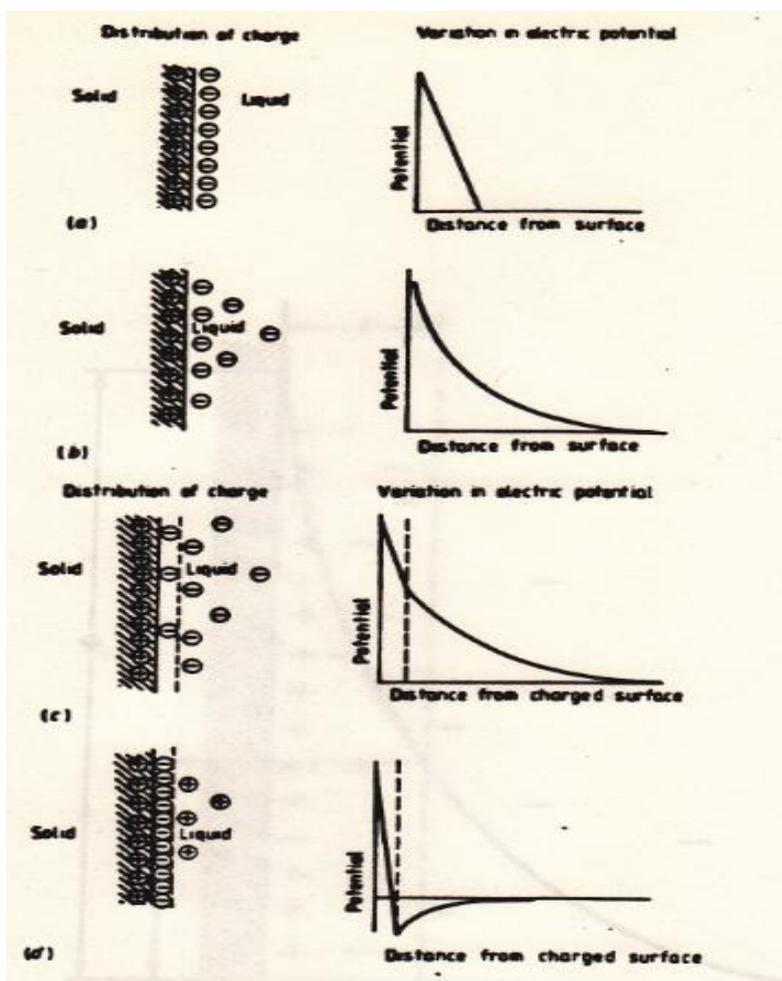
Figure 1: Models of the charged double layer at a solid-liquid interface.

a) **Helmholtz model**
b) **Gouy-Chapman model**
c) **Stern model**
d) **Stern model showing reversal of sign of charge surface**

The distribution of ions in the diffuse part of the layer is controlled by thermal diffusion and is therefore governed by the Boltzmann equation, i.e. for ions with the same polarity as the interface.

$$n_+ = n_0 \exp\left(\frac{zq\psi_x}{kT}\right) \qquad (4)$$

Here n_+ is the concentration of ions, Z is the ion valency , ψ_x is the potential at distance x from the surface, k is Boltzmann's constant , T is the absolute temperature , and q is the elementary electron charge. This leads to a potential which decreases exponentially from the surface

$$\psi_x = \psi_0 \exp(-kx) \qquad (5)$$

The extent of the layer is inversely proportional to x, which is given by

$$k^2 = \frac{2q^2 n_0}{\varepsilon_r \varepsilon_0 kT} = \frac{1}{\delta_0^2} \qquad (6)$$

$l/k = d_0$ is the Debye length of the double layer, i.e. a measure of the diffuse layer thickness.

The notions developed by Gouy and Chapman mode it is possible to explain the difference between the thermodynamic ψ_0 and electro kinetic ξ potentials and to discover the cause of some experimentally established relationships of electro kinetics. Before the appearance of Smoluchowski's[11], and Freundlich's[7] papers, the ψ and ξ potentials were regarded as identical in discussions of electro kinetic phenomena. Smoluchowski and Freoundlich proposed that the thermodynamic and ξ potentials should be distinguished and that the term electro kinetic potential should be applied to only the portion of the interphase potential difference located in the part of the double layer within which the liqid was invoved in tangential motion . As can be seen from Fig.2 ,the electro kintic potential is equal to the potential of the diffuse layer in the shear plane, this being the boundary of the liquid involved in electro osmosis slipping.

