Physicochemical Analysis at the Interface between Conductive Solid and Dielectric Liquid for Flow Electrification Phenomenon

M. EL-Adawy, T. Paillat, Y. Bertrand, O. Moreau, and G. Touchard

Abstract—At the solid-liquid interface, a charge zone called the Electrical Double Layer (EDL) appears. It is constituted of two zones of opposite sign, one in the solid and another one in the liquid. When a liquid flows through a pipe, an axial streaming current is generated. This current is due to the convection of the charges coming from the electrical double layer. The physicochemical reaction at the solid-liquid interface is one of the most important parameters which control the diffuse layer development inside the liquid, and consequently the space charge density. In this paper, we present an analysis for the physicochemical reaction in the case of liquid containing additives or impurities partially dissociated into positive and negative ions. The addition of impurities, in our case, is associated with changing in both the polarity and the value of streaming current. Thus, from this difference in streaming current, we will be able to identify the reagent of the oil with the solid material. Also, the effect of streaming potential on the EDL has been undertaken. Moreover, the fully developed space charge density at the wall is calculated with the help of streaming electrification experiments which are conducted in the case of non-fully developed EDL. This procedure can be conducted for the investigation of flow electrification in transformers with oil/metal configurations.

Index Terms—Electrical double layer, Flow electrification, Physicochemical reaction coefficient, Streaming current.

I. INTRODUCTION

FLOW electrification has been extensively studied, since over 50 years ago, due to its economical and theoretical interest. It has been found to be the cause of at least a dozen failures of large forced-oil cooled power transformers, and in a compact HVDC substation. This phenomenon, occurring in large power transformers, needs further study to obtain a reliable design of such transformers particularly with regard to the charge separation process occurring at the solid-liquid interface. Convection of liquid creates a streaming current and leads to a continuous charge separation process at the interface. When the solid is insulated from the ground, leakage impedances limit the accumulation of these charges at the wall. This leads to potential buildup and hence discharges on the surface of the solid material. Finally it may cause major discharge damage as in the case of large oil-cooled power transformers [1]-[3]. To date, however, there are a dozen failures of large oil-cooled power transformers attributed to this static electrification phenomenon. The predominant failure mode of these transformers involved internal flashover across considerable oil distances, and these failures have alarmed both transformer manufactures and users [4], [5].

From the theoretical point of view, many papers have been published which deal with the mathematical description of the EDL [6]-[8]. These papers describe the distribution of the potential or of the space charge density in the liquid. They are always based on one parameter, which depends only on the physicochemical proprieties of both the solid and the liquid. In most cases, this parameter is the zeta potential (ζ) or the Helmholtz potential (ψ_0). In other cases, it is the space charge density on the wall (ρ_{wd}) for a fully-developed EDL [9]. Also, the diffuse layer thickness (δ_o) and the EDL development time (τ) should be considered due to there effect on the EDL.

One of the most important parameters, the physicochemical reaction at the pipe wall-liquid interface which induces the flow electrification remains very difficult to quantify. When a liquid solution is in contact with a solid surface, a physicochemical reaction occurs at the solid-liquid interface, leading to the formation of an EDL. This layer is called "double" because charge of one sign collects on the solid surface, while the opposite charge remains in the liquid. The process of flow electrification is caused by the axial convection of the diffuse liquid part of the EDL appearing at the solid-liquid interface. Thus, the rate of the wall reaction and the resulting charge concentration in the liquid may control the flow electrification.

The purpose of this paper is to analyze the physicochemical

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reaction at the solid-liquid interface in the case of liquid containing additives or impurities partially dissociated into positive and negative ions. This reaction can be adsorption or corrosion (oxido-reduction) reaction depending on the type of solid which is in contact with the flowing liquid. Moreover, the fully developed space charge density at the wall is calculated with the help of streaming electrification experiments that are conducted in the case of non-fully developed EDL. This procedure can be conducted for the investigation of flow electrification phenomena in transformers with oil/metal configurations. Special attention should be considered for the evolution time of experiments to avoid changing the interfacial reaction rates and consequently the value of space charge density. Thus, it is very useful to carry out all the experiments in times which are close together [1].

II. Physicochemical Reaction

As soon as this liquid is in contact with the solid surface, the complex solid-liquid initially neutral becomes polarized under physicochemical reactions occurring at the interface. Such reactions generate a specific value of the streaming current depending on the type of the solid material and the chemical composition of the liquid, such as transformer oil, which is difficult to be characterized.

