

Flow Electrification Investigation Under the Effect of the Flow Parameters

P. Leblanc¹, T. Paillat¹, J. M. Cabaleiro², and G. Touchard¹

¹Electrofluidodynamic group, P' Institute, University of Poitiers, CNRS, France

²Laboratorio de Fluidodinamica (LFD), Facultad de Ingenieria, Universidad de Buenos Aires, Argentina

Abstract—When a liquid is in contact with a solid, a physicochemical reaction occurs at the solid-liquid interface leading to the formation of the electric double layer. Whenever the mobile portion of this charge inside the liquid is swept by the flow, a chemical reaction occurs and leads to the flow electrification process. According to the classical theory of this phenomenon, it is assumed that the laminar flow has no impact on the physicochemical behavior at the interface. However, recent studies have shown that this supposition is not consistent with the experimental results. The aim of this work is to highlight the impact of the flow parameters on the flow electrification phenomenon in the case of mineral oil flows through a stainless steel capillary. In this purpose, the capillary is shortened to keep the same contact duration between the solid and the liquid at different mean flow velocities.

Keywords—Electrical double layer, flow electrification, wall shearing stress, chemical reactions

I. INTRODUCTION

The flow electrification phenomenon has been studied for a long time [1]–[5]. Although all studies carried out until now on the influence of different parameters have provided a better understanding of this phenomenon, the physicochemical process occurring at the solid-liquid interface and responsible of the development of the electrical double layer remains poorly understood. Recently, some studies [6]–[8] have shown that the flow parameters, such as the wall shearing stress, probably play an important role on the global rate of the physicochemical reactions K_f . Indeed, different authors have proven that K_f is impacted by the mean flow velocity (U_m) of the liquid. The aim of this work is to confirm this behavior in the case of mineral oil flows through a stainless steel capillary with a specific experimental protocol.

II. METHODOLOGY

When a liquid is in contact with a solid, physicochemical reactions appear at the interface and lead to the formation of the electrical double layer (EDL). According to Stern's model (Fig. 1), the EDL is composed of two charge layers of opposite signs: one inside the solid and another inside the liquid. Within the liquid, two different regions are distinguished: one is located very close to wall where the electrical charges are not affected by the flow and is called the compact layer; the other one, called the diffuse layer, is composed of a space charge density (ρ) which decreases when one moves away from the wall according to the Boltzmann's law. For an infinite contact duration (τ_c) between the solid and the liquid, the EDL is fully developed. The space charge density at the interface (ρ_w) reaches a maximum value called ρ_{wd} . The diffuse layer's

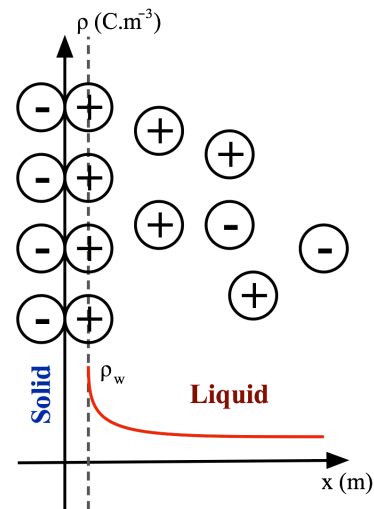


Fig. 1. Stern's model Electrical double layer.

thickness is often assimilated to the Debye length δ_0 [9]:

$$\delta_0 = \sqrt{\frac{\varepsilon_0 \varepsilon_L D_0}{\sigma_0}} \quad (1)$$

where ε_0 (8.854×10^{-12} F/m) and ε_L are the vacuum and the liquid's relative permittivity, respectively; D_0 is the mean ionic diffusion coefficient of the liquid charged species and σ_L is the liquid bulks electrical conductivity.