Figure 2: Typical variation in potential with distance from surface with no specific interaction . δ , thickness of immobile layer, σ_1 , immobile charge, σ_2 , electrokinetic charge.
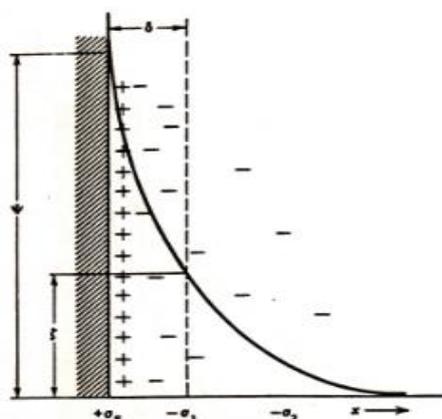
Experimental data show that the dependences of the thermodynamic and ξ potentials on the concentration of the potential-determined ions are different. The thermodynamic potential, ψ , is almost unaltered on introducing on electrolyte , whereas a decrease in double-layer thickness on introducing electrolyte should lead to a decrease in the electrokinetic potential as shown in Fig.3. with fairly high concentrations of electrolyte , the double layer thickness may possibly become less than the distance d from the shear plane to the wall, and the ξ potential may become infinitesimally small.

**Figure 3: Curves shown how the potential of the shear plane decreases with increasing electrolyte content: $c_3 > c_2 > c_1$, $\xi(c_1) > \xi(c_2) >> \xi(c_3)$,....., shear plane**
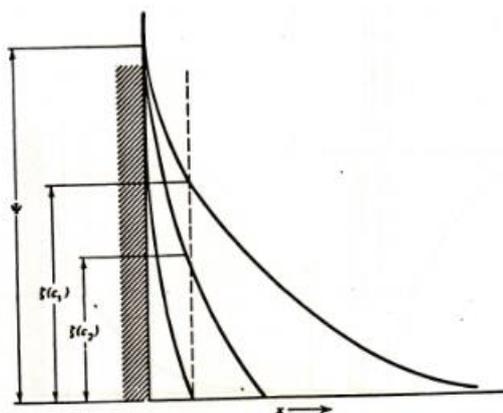
This explanation was given for the experimental data obtained on the decrease in ξ and on its eventual disappearance when a large quantity of concentrated electrolyte was introduced.

Since Stern considers that the ions cannot approach the surface to a distance less than d, the differentiates between the maximum value of the potentials in the diffuse layer ψ_d and the potential of the wall ψ_o. The ψ_d potential , introduced by Stern, is often called the stern potential (inner electric potential at the outer Stern plane). The surface charge and the single layer of adsorbed counter ions from something like a molecular condenser. As seen from the diagram in Fig.4, the surface charge σ_s is compensated for by the sum of charges of the molecular condenser σ_{st} and the outer diffuse layer σ_d , where as the total potential drop ψ_o consists of the potential drop in the diffuse region ψ_d and the potential difference between the sides of the molecular condenser $(\psi_o - \psi_d)$. It should be noted that the position of the shear plane in the model under consideration is still obscure. Some authors have assumed that it coincides with the boundary between the stern layer and the Gouy layer, at that $\psi_d = \xi$. In the general case, however, it may be regarded as located in the Gouy layer, as represented in Fig.4, where thr shear plane is designated by a dotted line. Comprehensive revieas and more details concerning the mathematical treatment of electroosmosis are provided in references[12-14]