Furthermore, even in a highly purified state, the hydrocarbon liquid oil can contain impurities that can undergo chemical reactions and affect the charge generated by flow electrification. Since these impurities are usually present in trace amounts, their composition tends to be unknown [10]. Also, additives may be used intentionally, e.g. to reduce the electric resistivity of the liquid or to significantly modify the EDL.

Different models have been proposed for modeling the chemical reaction process occurring at the solid-liquid interface which induces the EDL.

A. Corrosion Model

The corroding model [5] assumes that impurity or additive $A_L B_L$ is weakly dissociated in the liquid. Thus we have the following equation (dynamic dissociation equilibrium)

$$A_L B_L \underset{k_{r1}}{\overset{k_{d1}}{\rightleftharpoons}} A_L^+ + B_L^- \tag{1}$$

where k_{d1} and k_{r1} are the kinetic constants of the dissociation reaction. When an initially electrically neutral liquid comes in contact with a solid surface, a physicochemical reaction starts at the solid-liquid interface. This reaction depends on the nature of the solid and the liquid additive. Also, it is assumed that the solid surface is partly or totally composed of molecules $C_s D_s$ and undergoes the following reaction in presence of the liquid (dynamic dissociation equilibrium)

$$C_s D_s \underset{k_{r2}}{\overset{k_{d2}}{\rightleftharpoons}} C_s^+ + D_s^- \tag{2}$$

where k_{d2} and k_{r2} are the kinetic constants of the wall corrosion reaction. Then when the cations C_s^+ are in contact with the anions of the liquid B_L^-, a surface reaction can occur in the liquid region as follow: (ionic bound)
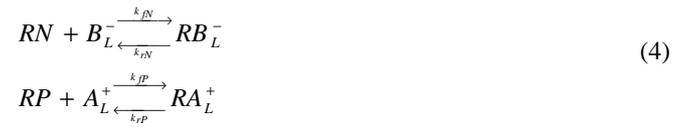
$$C_s^+ + B_L^- \underset{k_{r3}}{\overset{k_{d3}}{\rightleftharpoons}} C_s B_L \tag{3}$$

where k_{d3} and k_{r3} are the kinetic constants of the ions recombination reaction.

The balance sheet of the reactions (1), (2) & (3) is:

$$A_L B_L + C_s D_s \leftrightarrow A_L^+ + C_s B_L + D_s^- \tag{4}$$
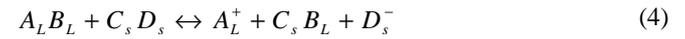
B. Adsorption Model

When a liquid is in contact with a solid, the preferential adsorption of charged species of the liquid by the solid induces the development of an EDL. However, in accordance with (1) and (2), this reaction can be modeled, in the solid region, as follow:

$$RN + B_L^- \underset{k_{rN}}{\overset{k_{fN}}{\rightleftharpoons}} RB_L^-$$

$$RP + A_L^+ \underset{k_{rP}}{\overset{k_{fP}}{\rightleftharpoons}} RA_L^+ \tag{4}$$

with RN and RP are the free active radicals of the cellulose, or the part of C_s^+ and D_s^-, that will react (electrostatic interaction) with B_L^- and A_L^+ respectively. RB_L^- and RA_L^+ are the occupied active radicals while $k_{fP/N}$ and $k_{rP/N}$ are the kinetic constants of adsorption and desorption reactions of positive and negative ions respectively.

Thus, the main difference between these two models is the mass transfer. For the corrosion model, the mass transfer is from the solid towards to the liquid, while for the adsorption model, it is from the liquid towards to the solid. Irrespective of the mechanism of transfer, these two models lead to the polarization of both the liquid and the solid at the equilibrium states. These reactions are equivalent to positive current at the solid-liquid interface, and they lead to an accumulation of positive ions in the liquid, while the negative ones react on the solid.

Concerning the mechanisms of mass transfer, they have different constants of reaction and consequently they have different kinetic development of the EDL. Also, these models haven't considered the effect of wall shearing stress on the interfacial process, especially for high laminar Reynolds numbers.

However, for these two models, most recent researches [1], [5], [11] indicated that the concentration of C_s^+ is not rate controlling and varies with the shearing stress. This means that the ionization process at the wall should be a function of the wall shearing stress and not only a chemical reaction.

Thereby, the currents are generated by differences in the ionic diffusivities and/or by differences in the ionic adsorption or corrosion rates at the wall. When these source terms give current of opposite sign it is possible for the polarity of the current generated in a pipe to change as the electrical conductivity of the liquid is increased [6].

