Flow electrification of liquids in rectangular channels - Comparison of different theoretical models

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Abstract— **This paper deals with flow electrification phenomenon of liquids in channels of rectangular cross section. Different theoretical models are described and compared. For all the models it is assumed that the flow and the diffuse layer are fully developed. The space charge density conveyed by the flow is computed. First, two cases are examined in the case of weak space charge density, the exact rectangular channel solution is compared with the approximate solution of two parallel planes. This comparison shows a rather small difference between the two models. Then, in the case of two parallel planes assumption, the charge conveyed is computed without any hypothesis on the magnitude of the space charge density and compared to the solution obtained for a weak space charge density commonly assumed. This comparison shows a big difference between the two models concerning the determination of the space charge density on the wall and therefore the zeta potential [1].**

Keywords: **Flow electrification of liquids, Diffuse layer, Theoretical models, Rectangular channels, Zeta potential**

I. INTRODUCTION

The phenomenon of flow electrification of liquids has been investigated for more than fifty years now [2-34]. Thus, the influence of different parameters on the flow electrification phenomenon of liquids is now well understood. Indeed, we now know, in the case of flow electrification in a pipe, the effect of the flow regime (specially the jump from laminar to turbulent), as well, how the magnitude of the charge transported varies with the Reynolds number of the flow and with the radius or the roughness of the pipe. The physicochemical process at the origin of the phenomenon has also been widely investigated but unfortunately it remains badly understood and is still under investigation. Moreover, very few investigations have been made on the influence of the shape of the channel and often an approximate solution available only for parallel planes and for the case of weak space charge density is employed. But, with the increasing of micro fluidic applications in various fields, it seems to be important to see the deviations between this approximate solution with the exact one or at least with a more realistic one. Thus, the purpose of this pa-

per is to compare the space charge density predicted near the wall of the duct using different models for the flow and the space charge density profiles. The results obtained are only available in the case of a fully developed laminar flow and, as well, a fully developed diffuse layer.

II. IMPORTANT PARAMETERS

Apart from the hydrodynamic parameters of the flow and the geometrical parameters of the tube or channel, two important parameters control the flow electrification phenomenon. One is the Debye length: $\delta_0 = \sqrt{\dfrac{\varepsilon D_0}{\sigma}}$, where ε is the permittivity of the liquid, D_0 is the mean diffusion coefficient of the ions (or dissociate impurities) and σ the liquid electrical conductivity, this parameter determines the non dimensional diameter of the duct. The other important parameter relates the intensity of ions exchange at the interface solid/liquid, we call it the space charge density on the wall: ρ_w . Indeed, in a liquid inside a duct, whatever the liquid and the material of the duct, an electrical double layer exists. Inside the liquid, the layer is in fact composed of two different layers, the compact layer close to the wall of the duct and the diffuse layer. The compact layer is so thin that it is not affected by the flow, thus, it is not taken into account in the phenomenon of flow electrification. The diffuse layer is generally characterize by the evolution of its space charge density. It is a volumetric quantity varying from the compact layer to the center of the duct. Close to the compact layer we usually call it the space charge density on the wall ρ_w , because the compact layer does not play a role in the flow electrification phenomenon. This parameter cannot be directly measured. It is generally obtained from experimental measurements of flow electrification. Indeed, we measure the streaming current at the exit of the duct for a fully developed laminar flow, and a totally developed diffuse layer. Then, from the streaming current and the flow rate we obtain the space charge density in the liquid which has been transported by the flow.

From the experimental value of the space charge density transported and depending on the model chosen for the velocity profile and the space charge density profile, we can computed the space charge density on the interface ρ_w corresponding to each model used.

Moreover, in all this study we consider the case of ions with the same diffusion coefficient D_0 and the same valence Z.

One model for the space charge density profile is the so called "weak space charge density model" [6]. The space charge density in the diffuse layer can be considered as "weak" when the difference between the number of positive and negative ions is very small compared to the total number of positive and negative ions. In that case the electrical quantities will verify the following equation.

$$\rho << \frac{kT\sigma}{e_0 Z D_0} \tag{1}$$

ρ being the space charge density, k the Boltzman coefficient, T the absolute temperature and e_0 .the elementary charge.

