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Impact of the laminar flow on the electrical double layer development

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ABSTRACT

As soon as a liquid comes into contact with a solid, physicochemical reactions occur at the solid-liquid interface to form an electrical double layer. According to the classical theory, it is assumed that the laminar flow has no impact on the global rate of the physicochemical reactions (K_f). However, recent studies have shown that this assumption is not consistent with the experimental results. It seems that K_f is impacted by the flow velocity of the liquid. The aim of this work is to compare the experimental behavior with the numerical simulation taking this variation into account.

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1. Introduction

The flow electrification phenomenon has been studied for a long time [1–3]. Basically, in the dielectric liquid, impurities are assumed to be responsible of the electrical double layer's (EDL) development at the solid-liquid interface. The interface becomes polarized with two opposite charge layers: one inside the liquid and the other within the solid. When a liquid flow is induced, part of the charge inside the liquid is swept, leading to the flow electrification phenomenon. Indeed, the chemical imbalance caused by the flow triggers the creation of new electrical charges within the dielectric liquid (Q_L) and opposite charges in the solid (Q_S). However, the physicochemical reactions at the origin of the EDL are still poorly understood. Most models proposing a better understanding of this phenomenon, such as the adsorption model [4], the corrosion model [5] or a combination of both [6], assume that the global rate of this physicochemical reaction K_f is independent of the flow parameters. However, recent studies [7–9] have shown that this assumption is not consistent with the experimental results.

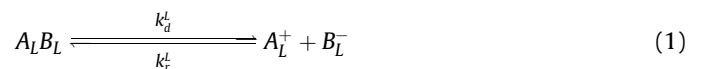
Using the finite volume CFD tool Code-Saturne[®] from EDF (Electricité de France), a module has been developed in order to simulate an adsorption model. The equations of this model [10] are coupled to the Navier-Stokes equation. This work presents a numerical analysis between both cases (K_f constant and variable)

based on the experimental results. The simulation was carried out using the variation of K_f as a function of the mean velocity observed during the experimental campaign on the flow electrification of mineral oil through a stainless steel duct [8].

2. Adsorption model

2.1. Scheme of the physicochemical adsorption model

The model assumes that the neutral impurities present within the liquid are dissociated in two ionic species: positive and negative. In the case where only one type of impurities reacts at the solid-liquid interface, the chemical reaction is given as follows:



where k_d^L and k_r^L are the kinetic constants of the dissociation/recombination reactions. Before the liquid comes into contact with the solid, the liquid and the solid are electrically neutral. The ionic species may react according to the adsorption model (Fig. 1).

Close to the interface, the solid medium is composed of many preferential adsorption sites. Thus, this model assumes that the negative ionic chemical species B_L^- are adsorbed at the solid surface by these sites, called C_S , to form the occupied site $C_S B_L^-$:

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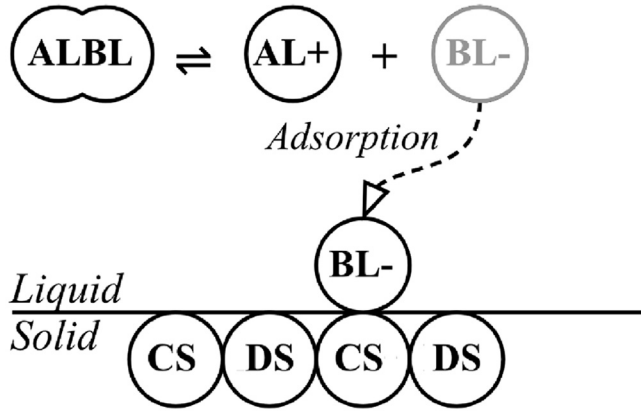


Fig. 1. Scheme of the physicochemical mechanism occurring at the solid-liquid interface for the adsorption hypothesis.

$$C_S + B_L^- \xrightleftharpoons[k_{rn}]{k_{fn}} C_S B_L^- \quad (2)$$

where k_{fn} and k_{rn} represent rates of negative adsorption and desorption, respectively. As in the majority of situations observed within experiments, in this model, the liquid is charged positively and the solid is negatively.