It has been shown previously that a reasonable model for streaming current phenomena may be deduced from physicochemical reactions at the wall between liquid additives or impurities and ions coming from the solids. Starting from the principle of charge conservation, and taking into account the equilibrium conditions for these reactions, we can determine the value of physicochemical reaction coefficient K_f as in [5].

C. Calcium Alkyphenate OLOA 218 A

To control the additive which is responsible of the EDL development in the oil, we introduced some additives such as *OLOA 218* and *OLOA 219*. These types have been selected because they have very well known structure [12], and they are highly soluble in the oil. Moreover, adding these types into some liquids like heptane, there will be changing the EDL sign such has been observed in [9].

Bouregois [13], [14] has observed that, in the case of oil-cellulose material (pressboard, paper …) interface, when adding certain additives to the pure oil, there is inversing in the sign of the measured electric charge and consequently the sign of the streaming current. This phenomenon of inversion of sign has been occurred with different types of additives but all these additives have in common the presence of non-binding pairs of electrons in the external orbital of several atoms (*i.e.* electron pairs not involved in a covalent bond).

From the molecular structure of *OLOA 218* shown in Fig. 1, it can be seen that the most reactive parts are the *Ca-O* groups. Thus, the *Ca* atoms establish a covalent bound with the *O* from the aromatic cycles, and an ionic bond with the other one. As a result, there is appearing of negative *OH* ions in the oil. Beside, both *O* atoms bear non-binding electron pairs (2 and 3 respectively). The benzenic cycles of the molecule are stable and will not ready undergo additional chemical reactions. But we can note the presence of a disulfide bridge between these 2 aromatic cycles, where each of the sulfur atoms bear 2 non-binding electron pairs. Also these types of additives have very high solubility in the oil due to the two long hydrocarbon chains $C_{12}H_{25}$.
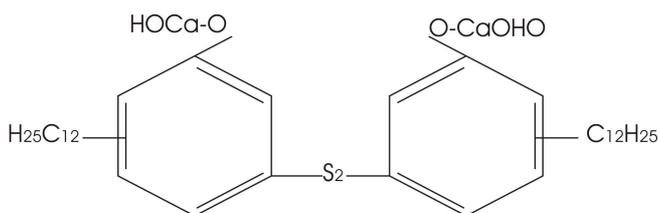
III. THEORETICAL MODEL

Inside the liquid, the Stern model [5] distinguishes two different regions: the compact layer so close to the wall that its electrical charges are not affected by the liquid flow, and the diffuse layer for which the space charge density ρ_w decreases when one moves away from the wall according to Boltzmann's law. At the wall, there is a maximum value of space charge density ρ_{wd}. The streaming current is consequently due to the convection of charges coming from the diffuse layer inside the liquid. Thus, it depends mainly on the properties of the electrical layer: diffuse layer thickness or Debye Length δ_o, space charge density at the wall and the EDL development time τ [5].

So the classical form for the wall current is the following [2]:

$$i_w = K_f \left(\rho_{wd} - \rho_w \right) \tag{6}$$

The coefficient K_f is related to the intensity of the physicochemical reactions (in the classical model it is considered to be only dependent of the liquid-solid couple). Previous experiments [15] show that the space charge development time in the diffuse layer is much faster than the evolution of the wall space charge density. This means that the relaxation time $\tau_c = \varepsilon/\sigma_0$ is much smaller than the time needed for the development of the diffuse layer at the interface, where ε is the liquid's dielectric constant and σ_0 its bulk conductivity. Under these considerations, the space charge density profile is a quasi-static profile. Thus, it is reasonable to assume that all along the channel axis, the space charge density profile is similar to a fully developed diffused layer profile.

To carry out our calculations for space charge density and coefficient of physicochemical reaction, consider a rectangular cross section channel of half thickness a and width D as shown in Fig. 2, where $a << D$, which allowing us to treat the configuration as a parallel plate capacitor with infinite plane electrodes.

If we assume that the time needed for the formation of the diffuse layer profile (in the X-direction) is much shorter than the time needed for the development of space charge at the interface, and also shorter than the residence time of a particle in the duct during the convection, the space charge density in the liquid (in the case of weak space charge density) is given by [1]:
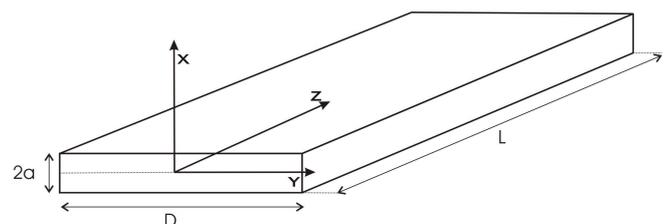

Fig. 1. The structure of OLOA 218.