Figure 4: a)Typical variation in potential and charge distribution with formation of stern layer; $\left|\sigma_{st}\right|$ is small compared with σ .b) when $\left|\sigma_{st}\right| > \sigma$, the ψ_d and ξ change sign ….., d plane , ……, shear plane.
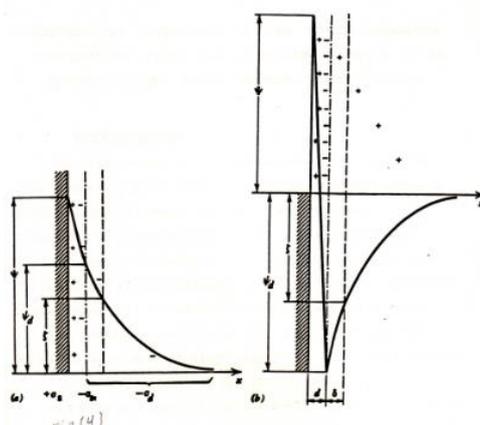
Postulation of a Novel Electrokinetic Effect Called Field Effect Electroosmosis:

As it was shown in previous section, up to now , the zeta potential, ξ, in a capillary has been manipulated as a function of pH(surface ionization) and the ionic concentration (specific adsorption). A novel phenomenon would be postulated by us in which the zeta potential in a capillary can be controlled by an external field. If a thin wall capillary is coated with a metallic conductor on the external surface and a voltage and a voltage V_G is applied between the metal electrode and the electrolyte, Fig.5 electroosmotic flow can be controlled. By changing V_G the voltage drop across the double layer changes, and this change includes the change in zeta potential, ξ, which in turn causes a modification in the electroosmotic flow. We name this phenomenon "external field effect electroosmosis" or just simply " field effect electroosmosis " . Therefore , zeta potential , ξ (pH,C,V_G), is function of three variables: pH,C, the ionic concentration in the electrolyte , and V_G. To understand this approach , there is a need to model the metal-electrolyte system in more detail, which has been addressed in the following sections of this manuscript.

Figure 5: a) A capillary covered with metallic coating. b) Cross section of Metal-Insulator-Electrolyte Field Effect Device Electro osmosis.
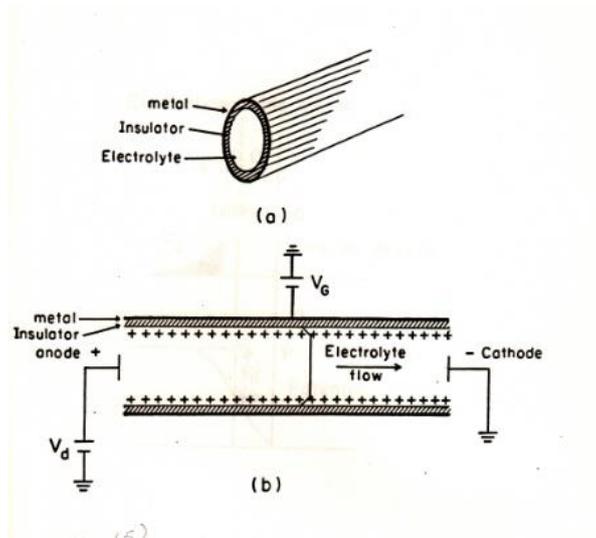
Ideal Metal-Insulator-Electrolyte Structures

In this section, the ideal metal-insulator-electrolyte(MIE) system is analyzed for the totally blocked interface[15]. The non-ideal case is studied in the following section. The ideal MIE system is similar to what Siuetal. [15] have defined as the totally blocked interface of an insulator/electrolyte. In an ideal MIE, there is a complete absence of interfacial reactions between the electrolyte and oxide or , in other words, there is neither specific adsorption nor surface ionization. Since the interfacial electrochemical processes are absent, the charge and potential distribution in this MIE system are dictated solely by electrostatic considerations. As showm in Fig.6, the metal electrode is chosen to be ground and the applied voltage, V_G, is applied to the reference electrode. The reference electrode is chosen as a nonpolarizableone , where the voltage drop across it is negligible, so it can be assumed that the V_G is applied to the electrolyte. The charge per unit area and the potential in the electrolye space-charge region are related by the Poisson-Boltzmann equation[16]. From Gauss's law and the solution to this equation, we find that for an electrolyte the charge per unit area in the Gouy-Chapman space-charge region is given by

$$\sigma_d = (8\varepsilon_e kTn_0)^{1/2} \sinh[q(V_G - \varphi_d)/2kT] \qquad (7)$$