In all this study when we assume the hypothesis of weak space charge density it means that we assumed that equation (1) is available in the whole diffuse layer.

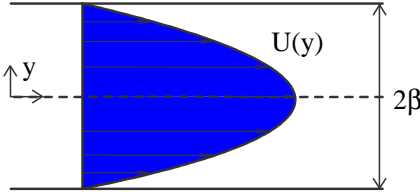## III. Two parallel planes model with the weak space charge density assumption
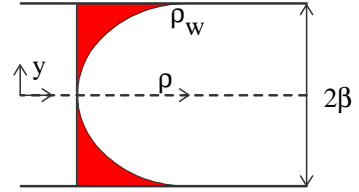

Fig. 1 Velocity profile   Fig2. Space charge density profile

The velocity is given by:

$$U(y) = \frac{3}{2} U_m \left(1 - \frac{y^2}{\beta^2} \right) \tag{2}$$

U_m being the mean velocity and β the half width of the duct. With the assumption of weak space charge density, the space charge density profile is given by [2]:

$$\rho(y) = \rho_w \frac{\cosh(y/\delta_0)}{\cosh(\beta/\delta_0)} \tag{3}$$

The streaming current and the flow rate (per unit width) can be then calculated:

$$\int_{-\beta/\delta_0}^{+\beta/\delta_0} \rho(y)\ U(y) dy \qquad \int_{-\beta/\delta_0}^{+\beta/\delta_0} U(y) dy \tag{4},(5)$$

Finally, the space charge density in the liquid which has been transported by the flow is the ratio of the streaming current to the flow rate:

$$Q = \frac{\displaystyle\int_{-\beta/\delta_0}^{+\beta/\delta_0} \rho(y)\ U(y) dy}{\displaystyle\int_{-\beta/\delta_0}^{+\beta/\delta_0} U(y) dy} \tag{6}$$

Referring to the expression of $\rho(y)$ in equation (3) we will call this value Q_{ch}

After some calculus we find:

$$Q_{ch} = \frac{3\rho_w}{(\beta/\delta_0)^2}\left[1 - \frac{\tanh(\beta/\delta_0)}{(\beta/\delta_0)}\right] \tag{7}$$

Thus, measuring the streaming current at the exit of the duct and knowing the flow rate we obtain the space charge density in the liquid which has been transported by the flow (equation (6)), we call it the space charge density transported Q_{exp}. Then, knowing Q_{exp} from experiment we compute (with the help of equation (7)) the value of ρ_w. We call it ρ_{wpp} because it is obtained with the parallel planes model:

$$\rho_{wpp} = \frac{Q_{exp}\,(\beta/\delta_0)^2}{3\left[1 - \dfrac{\tanh(\beta/\delta_0)}{(\beta/\delta_0)}\right]} \tag{8}$$

The expression given in equation (8) is the most commonly used because of its simplicity but it is based on two important assumptions rarely satisfied. The first one is the hypothesis of two parallel planes instead of a rectangular channel, this hypothesis could be realistic if and only if one size of the rectangular channel section is much greater than the other one. The second one is the hypothesis of weak space charge density, this is the case if the non dimensional space charge density in the whole diffuse layer is much smaller than 1 (equation (1)). Unfortunately we have often found in many experiments that it is not the case [12].

Following, still assuming the case of weak space charge density, we are going to compare the previous expression with one obtained in a duct of rectangular cross section.

IV. CHARGE TRANSPORTED IN A RECTANGULAR CHANNEL WITH THE WEAK SPACE CHARGE DENSITY ASSUMPTION
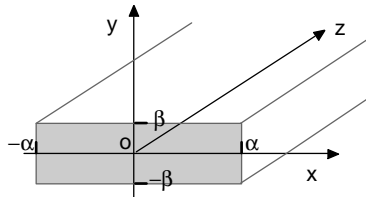
A. *Laminar flow in a rectangular channel*

Fig.3 Rectangular channel

We consider a rectangular channel with an axis in the z direction. The size is 2α in the x direction and 2β in the y direction. The Navier-Stokes equations for a laminar unidirectional flow of a non compressible fluid inside a tube of any kind of cross section can be written:

$$\begin{cases} \Delta U = \dfrac{1}{\mu}\dfrac{dP}{dz} \\[2mm] \dfrac{\partial U}{\partial z} = 0 \end{cases} \tag{9}$$

U being the velocity in z direction, μ the dynamic viscosity and P the pressure.