The flow electrification appears whenever the diffuse layer is swept by the flow; the current induced by the convection is called "streaming current" (I_{ss}). The flow causes a chemical imbalance of the electrical double layer that is compensated by the creation of new electrical charges within the liquid (Q_L) and opposite charges within the solid (Q_S), in the form of a wall current density, i_w (3). Thus, the electrical double layer develops progressively all throughout the duct. Most models of this phenomenon [10]–[12] assume that the global rate of the physicochemical reactions K_f and ρ_{wd} are intrinsic properties

Corresponding author: Paul Leblanc
e-mail address: paul.leblanc@univ-poitiers.fr

of the solid-liquid couple, constant and depend only of the physicochemical reactions.

$$i_w = K_f (\rho_{wd} - \rho_w) \quad (2)$$

In order to calculate the streaming current and the charge transported by the laminar flow (Poiseuille profile), we consider a capillary (cylindrical pipe) with a radius R and a length L . In this case, where we assumed that the space charge density at the entry of the capillary is null, Touchard [9], [13] gives expressions of the streaming current (I_{ss}) and the convected charge (Q_L) as a function of the mean flow velocity (U_m) and the contact duration ($\tau_c = L/U_m$) between the solid and the liquid as follows:

$$I_{ss}(\tau_c) = A\rho_{wd}U_m \left[1 - \exp(-BK_f\tau_c) \right] \quad (3)$$

$$|Q_L(\tau_c)| = \frac{A\rho_{wd}}{\pi R^2} \left[1 - \exp(-BK_f\tau_c) \right] \quad (4)$$

with A and B , two constants given by:

$$A = 8\pi\delta_0^2 \frac{I_2(R/\delta_0)}{I_0(R/\delta_0)}, \quad B = \frac{2\pi R}{A} \quad (5)$$

I_0 and I_2 are the modified Bessel functions of zero and second order, respectively. Both expressions are composed of two terms: the magnitude term controlled by w_d and the temporal term controlled by K_f . The amount of convected charge and generated charge is equal but of opposite sign ($|Q_S| = |Q_L|$).

For a contact duration sufficiently long to allow the EDL to fully develop, the temporal term may be neglected and the magnitude of streaming current becomes directly proportional to the mean flow velocity. In order words, the charge transported (Q_L) and the charge generated within the solid (Q_S) are constant. Thus, the parameter ρ_{wd} can be determined with the following equation:

$$\rho_{wd} = \frac{I_{ss}|_{\tau_c \rightarrow +\infty}}{AU_m} \quad (6)$$

In the same way, if the contact duration is kept constant, the streaming current (3) is also proportional to the mean flow velocity (Q_L and Q_S are constant). Regardless of the magnitude of the mean flow velocity's value or the length of the capillary, for a constant ratio L/U_m , the electrical double layer reaches the same development state. Using the parameter ρ_{wd} determined with the previous method (6), the parameter K_f can then be determined from the experimental streaming current values as follows:

$$K_f = -\frac{1}{B\tau_c} \ln \left(1 - \frac{I_{ss}}{A\rho_{wd}U_m} \right) \quad (7)$$

An experimental bench setup, developed in the laboratory, allows accurate measurements of streaming currents for dielectric liquids through stainless steel capillary.

III. EXPERIMENTAL SETUP

The experimental setup (Fig. 2) was developed a few years ago at Institut Pprime. It consists of a syringe pump (Fig. 2-1) on which is mounted a glass syringe (Fig. 2-2). A Nitrogen bottle (Fig. 2-3) allows compressing the oil in the bottle (5 L) to supply the syringe (Fig. 2-4). When the syringe is

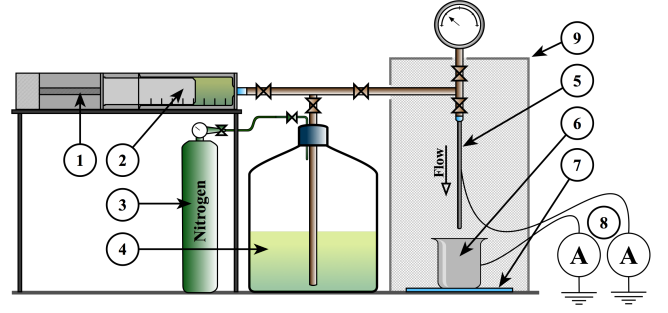


Fig. 2. Experimental setup 1: Syringe Pump, 2: Glass syringe, 3: Nitrogen bottle, 4: Oil bottle, 5: Stainless steel capillary, 6: Stainless steel beaker, 7: PTFE plate, 8: Pico-amperometers, 9: Faraday cage.