2.2. Electric charge transport mechanisms

All the chemical species, positive (P), negative (N) and neutral (O) are transported within the liquid by three mechanisms: diffusion (Diff.), migration (Mig.) and convection (Conv., when the liquid flow is induced). The flux density can be expressed with the charge species "i" in the liquid (L) as:

$$\vec{\Gamma}_i^L = \underbrace{-D_i^L \nabla n_i}_{Diff.} \pm n_i \underbrace{\frac{e_0 z_i D_i^L}{kT} \nabla \varphi}_{Mig.} + \underbrace{n_i \vec{u}}_{Conv.} \quad (3)$$

where, The general conservation equation can be expressed as follows:

$$\frac{\partial n_i}{\partial t} + \nabla \cdot \vec{\Gamma}_i^L = G_i - R_i \quad (4)$$

where G_i and R_i represent the generation and recombination species due to the chemical reactions. For example, the chemical reactions of B_L^- within the liquid can be expressed in more explicit forms:

$\vec{\Gamma}_i^L$	Ionic flux density vector ($m^2 \cdot s^{-1}$)
D_i^L	Diffusion coefficient for each species ($m^2 \cdot s^{-1}$)
n_i	Number density for each species ($A_L B_L, A_L^+, \dots$)
e_0	Elementary charge (C)
Z_i	Valence of the ion, taken as 1
k	Boltzmann's constant
T	Absolute temperature (K)
φ	Electrical potential (V)
\vec{u}	Velocity vector ($mg \cdot s^{-1}$)

$$G_{B_L^-} = k_d^L n_0^L \quad (5)$$

$$R_{B_L^-} = k_r^L n_p^L n_N^L \quad (6)$$

where n_0^L, n_p^L, n_N^L are the neutral, positive and negative concentrations within the liquid, respectively. At the interface, a normal ionic flux density is imposed for the species B_L^- (Γ_{wall}^-) to allow the adsorption and desorption reactions:

$$\Gamma_{wall}^- = \left[\overbrace{k_{fn} n_N^L n_p^S}_{Adsorption} - \overbrace{k_{rn} n_N^{SL}}_{Desorption} \right] \quad (7)$$

where n_p^S and n_N^{SL} are the surface concentrations of the free (C_S) and occupied ($C_S B_L^-$) adsorption sites, respectively. Finally, all these equations are coupled to the Poisson's equation:

$$\nabla \cdot (\epsilon_L \vec{E}) = e_0 \times (z_p n_p^L - z_N n_N^L) = \rho_L \quad (8)$$

where ϵ_L is the permittivity within the liquid, ρ_L is the space charge density. In the liquid region, the convection of the species are coupled to the Navier-Stokes equation. The simulation can be decomposed into two steps:

- **Static development of the EDL:** without fluid motion ($\vec{u} = 0$), the system reaches an equilibrium depending on different chemical kinetics.
- **Dynamic development of the EDL:** a flow is induced at the entry of the duct from the static equilibrium. The flow electrification process appears and the impact of the flow is studied.

2.3. Initial values and reaction rates of impurities

The initial concentration n_0 for the A_L^+ and B_L^- species in each region is estimated from the liquid conductivity as follows:

$$\sigma_0 = \frac{e_0^2}{kT} (D_p^L |z_p|^2 + D_N^L |z_N|^2) \times n_0 \quad (9)$$

Thus, the different chemical reactions of ionized impurities in the liquid can be determined considering that the space charge density is initially zero and the ionization fraction is equal to 0.1. The initial data values are summarized in Table 1:

Table 1 Physical constants and initial values of reaction rates.

Constant	Symbol	Value
Liquid conductivity	σ_0	$5 \times 10^{-12} S \cdot m^{-1}$
Liquid permittivity	ϵ_L	$2.2e_0$
Vacuum permittivity	ϵ_0	$8.85 \times 10^{-12} F \cdot m^{-1}$
Dynamic viscosity	ν	$0.0014 kg \cdot m^{-1} \cdot s^{-1}$
Liquid mass density	ρ_m	$836.9 kg \cdot m^{-3}$
Ionic valence	z	1
Elementary charge	e_0	$1.6 \times 10^{-19} C$
Ionic diffusion in liquid region	$D_{p,N,O}^L$	$2.5 \times 10^{-11} m^2 \cdot s^{-1}$
Initial concentration (A_L^+, B_L^-)	n_0	$1.58 \times 10^{16} m^{-3}$
Initial neutral ions concentration	n_{oi}	$1.44 \times 10^{18} m^{-3}$
Recombination rate in the liquid	k_d^L	$1.65 \times 10^{-17} m^3 \cdot s^{-1}$
Dissociation rate in the liquid	k_r^L	$2.85 \times 10^{-3} s^{-1}$

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