From Fig.6, a charge neutrality equation could be written,

$$\sigma_d + \sigma_m = 0 \qquad (8)$$

And $C'_o = \dfrac{C_H C_o}{C_H + C'_o}$ since $C_H \gg C_o$ and $C'_o \cong C_o$ $\qquad (9)$

Since $C_d \gg C_o$ then $\varphi_d - \varphi_o \ll \varphi_o$, or $\varphi_d \approx \varphi_o$ $\qquad (10)$

And $\varphi_o C_o \approx -\sigma_m$ $\qquad (11)$

$-\sigma_m = \sigma_d \approx C_o \varphi_d$ $\qquad (12)$
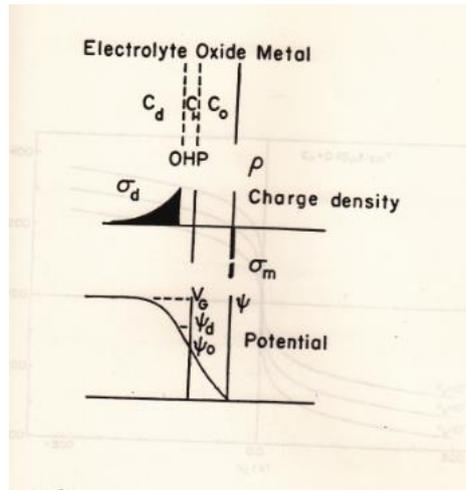
Thus, from eq.(7) to (12) we find

$$C_o\varphi_d = -(8\varepsilon_e kTn_0)^{1/2} \sinh[q(\varphi_d - V_G)/2kT] \quad (13)$$

Rearranging eq. (13) , we obtain

$$V_G = \frac{2kT}{q}\sinh^{-1}(\frac{\varphi_d C_o}{(8\varepsilon_e kTn_o)^{1/2}}) + \varphi_d \qquad (14)$$

$$\xi \approx \varphi_d - V_G = -\frac{2kT}{q}\sinh^{-1}(\frac{\varphi_d C_o}{(8\varepsilon_e kTn_o)^{1/2}}) \qquad (15)$$

Figure 6: The charge and potential profile in a totally blocked MIE system.

Since the difference between V_G and φ_d is very small(in the range of millivolts) , where as the difference between V_G and φ_d are several volts, by substituting φ_d in the paranthes is of eq.(15) with V_G, the calculations are simplified. Thus the following equation is found for the zeta potential:

$$\xi \approx \frac{2kT}{q}\sinh^{-1}(\frac{\varphi_d C_o}{(8\varepsilon_e kTn_o)^{1/2}}) \qquad (16)$$

For calculation of the inverse hyperbolic sine, it is convenient to use the following identity,

$$\sinh^{-1} x \approx Ln[x + (x^2 + 1)^{1/2}] \qquad (17)$$

Using eq. (18) and (19) , the zeta potential is plotted versus V_G in Fig.7. In interpreting this relation , it should be noted that since we are considering a limiting situation in which no interfacial processes occur, only electrostatic factors affect the overall response. It can be seen that by controlling V_G the zeta potential could be controlled. When V_G is close to zero , changing V_G causes a bigger change than when V_G has larger values. Also, it can be seen that the polarity of the zeta potential can be controlled through V_G. By lowering the ionic concentration , the change in V_G causes a relatively greater change in the zeta potential. To final C-V characteristics of the ideal MIE system, since C_d, C_H and C_o are in series, the resulting equation is

$$C_o' \approx \frac{C_H . C_o}{C_o + C_H} \text{ since } \qquad C_H >> C_o, \qquad \text{then} \qquad C_o' \approx C_o \qquad \text{and} \qquad C_T \approx \frac{C_d C_o'}{C_d + C_o'}$$

(18)