TABLE I
PROPERTIES OF THE OIL

Type	Mineral oil
σ_L (pS/m)	0.758
ε_L	2.143
D_0 (m ² /s)	2.5×10^{-11}
δ_0 (μ m)	25.01
ρ_m (kg/m ³)	870
ν (m ² /s)	14.93×10^{-6}

completely filled, the liquid is injected throughout the capillary (Fig. 2-5). The liquid is collected afterwards in a stainless steel beaker (Fig. 2-6). A polytetrafluoroethylene (PTFE) plate (Fig. 2-7) is placed under the beaker in order to electrically isolate it from the ground. The capillary and the beaker are placed inside a Faraday cage (Fig. 2-9). Two grounded pico-amperometers (Fig. 2-8), connected from the capillary and the beaker, allow measuring the electric charge transported by the liquid (I_s) and the opposite electric charge generated within the capillary (I_c).

To highlight the possible effects of the flow parameters on the global rate of the physicochemical reactions, the same capillary of 1.9 mm of radius and 360 mm of initial length is progressively shortened while keeping the same contact duration τ_c between the solid and the liquid. Indeed, it is important to ensure that the nature of the solid-liquid couple remains identical during the experimentation. Thus, the shorter the capillary, the lower the mean flow velocity to adjust τ_c . The liquid used, during the experimental campaign, is a mineral oil and the capillary is made of stainless steel. Table I summarizes the different properties of the oil.

The syringe pump allows controlling the mean flow velocity from 0.058 m/s to 1.76 m/s with an error of ± 0.002 m/s. A preliminary study was carried out and showed a good repeatability in the current's measurement; less than 1% of error upon the average value measured (based on 10 measurements).

The typical evolution of the streaming and capillary currents at $U_m = 1.23$ m/s is shown Fig. 3. As soon as the mineral oil is put into the motion, the streaming current increases and reaches its stationary state I_{ss} . This current is positive and in the order of pico-amperes; the charge generated within the oil is positive. The capillary current is opposite in sign; in the same way, it increases (in absolute value) and reaches

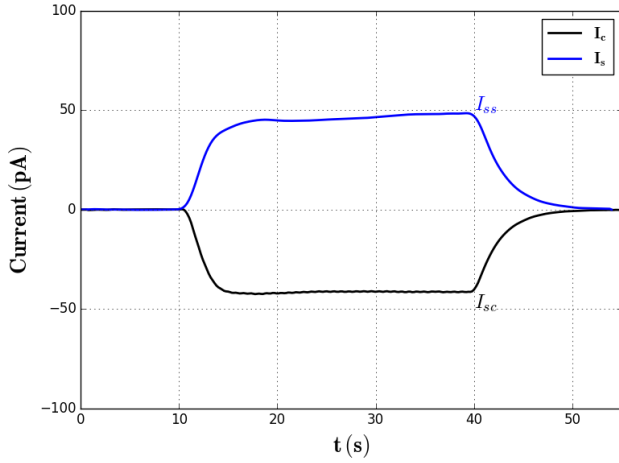


Fig. 3. Temporal evolution of the streaming and the capillary currents at $U_m=1.23$ m/s.

also a stationary state I_{sc} . Theoretically, both currents must be identical and lead to the same stationary state value ($|I_{ss}| = |I_{sc}|$).

However, we can observe slight differences. Firstly, in terms of magnitude, even if the oil system is grounded in order to avoid the creation of charge before the capillary, a small amount of charge within the oil is injected at the entrance of the capillary. Thus, the streaming current is slightly higher than the capillary current. Secondly, in terms of temporal evolution, a little time delay appears between both current. Indeed, the oil passes through the capillary at first before being collected by the beaker. The objective being to measure the charge generated throughout the capillary, the analysis is performed from the stationary capillary current I_{sc} .

IV. RESULTS

A. Determination of the space charge density at the interface for a fully developed electrical double layer

With the initial capillary length (longest length), at low velocity values, the stationary capillary current I_{sc} is measured as a function of the mean flow velocity, as shown in Fig. 4. The stationary current increases proportionally with the mean flow velocity. In this case, the contact duration seems to be long enough for allowing a full development of the EDL. In addition, a non-zero crossing of this curve is observed. This observation was also reported and analyzed in literature [8], [13], [14]. However, its origin is not clearly understood. In this context, the space charge density ρ_{wd} is determined from the slope α , in accordance with the experimental values, as follows:

$$\alpha = A\rho_{wd} \quad (8)$$

According to Eq. (8), the value of ρ_{wd} is calculated and is equal to 1.8 mC/m³.