Fig. 2. Rectangular channel and associated coordinate system.

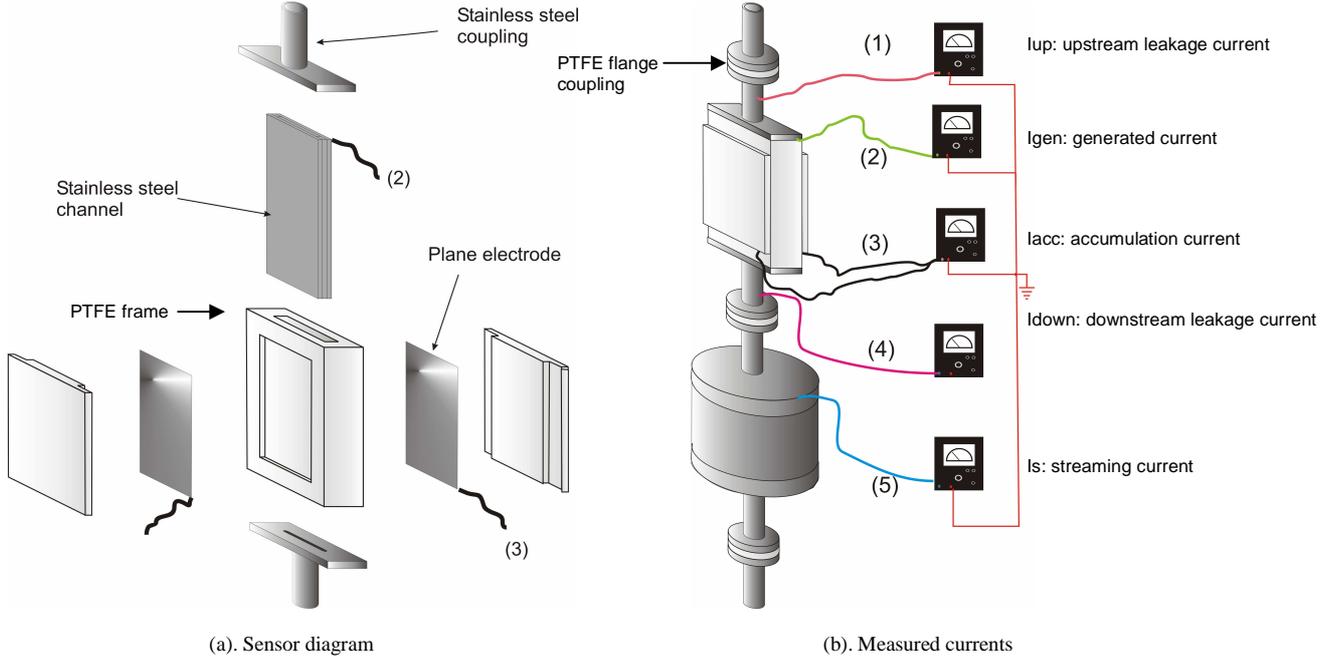(a). Sensor diagram   (b). Measured currents

Fig. 3. Experimental setup showing the sensor diagram and measuring schemes for the current

$$\rho(x,z) = \rho_w(z)\frac{\cosh(x/\delta_0)}{\cosh(a/\delta_0)}, \qquad \delta_0 = \sqrt{\frac{\varepsilon D_0}{\sigma_0}} \qquad (7)$$

where D_0 is the mean diffusion coefficient. The velocity profile for a laminar flow in the channel is:

$$U(x) = \frac{3}{2}U_m\left(1 - \frac{x^2}{a^2}\right) \qquad (8)$$

where U_m is the mean flow velocity. The streaming current due to the charge convection could be obtained through the following surface integral:

$$I_s(z) = \int_{-D/2}^{D/2}\int_{-a}^{a}\rho(x,z)\ U(x)\ dxdy = 2DC_1\rho_w(z) \qquad (9)$$

where C_1 can be determined as follow:

$$C_1 = C_1(U_m) = 3\left(\frac{\delta_0}{a}\right)^2\left[a - \delta_0\tanh(a/\delta_0)\right]\ U_m \qquad (10)$$

For a given liquid and channel configuration parameters, C_1 is a function of the mean flow velocity U_m. Starting from the principle of charge conservation, one can obtain the following expression for surface charge density ρ_w [1], [15]:

$$\rho_w(z,U_m) = \rho_{wd}\left[1 - \exp\left(\frac{-K_f z}{C_1}\right)\right] \qquad (11)$$

Equation (11) shows the dependence of surface charge density, and consequently the streaming current, on both the mean flow velocity and the value of chemical reaction coefficient. Also, it can be used for the determination of space charge density associated with flow electrification experiments for the fully-developed EDL.