Where C_o is the oxide layer capacitance, C_H is the inner Helmholtz layer capacitance and C_d is the Diffuse layer capacitance. C_d could be found by taking the derivative of eq.(7),

as $C_d = -\frac{\partial d_d}{\partial \varphi_d} = (\frac{2\varepsilon_e n_o q^2}{kT})^{\frac{1}{2}} \cosh[q\frac{(\varphi_d - V_G)}{2kT}]$ (19)

substituting eq. (19) in (18) , C_T is obtained as follows :

$$C_T = \frac{C_o(\frac{2\varepsilon_o n_o q^2}{kT})^{\frac{1}{2}} \cosh[\frac{q(\varphi_d - V_E)}{2kT}]}{C_o + (\frac{2\varepsilon_e n_o q^2}{kT})^{\frac{1}{2}} \cosh[\frac{q(\varphi_d - V_G)}{2kT}]}$$

(20)

The difference $(\varphi_d - V_G)$ is the zeta potential , so by inserting eq. (16) into (20), C_T is found with respect to V_G.

$$C_T = \frac{C_o(\frac{2\varepsilon_o n_o q^2}{kT})^{\frac{1}{2}} \cosh \sinh^{-1}[\frac{V_G C_o}{(8\varepsilon_e kTn_o)^{\frac{1}{2}}}]}{C_o + (\frac{2\varepsilon_e n_o q^2}{kT})^{\frac{1}{2}} \cosh \sinh^{-1}[\frac{V_G C_o}{(8\varepsilon_e kTn_o)^{\frac{1}{2}}}]}$$

(21)

By using eq. (17) and the definition of $\cos x = \frac{(e^x + e^{-x})}{2}$, by simple algebraic manipulation the following identity is found:

$$\cos(\sinh^{-1} x) = x + \frac{1}{x + (x^2 + 1)^{\frac{1}{2}}}$$

(22)

Applying identity (22) to eq. (21), it can be converted to

$$C_T = \frac{\alpha_1 C_o^3 V_G^2 \alpha_2^2 + \alpha_1 \alpha_2 V_G C_o^2 ((V_G C_o \alpha_2)^2 + 1)^{\frac{1}{2}} + \alpha_1 C_o}{C_o^2 V_G \alpha_2 + C_o ((V_G C_o \alpha_2)^2 + 1)^{\frac{1}{2}} + \alpha_1 \alpha_2^2 V_G^2 C_o^2 + \alpha_1 \alpha_2 V_G C_o ((V_G C_o \alpha_2)^2 + 1)^{\frac{1}{2}} + \alpha_1}$$

(23)

Where

$$\alpha_1 = (\frac{2\varepsilon_e n_o q^2}{kT})^{\frac{1}{2}} \qquad , \qquad \alpha_2 = \frac{1}{(8\varepsilon_e kTn_o)^{\frac{1}{2}}}$$

C_T is plotted versus V_G in Fig. 8. At voltage close to zero , C_d is dominant. As the concentration of the electrolyte decreases, the C_d value becomes smaller at voltages around zero , and its effect becomes more significant on the total capacitance, C_T.

Figure 7: Change in zeta potential ξ as a function of V_G for various ionic concentrations for ideal case.
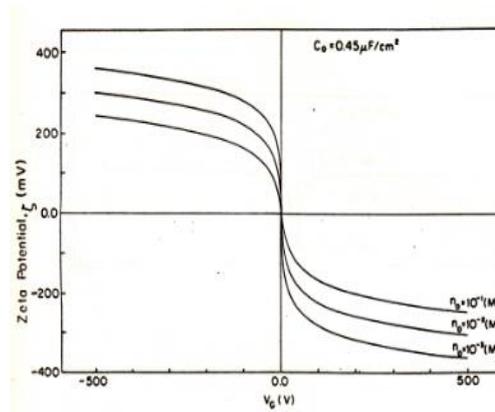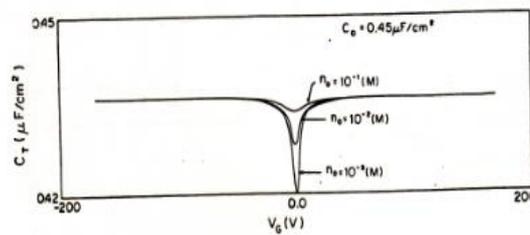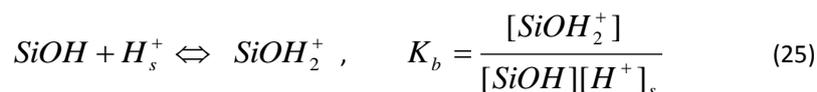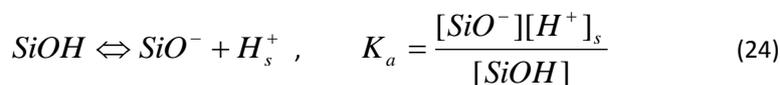

Figure 8: Theoretical C-V response of an ideal MIE (Metal-Insular-Electrolyte) for various ionic concentration.