On the limit of the domain (wall channel) we have Dirichlet conditions and the solution can be obtained with Green functions. Several different solutions exist.

1) First solution with cosines and hyperbolic cosines series [35]

$$U = -\frac{16\alpha^2}{\pi^3}\frac{1}{\mu}\frac{dP}{dz}\sum_{n=1}^{\infty}(-1)^n\left[1-\frac{\cosh\left(\dfrac{(2n-1)\pi y}{2\alpha}\right)}{\cosh\left(\dfrac{(2n-1)\pi\beta}{2\alpha}\right)}\right]\left[\frac{\cos\left(\dfrac{(2n-1)\pi x}{2\alpha}\right)}{(2n-1)^3}\right] \tag{10}$$

2) Second solution with cosines and hyperbolic cosines series [36]

$$U = -\frac{1}{2\mu}\frac{dP}{dz}\left[\beta^2 - y^2 + \frac{4}{\beta}\sum_{n=1}^{\infty}\frac{(-1)^n}{\left(\dfrac{\pi(2n-1)}{2\beta}\right)^3}\frac{\cosh\left(\dfrac{\pi(2n-1)x}{2\beta}\right)\cos\left(\dfrac{\pi(2n-1)y}{2\beta}\right)}{\cosh\left(\dfrac{\pi(2n-1)\alpha}{2\beta}\right)}\right] \tag{11}$$

3) Solution with sinus series [37]

$$U = -\frac{1}{\mu}\frac{dP}{dz}\sum_{k=0}^{\infty}\sum_{l=0}^{\infty}16\frac{\sin\left(\dfrac{(2k+1)\pi(x+\alpha)}{2\alpha}\right)\sin\left(\dfrac{(2l+1)\pi(y+\beta)}{2\beta}\right)}{\left[(2k+1)(2l+1)\pi^2\right]\left[\dfrac{(2k+1)^2\pi^2}{4\alpha^2}+\dfrac{(2l+1)^2\pi^2}{4\beta^2}\right]} \tag{12}$$

This solution is available inside the domain: $x \in \left]-\alpha,+\alpha\right[$ and $y \in \left]-\beta,+\beta\right[$, but not on the wall ($x = \pm\alpha$ or $y = \pm\beta$) for which U=0.

This solution is not so rapidly convergent than the previous ones, we used 1000 terms for this one, which gives a satisfactory answer.

Even if this solution is not so rapidly convergent than the solutions using hyperbolic cosines we use it for two reasons. The first one is the strong increasing of the hyperbolic cosine with the argument of the function which leads rapidly to underflows or overflows during the computation however this difficulty can be corrected using approximation. The second and real reason is the similarity of the sinus solution with the solution existing for the space charge density.

The mean velocity for the solution with sinus is given by:

$$U_{moy} = -\frac{1}{\mu}\frac{dP}{dz}\sum_{k=0}^{\infty}\sum_{l=0}^{\infty}\frac{64}{\left[(2k+1)^2(2l+1)^2\pi^4\right]\left[\frac{(2k+1)^2\pi^2}{4\alpha^2}+\frac{(2l+1)^2\pi^2}{4\beta^2}\right]} \qquad (13)$$

The maximum velocity which is for x=0 and y=0 is given by:

$$U_{max} = -\frac{1}{\mu}\frac{dP}{dz}\sum_{k=0}^{\infty}\sum_{l=0}^{\infty}\frac{16(-1)^k(-1)^l}{\left[(2k+1)(2l+1)\pi^2\right]\left[\frac{(2k+1)^2\pi^2}{4\alpha^2}+\frac{(2l+1)^2\pi^2}{4\beta^2}\right]} \qquad (14)$$

Finally, the flow rate is given by:

$$F = -\frac{1}{\mu}\frac{dP}{dz}\sum_{k=0}^{\infty}\sum_{l=0}^{\infty}\frac{256\alpha\beta}{\left[(2k+1)^2(2l+1)^2\pi^4\right]\left[\frac{(2k+1)^2\pi^2}{4\alpha^2}+\frac{(2l+1)^2\pi^2}{4\beta^2}\right]} \qquad (15)$$