Many papers [16]-[23] have been published that deal with the methods for the determination of streaming current in the case of fully developed EDL, in fact, for an infinite solid-liquid contact time. For a partially developed double layer, it is difficult to determine the value of streaming current because the double layer is not completely formed, this is the reason which we called it partially developed EDL. Thus, it is seems to be better to determine the value of space charge density experimentally and then using a simplified procedure [1], [11] to determine the value of streaming current. Analyzing experimental results which were made in a metallic capillary [5], it appears that the coefficient K_f of the process is a function of the wall shearing stress. In other words, this means that the physico-chemical process on the wall must be a function of wall shearing stress and not only a chemical reaction.

IV. EXPERIMENTAL SETUP

The facility used to obtain our experimental results was developed a few years ago as a part of the research program of "Electricité de France" and the University of Poitiers, France. It consists of a closed loop where transformer oil flows through a sensor which is consisted of a rectangular stainless steel channel inserted in a PTFE frame as shown in Fig. 3a. Charge leakage takes place towards two stainless steel couplings placed at both extremities of the sensor, and insulated from the rest of the loop by PTFE flange couplings.

Two plane electrodes are placed facing the external surfaces of the stainless steel channel, beyond 2 mm of PTFE, to measure a mainly capacitive current I_{acc}, which is related to the charge trapped inside the stainless steel channel (accumulation charges). A precise description of this facility and the sensor is described in detail in [24]. A small modification has been added to this sensor by connecting the stainless steel channel to a wire in order to grounding it through the ammeter 2, as shown in Fig. 3b, (grounded metal configuration) and measure the generated current or to leave it floated (isolated metal configuration). This enables us to study the effect of the potential on the EDL. In this work, the streaming current is produced by oil flowing through the rectangular channel (3×30 mm cross section, and 300 mm in length), as a function of time interval (at rest) between two consecutive measurements for a given flow velocity, and as a function of oil mean flow velocity.

V. STREAMING CURRENT MEASUREMENT

The typical experimental measured values of currents as a function of time during flow and after the flow is stopped are shown in Fig. 4 for the isolated metal. Fig. 5 shows these currents for the grounded metal one. After a time dependent on the initial conditions, the absolute value of the streaming current reaches a maximum, then starts to drop more and less rapidly to constant value. In fact, the peak value represents the convection of a diffuse layer, developed at rest during a time t. Once the flow begins, the diffuse layer evolves towards a dynamic equilibrium, which does not depend upon initial conditions. The measurements, for the same couple of solid-liquid, of steady-state value of streaming current as a function of mean flow velocity (dynamic case [1]) are shown in Fig. 6 for the two configurations with the extrapolation of these measurements. Also, it shows a remarkable dependency on the mean flow velocity as reported before [1]-[9]. In these series of experiments, the oil proprieties were: σ_0=7.5×10^{-12} S/m, D_0=4×10^{-11} m^2/s, and ε=1.95×10^{-11} F/m. These values give a double layer thickness of δ_0= 10.20 µm. The space charge
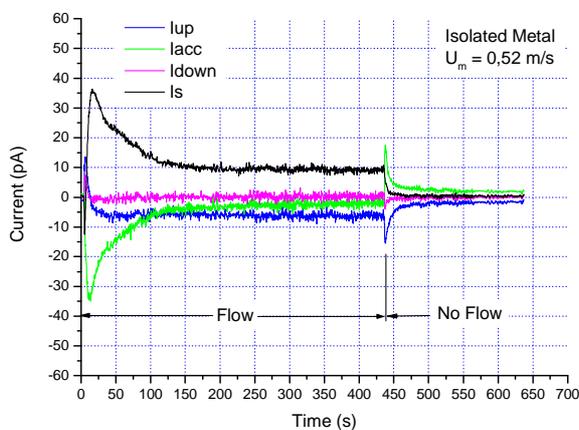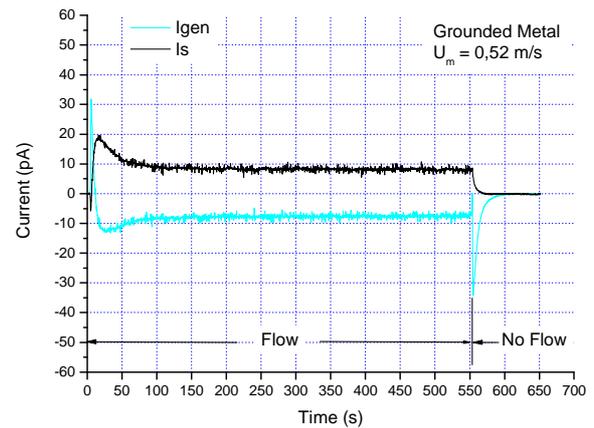