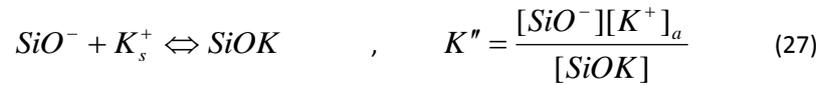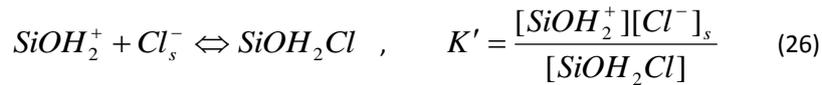## Nonideal Metal-Insulator-Electrolyte Structures

Glass is used as the insulator because it has a very high electric field breakdown (the reason for the selection of a dielectric with high electric field breakdown is given later) , and there exists the well established site binding model of Yates etal[17].For oxides and electrolytes interaction.

SiO_2 interacts with the electrolyte by surface adsorption processes that are in equilibrium with the diffused charges in the Gouy-Chapman layer. The interactions according to the site binding model of Yates etal. [17] are as following: (i) when the SiO_2 is in contact with an equeaus solution , its surface hydrolyzes to from silanol surface groups. These groups may be positively charged , negatively charged, or neutral , depending on the electrolyte pH.For SiO_2 , the surface group density is of the order of $5 \times 10^{14} cm^{-2}{}^{18}$ and the chemical equilibria can be described by the following equations,

$$SiOH \Leftrightarrow SiO^- + H_s^+ \ , \qquad K_a = \frac{[SiO^-][H^+]_s}{[SiOH]} \qquad (24)$$

$$SiOH + H_s^+ \Leftrightarrow SiOH_2^+ \ , \qquad K_b = \frac{[SiOH_2^+]}{[SiOH][H^+]_s} \qquad (25)$$

$[H^+]_s$ is the Concentration of protons at the surface. $[SiO^-], [SiOH]$ and $[SiOH_2^+]$ are the surface sites at the oxide.

(ii) In addition to the amphoteric dissociation mentioned previously, the supporting electrolyte also can from ion pairs with the charged surface sites. This process will be referred to as surface complexation. It is illustrated below using KCl as a typical, univalent supporting electrolyte.

$$SiOH_2^+ + Cl_s^- \Leftrightarrow SiOH_2Cl \quad , \qquad K' = \frac{[SiOH_2^+][Cl^-]_s}{[SiOH_2Cl]} \qquad (26)$$

$$SiO^- + K_s^+ \Leftrightarrow SiOK \qquad , \qquad K'' = \frac{[SiO^-][K^+]_a}{[SiOK]} \qquad (27)$$

$[SiOH_2Cl]$ and $[SiOK]$ are the surface sites at the oxide. The total number of sites per unit area, is given by

$$N_s = [SiOH] + [SiOH_2^+] + [SiO^-] + [SiOH_2Cl] + [SiOK] \qquad (28)$$

The concentration of $[Cl^-]_s, [K^+]_s$ and $[H^+]_s$ at the surface could be related to the bulk electrolyte by Boltzmann distribution given by the following equations,

$$[H^+]_s = [H^+]\exp(^{-q\varphi_a}\!/_{kT}) \qquad (29)$$

Where $\varphi_a = \varphi_o - V_G$

$$[Cl^-]_s = M\exp[\frac{q\psi_b}{kT}] \qquad (30)$$

$$[K^+]_s = M\exp[\frac{-q\psi_b}{kT}] \qquad (31)$$

Where $\varphi_b = \varphi_\beta - V_G$ and M is the concentration of bulk electrolyte. By solving the above equations, a complex analytical function can be found which relates V_G to ξ.[19]

In reference 19 only the theory is acceptable the experimental result prove to auther to be irriproduceable. This theory is made on previous theories and mathematical modeling in the field.