B. Behavior of the global rate of the physicochemical reactions K_f

The charge generated within the capillary is presented in Fig. 5 as a function of the contact duration ($\tau_c = L/U_m$) for

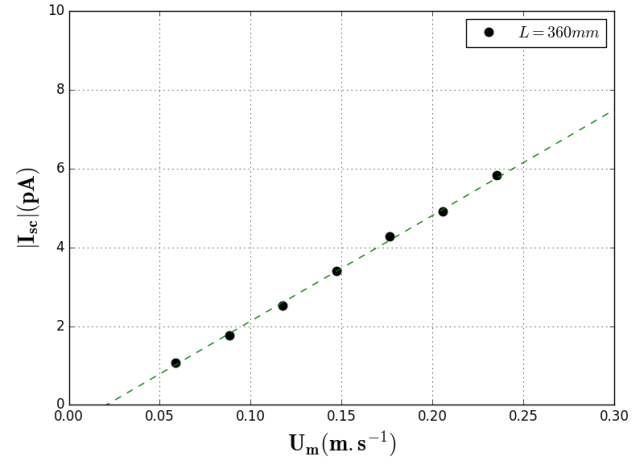


Fig. 4. Capillary current vs. mean flow velocity.

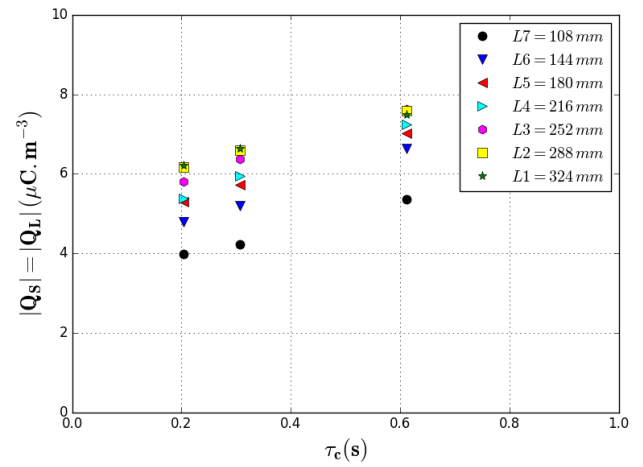


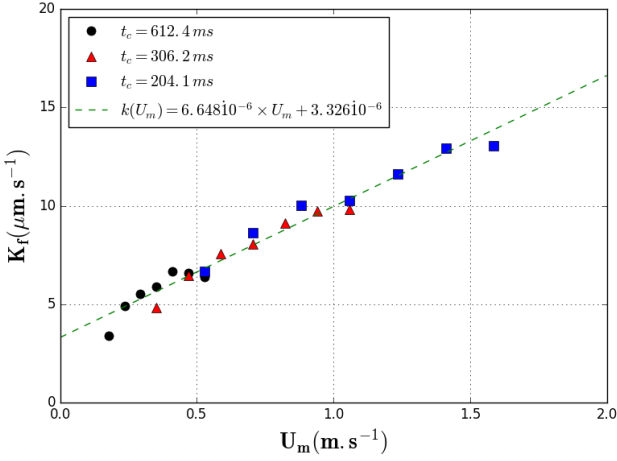
Fig. 5. Charge generated within capillary vs. contact duration.

different duct lengths. All the measurements were performed for three durations: 204.1, 306.2 and 612.4 ms. Regardless of the contact duration τ_c , the magnitude of Q_S is progressively reduced when the capillary is shortened. Contrary to the theoretical prediction, the charge transported is not constant. This rupture in the behavior between the theoretical model and the experimental measurement shows that the dynamics of the electrical double layer's development are also impacted by the mean flow velocity.

Therefore, this result suggests that the global rate of the physicochemical reactions also depends of the mean flow velocity. According to Eq. (4), the experimental value of K_f can be expressed as follows:

$$K_f(U_m) = -\frac{1}{B\tau_c} \ln \left(1 - \frac{\pi R^2}{A\rho_{wd}} |Q_S(L/U_m)| \right) \quad (9)$$

In accordance with Fig. 6, the global rate of the physicochemical reactions increases proportionally with the mean flow velocity. Regardless of the contact duration, its behavior remains the same. Thus, a rather good agreement can be found


 Fig. 6. Evolution of K_f vs. mean flow velocity.

with the following linear interpolation:

$$K_f(U_m) = 6.648 \times 10^{-6} U_m + 3.326 \times 10^{-6} \quad (10)$$

The y -intercept ($K_0 = 3.326 \times 10^{-6}$ m/s) seems to represent the global rate of the physicochemical reaction without flow.