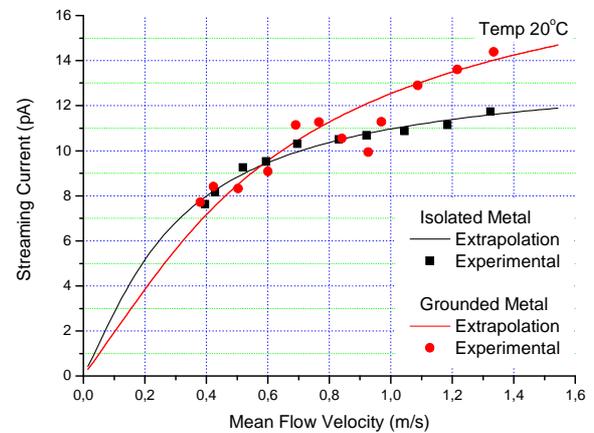Fig. 5. Measured currents versus time for the grounded metal configuration.

Fig. 6. Comparison of the stationary values of streaming currents for the two configurations.

density at the wall for a fully developed double layer can be calculated according to the simplified procedure which is described in detail in [1]. However these values of ρ_{wd} are of 2.27×10^{-3} and 1.42×10^{-3} C/m^3 for isolated and grounded metal respectively. These values are of the same order of magnitude than those reported in previous flow electrification studies concerning hydrocarbon liquids [1], [2], [11], [15]. Regarding the physicochemical reaction coefficient K_f, its value was 3.40×10^{-7} and 8.71×10^{-7} m/s for isolated and grounded metal respectively. From these results, we can note that, the potential build-up on the metal part affects both the space charge density and the physicochemical reaction coefficient. Also, there are changing in both the values and the comportments of the streaming currents as the flow velocity increases as shown in Fig. 6. To confirm these results, the application of different potential values has been undertaken as shown in Fig. 7 and it leads to the same effect which is discussed before.

During tests, the streaming current's peak value varies depending on the time at which the flow was stopped between two consecutive runs, for a given flow rate. Also, the peak value increases with time to reach a constant value after long time (t→∞). These curves can be extrapolated to obtain the

Fig. 4. Measured currents versus time for the isolated metal configuration
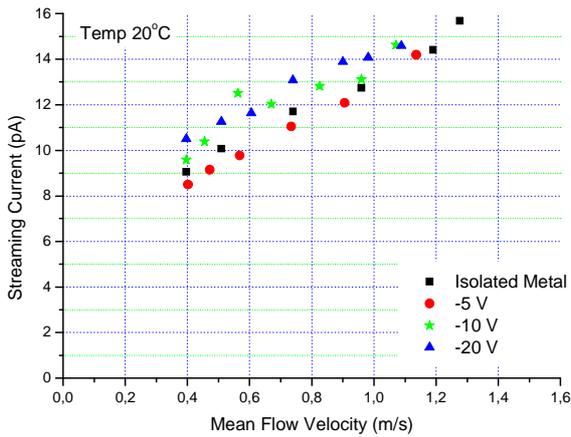
Fig. 7. Effect of the application of different potentials on the stationary values of currents.

steady state value as shown from Fig. 8. Also, there is a difference between the peaks values of the streaming currents for both configurations and consequently this difference affects the values of space charge density for each case.