Figures 9 and 10 depict the zeta potential as a function of V_G for different pH's and different concentration of KCl solution. For these figures the oxide thickness of $1\mu m$ has been chosen. The V_G in Figures 7 and 8 range from-500 to +500 volts and these voltages can create a maximum electric field of $5\times10^6 \, V\!/_{cm}$, Which is below the dielectric strength of SiO_2, $1\times10^7 \, V\!/_{cm}$. In order to manipulate the voltage distribution in the double layer , one needs to apply electric field with in the same order of magnitude as the electric field in the double layer. That is why an insulator with high dielectric breakdown like SiO_2 is required. For higher Oxide thickness the voltage V_G must be increased in order to generate ligh enough electric field for the manipulation of the zeta potential , ξ.

Figure 9: Change in zeta potential ξ as a function of V_G for different pH and constant concentration with the oxide thickness of 1 (μm).
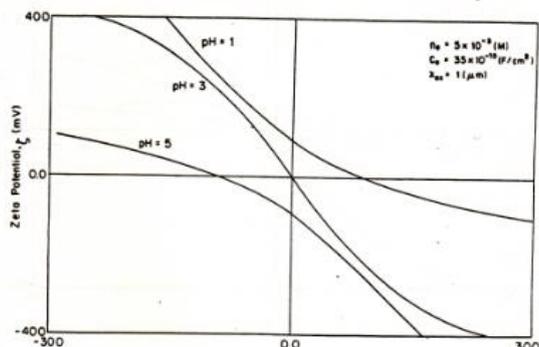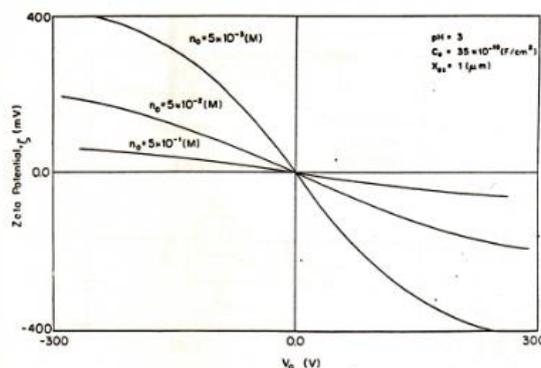

Figure 10: Change in zeta potential as a function of V_G for different concentrations and constant pH with the oxide thickness of 1 ((μm).

The suitable range for voltage per oxide thickness would be $\pm 500V/\mu m$. For instance, for 10 μm and 100 μm the proper ranges of V_G would be $\pm 5KV$ and $50KV$, respectively.

Field Effect Steaming Potential

The first new electrokinetic phenomenon we presented , was Field Effect Electroosmosis which could be seen from eq. 1. If ξ is function of two variables concentration of electrolytes and pH, electroosmotic velocity could be controlled by these two variables. We postulated that there is a third variable, V_G ,the potential applied perpendicular to double layer of a capillary, which zeta potential is function of it , V_G , . Therefore by changing the V_G zeta potential is changed and consequently the electroosmotic velocity is changed.

The equation 2 describe the potential arising in the capillary under the effect of a pressure p and a specific conductivity of the liquid. We can postulate another new Electrokinetic phenomenon called Field Effect Streaming potential. This new effect similar to Field Effect Electroosmosis is stablished as zeta potential

is function of three variables , concentration of electrolyte , pH and , V_G , the potential perpendicular to the double layer in the capillary. V_G is the new proposed variable that zeta potential is function of. According to eq. (2) if zeta potential is charged by V_G the streaming potential , ΔV_{str} is charged.

CONCLUSION

The metal-insulator structure has been modeled mathematically. It is shown that the zeta potential , the potential at the diffuse layer of the double layer, can be changed by varrying the voltage applied across metal-insulator-electrolyte(MIE). By using the MIE system with a capillary , two new electrokinetic effects can be postulated. These two effects are called Field-Effect-Electroosmosis and Field-Effect-Streaming potential.

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