V. DISCUSSION

In order to explain this behavior, Cabaleiro *et al.* [13] focused on the effect of the porous nature of a pressboard channel. Indeed, increasing the velocity leads to increase the effective contact surface. However, in the case of our metallic solid, this analysis appears to be insufficient to explain this phenomenon. Some authors [6]–[8] suggest that the variation of K_f is induced by the wall shearing stress, τ_w . In our case, the expression of τ_w can be expressed in a simple form:

$$\tau_w = \frac{4U_m \rho_m \nu}{R} \quad (11)$$

where ρ_m and ν are the mass density and the kinematic viscosity of the liquid, respectively. The wall shearing stress is directly proportional of the mean flow velocity. Recently, a study emphasized the impact of the wall shearing stress on the flow electrification process with different type of Newtonian and non-Newtonian liquids [15]. This work showed that, in the case of silicone oil and for a constant liquid conductivity, the evolution of the space charge density and the global rate of the chemical reactions are weakly impacted by the variation of the liquids viscosity. Furthermore, based on the streaming current measurements published through this work, we can replot I_{ss} as a function of τ_w . Fig. 7 brings up that when the wall shearing stress is kept constant, the streaming current is almost the same regardless of the initial viscosity and/or the magnitude of the mean flow velocity. Clearly, K_f seems to be strongly linked to the wall shearing stress imposed at the interface. Several scenarios can explain this behavior.

Firstly, we can consider that the wall shearing stress influences the different concentrations of the chemical species at the interface. Most models consider that there is no restriction for the chemical processes at the interface but this situation

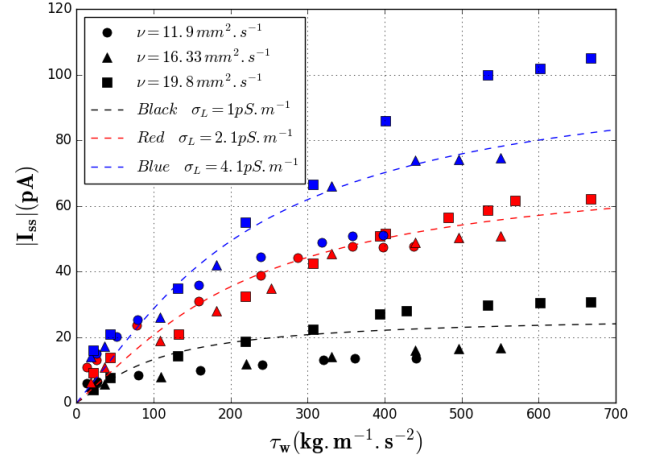


Fig. 7. Evolution of streaming current vs. wall shearing stress for the silicone oil at different viscosity and conductivity [15].

does not correspond to the reality. Indeed, even if the capillary is a metallic solid, we can assume that its interface is rough. Thus, increasing the wall shearing stress enhances slightly the penetration of oil within the roughness at the interface and enhances the chemical exchanges at the interface.

Secondly, it is quite possible that the local viscous dissipation Φ_w at the interface is high enough to allow any increase of the local temperature.

$$\Phi_w = \frac{\tau_w^2}{\rho_m \nu} \quad (12)$$

Assuming that the viscous dissipation is completely dissipated by conduction effect within the diffuse layer's volume, the local elevation temperature can be approached. This first estimation gives an elevation of the temperature of the order of one degree. Even if this variation appears to be low, it could be sufficient to impact the different electrical parameters, such as the local ionic mobility and the local conductivity. Thus, the physicochemical equilibrium of the electrical double layer can be impacted; hence modifying the global rate of chemical reactions.

VI. CONCLUSION

In this paper, the experimentation brings up that the process which governs the electrical double layers development cannot be reduced to a simple physicochemical reaction. It will be essential in further models to take into account the effect of the wall shearing stress on the flow electrification process and the possible variation of the local temperature.

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