With (9), and considering that the profile of $\rho_w(z)$ is constant along the channel (at rest), the value of space charge density at the wall can be obtained as described in [1]. In these series of experiments, the oil properties were: $\sigma_0=8.8\times10^{-12}$ S/m, $D_0=4\times10^{-11}$ m^2/s, and $\varepsilon=1.95\times10^{-11}$ F/m. The difference in oil conductivity is attributed to the fact that the interface between the solid and liquid has been changed and can be explained as follows: the strong dependency of space charge density on the reaction coefficient K_f has been noticed in (11), and the reaction rate affects the magnitude, but not the polarity, of the space charge perturbation by the flowing current. Also, the past history of an experiment causes the interface to be slightly different, for example, the passage of current through the interfaces may cause the formation of an oxide which will have a different thickness on each interface [10]. Consequently, it will be useful to carry out all the experimental measurements through times which are close together, as soon as possible, to avoid divergence in the
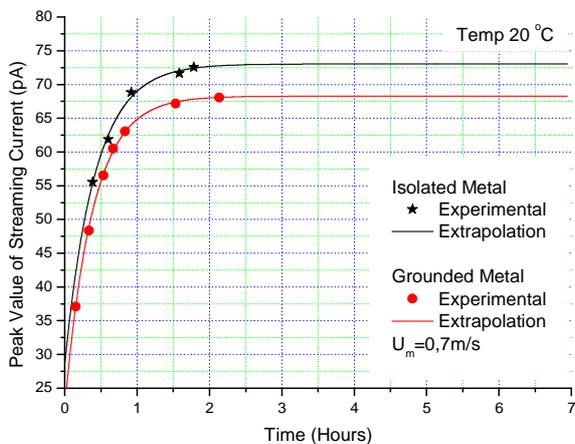
calculation of space charge density [1].

These values give a double layer thickness of $\delta_0=9.41$ μm. The fully developed space charge density ρ_{wd} are of 5.47×10^{-3} and 5.11×10^{-3} C/m^3 for isolated and grounded metal respectively. Comparing these values of space charge density with those are obtained before, we can note the effect of potential build-up on the metal part which cause these changes in the values of space charge density.

VI. EFFECT OF IMPURITIES OLOA 218

The oil under test has been doped with 30 ppm of additives *OLOA 218* and it was noted that there are changes of amplitudes and signs of all currents as shown in Fig. 9. This effect has been tested also for different flow velocities as shown in Fig. 10. In these series of experiments, the proprieties for the additived oil are: $\sigma_0=2.6\times10^{-11}$ S/m, $D_0=4\times10^{-11}$ m^2/s, and $\varepsilon=1.95\times10^{-11}$ F/m. These values give a double layer thickness of $\delta_0=5.48$ μm. The space charge density ρ_{wd} are of -2.15×10^{-2} and -1.11×10^{-2} C/m^3 for isolated and grounded metal respectively. Regarding the
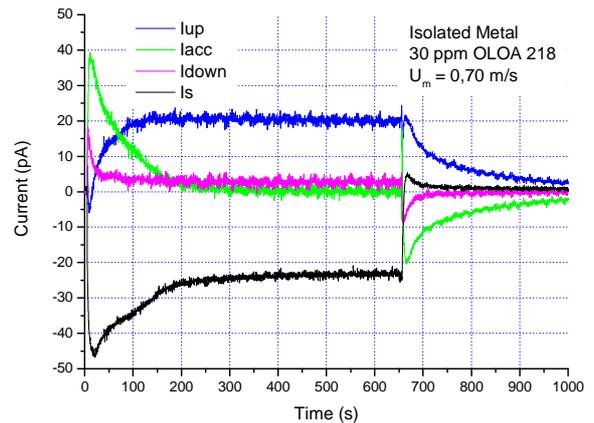

Fig. 9. Measured current versus time for the isolated metal configuration and oil doped with 30 ppm OLOA 218.

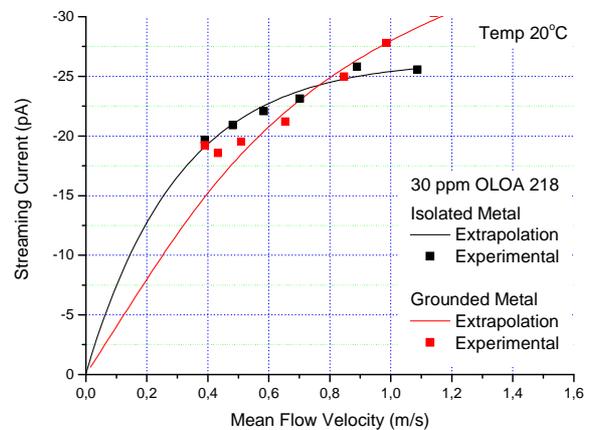Fig. 8. Peak streaming current values as a function of time interval between consecutive runs.