As an example, we can see in Fig.4 the velocity profile in the case of a channel of square cross section ($\alpha = \beta$). The three solutions give the same results.
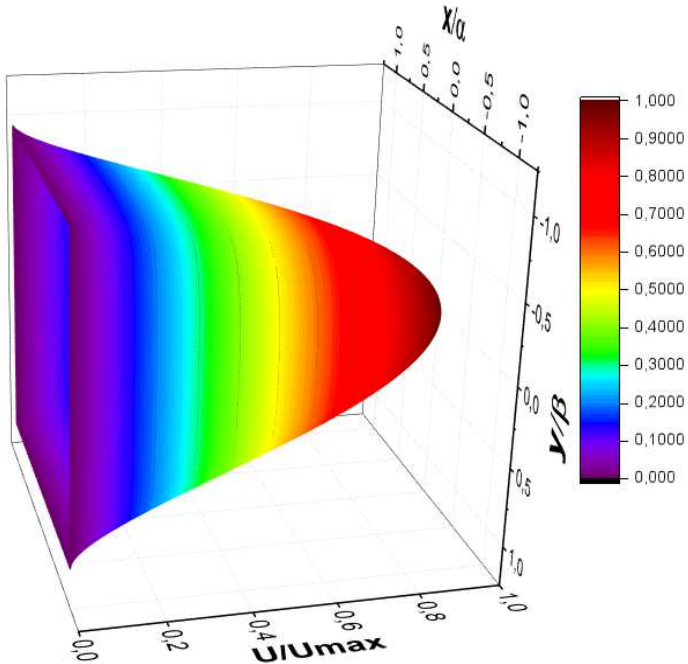

Fig.4. Velocity profile in a square cross section channel

B. Space charge density in a rectangular channel

With the assumption of weak space charge density and taking appropriate reference quantities we get the following system of non dimensional equations [6]:

$$\begin{cases} \overrightarrow{grad}_+ \rho_+ + \gamma^2 \, \overrightarrow{grad}_+ \varphi_+ = 0 \\ \Delta_+ \varphi_+ = -\rho_+ \end{cases} \tag{16}$$

ρ_+ being the non dimensional space charge density, φ_+ the non dimensional potential and γ is a coefficient taking into account the diffusion coefficient of ions:

$$\gamma = \frac{2\sqrt{D_P D_N}}{(D_P + D_N)} \tag{17}$$

D_P and D_N being respectively the diffusion coefficient for positive and negative ions. In practice γ is always close to 1. The previous system can be written as follows:

$$\overrightarrow{grad}_+ \left[-\Delta_+ \varphi_+ + \gamma^2 \varphi_+ \right] = 0 \tag{18}$$

Which is equivalent to:

$$\left[-\Delta_+ \varphi_+ + \gamma^2 \varphi_+ \right] = C \tag{19}$$

C being an integration constant.

We will take C equal to the non dimensional space charge density on the wall of the channel:

$$\left[-\Delta_+ \varphi_+ + \gamma^2 \varphi_+ \right] = \rho_{w+} \tag{20}$$

This implies $\varphi = 0$ on the wall of the channel

The solution of this equation is, again, obtained with the help of Green function.

$$\rho_+(x_+, y_+) = \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} 16 \rho_{w+} \frac{\left[\frac{(2k+1)^2 \pi^2}{4\alpha_+^2} + \frac{(2l+1)^2 \pi^2}{4\beta_+^2} \right] \sin\left(\frac{(2k+1)\pi(x_+ + \alpha_+)}{2\alpha_+} \right) \sin\left(\frac{(2l+1)\pi(y_+ + \beta_+)}{2\beta_+} \right)}{\left[(2k+1)(2l+1)\pi^2 \right] \left[\frac{(2k+1)^2 \pi^2}{4\alpha_+^2} + \frac{(2l+1)^2 \pi^2}{4\beta_+^2} + \gamma^2 \right]} \tag{21}$$