Fig. 10. Comparison of the stationary values of steaming currents for the two configurations and oil with 30 ppm OLOA 218.

physicochemical reaction coefficient K_f, its value is 7.97×10^{-8} and 2.40×10^{-7} m/s for isolated and grounded metal respectively. By comparing the results with and without impurities, K_f is changed depending on the additive concentration. This means that physicochemical reactions at the wall are smaller when the liquid contains impurities or additive ions [9]. Again, there are changing in the values and the comportments of the streaming currents as the flow velocity increases as shown in Fig. 10. The peak value of streaming current versus the time at which the flow was stopped between two consecutive runs for the isolated metal case and with oil doped with 30 ppm *OLOA 218* additives is shown in Fig. 11. Similar results have been obtained when changing the type of additives to *OLOA 219*.
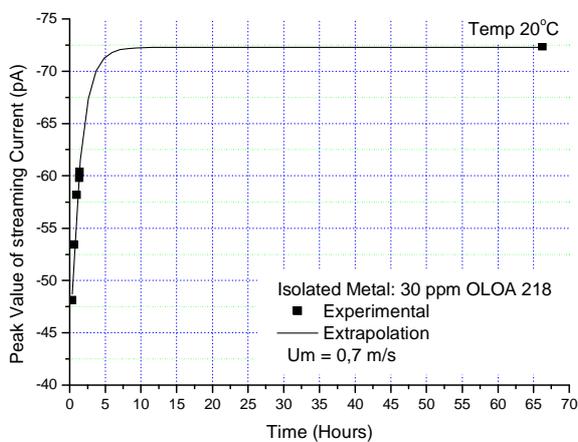

Fig. 11. Peak streaming current values as a function of time interval between consecutive runs for oil with 30 ppm OLOA 218.

Comparing this case with those before the additions of impurities, you can note that there is a change of the amplitude and the sign of the streaming current and consequently the values of space charge density and physicochemical coefficient. Regarding these changes after adding the impurities, we can confirm that, this is the effect of impurities on the physicochemical reaction at the metal-liquid interface. This reaction can be either corrosion or adsorption reaction or a combination between them.

This trend is attributed to the fact that, because of the unavoidable trace amounts of water in oil, the dissociation of ionic bond Ca^+OH^- in the additive is possible, leading to the appearance of positive $R–Ca^+$ and OH^- negative ions in the liquid.

VII. DISCUSSION

Before adding the impurities to the oil, the streaming current was positive and the charge accumulated at the solid surface was negative.

The modification, which has been made in the sensor, enabled us to confirm that the potential build-up on the metal part affects the space charge density, the physicochemical reaction coefficient. Also, there is changing in the profile of streaming current when fixing the potential of the metal part especially for higher flow velocities.

After the addition of impurities OLOA 218, there are signs reversal of both the streaming current and the charge accumulated at the solid surface and this trend could be explained depending on the type of reaction as follow:

If we considered the corrosion reaction, the negative ions of the solid D_s^- will undergo chemical reaction with the positive ion Ca_2^+ leaving inside the liquid the negative ions O_2^-. These reactions lead to an accumulation of positive ions in the solid and hence negative streaming current.

For the adsorption reaction, the positive ions Ca_2^+ are preferentially adsorbed by the solid material to react within it leaving negative ions in the liquid. Hence, this reaction leads to accumulation of negative ions in the liquid.

However, from these different values of streaming currents before and after the addition of impurities, we were able to identify the reagent of the oil with the solid material.

VIII. CONCLUSION

In this paper, we presented an analysis for the physicochemical reaction in the case of dielectric liquid containing additives or impurities partially dissociated into positive and negative ions which is in contact with a conductive solid material. This reaction can be corrosion, adsorption or combination between them depending on the solid which is in contact with the flowing liquid. However, before the addition of impurities, these reactions are equivalent to positive current at the solid-liquid interface, and they lead to an accumulation of positive ions in the liquid, while the negative ones react on the solid.

The addition of impurities of very well known structure, such as *OLOA 218* and *OLOA 219* leads to changing the amplitude and the sign of the streaming current. This difference in the values of streaming currents helps us to identify the reagent of the oil with the solid material.

The modification, which has been added to our facility, enabled us to study the effect of the potential on the EDL and consequently on both the streaming current and space charge density. We noticed that there are changing in both the values and the comportments of the streaming currents as the flow velocity increases.

The calculated space charge density for a fully developed EDL is determined in the case of non-fully developed EDL. Also, with respect to the evolution time of experiments, it was usefully to carry out all the experiments in times which are close together to avoid changing in the value of space charge density which depends upon the interfacial reaction rates.

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