This solution is available on the open intervals:

$$x_+ \in \left] -\alpha_+, +\alpha_+ \right[\quad \text{and} \quad y_+ \in \left] -\beta_+, +\beta_+ \right[$$

On the wall, i.e. $x_+ = \pm\alpha_+$ or $y_+ = \pm\beta_+$, the solution is: $\rho_+ = \rho_{w+}$

In dimensional form we have:

$$\rho(x,y) = \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} 16\rho_w \frac{\left[\dfrac{(2k+1)^2\pi^2}{4\alpha^2} + \dfrac{(2l+1)^2\pi^2}{4\beta^2}\right]\sin\!\left(\dfrac{(2k+1)\pi(x+\alpha)}{2\alpha}\right)\sin\!\left(\dfrac{(2l+1)\pi(y+\beta)}{2\beta}\right)}{\left[(2k+1)(2l+1)\pi^2\right]\left[\dfrac{(2k+1)^2\pi^2}{4\alpha^2} + \dfrac{(2l+1)^2\pi^2}{4\beta^2} + \dfrac{\gamma^2}{\delta_0^2}\right]} \quad (22)$$

As an example, we can see in Fig.5 the space charge density profile in the case of a channel of square cross section ($\alpha = \beta$) and for $\delta_0 = \alpha = \beta$ and $\gamma = 1$.
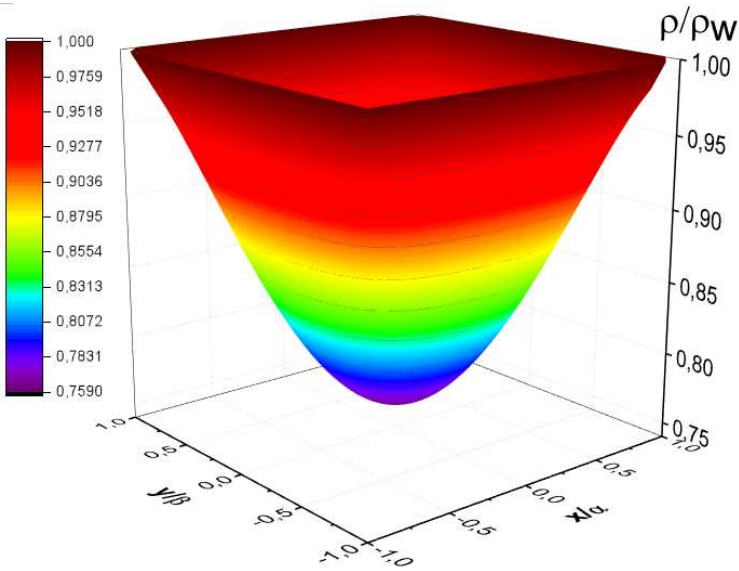

Fig.5. Space charge density profile in a square cross section channel for weak space charge assumption

C. Space charge density transported by a laminar flow in a rectangular channel

Knowing the evolution of the velocity and the space charge density in the case of a rectangular channel we compute equation (6), then we determine the space charge density on the wall from the experimental value of the space charge density transported. We will call it ρ_{wrect} because it is the case of channel of rectangular cross section. We can see in Fig. 6 the ratio of the value obtained with the assumption of parallel planes to the value obtained with the rectangular cross section channel solution. This ratio is computed for 10 different values of α/β varying from 1 to 10 and 100 values of β/δ_0 varying between 1 to 100. We can see that when α/β is large the results obtained with the two models are quite close, this is normal because, for such cases, the cross section is more like parallel planes. On the other hand, for a square cross section of the duct the ratio increases with

β / δ_0 up to around 2. This ratio of 2 can be explained by the double surface in contact with the liquid than in the case of two parallel planes models. Any way, this difference remains rather small. Thus, in the case of weak space charge density, assuming a rectangular channel as two parallel planes seems reasonable.
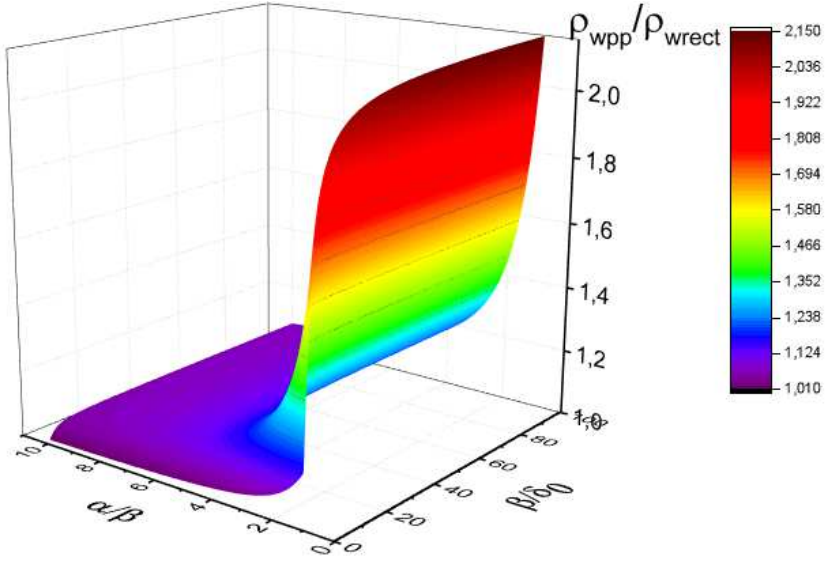

Fig.6. Comparison between the paralell plane and rectangulal model for weak space charge assumption

V. TWO PARALLEL PLANES MODEL WITHOUT WEAK SPACE CHARGE ASSUMPTION

Now, we are going to compute the space charge density transported by a laminar flow through parallel plates for which β / δ_0 is large enough in order to consider that the expression of the space charge density on each plate is the same that the one existing on a single plate (in a semi infinite domain). The space charge density on a single plate without any assumption on the magnitude of the space charge density has already been computed [38]. For this computation we take the origin on the plate ($y = \beta$ corresponds to the median plane).

$$\rho = 2\,\rho_{ref}\,\frac{\cosh(y_+ + C)}{\sinh^2(y_+ + C)} \qquad \text{with: } C = \text{ArgCosh}\left[\frac{1}{\rho_{w+}} + \sqrt{1 + \frac{1}{\rho_{w+}^2}}\right] \qquad (23)$$

ρ_{ref} being the space charge density of reference ($\rho_+ = \rho / \rho_{ref}$)

With y=0 on the plate the velocity is given by:

$$U = \frac{3}{2} U_m \left(\frac{2y}{\beta} - \frac{y^2}{\beta^2} \right) \tag{24}$$

U_m being the mean velocity. Finally, the non dimensional space charge density transported by the flow is:

$$Q_+ = \frac{3}{\beta_+} \int_0^{\beta_+} \frac{\cosh(y_+ + C)}{\sinh^2(y_+ + C)} \left(\frac{2y_+}{\beta_+} - \frac{y_+^2}{\beta_+^2} \right) dy_+ \tag{25}$$

We will call this value Q_{real+} as it is more close to the real value than the expression given by the weak space charge assumption.
Integration gives:

$$Q_{real+} = \frac{6}{\beta_+} \left[\frac{-1}{\sinh(\beta_+ + C)} + \frac{1}{\beta_+} \left(\ln\left(\tanh\left(\frac{\beta_+ + C}{2} \right) \right) - \ln\left(\tanh\left(\frac{C}{2} \right) \right) \right) \right] +$$
$$\frac{3}{\beta_+ \sinh(\beta_+ + C)} - \frac{6}{\beta_+^2} \ln\left(\tanh\left(\frac{\beta_+ + C}{2} \right) \right) +$$
$$\frac{6}{\beta_+^3} \left[\ln\left(\tanh\left(\frac{\beta_+ + C}{2} \right) \right) \ln\left(1 + \tanh\left(\frac{\beta_+ + C}{2} \right) \right) + li2\left(1 - \tanh\left(\frac{\beta_+ + C}{2} \right) \right) + li2\left(- \tanh\left(\frac{\beta_+ + C}{2} \right) \right) \right] \tag{26}$$

In this equation li2 is the dilogarithm function. The value of ρ_w obtained with this model must be compared to the value obtained with the weak space charge density assumption:

$$Q_{ch+} = \frac{3\rho_{w+}}{(\beta_+)^2} \left[1 - \frac{\tanh(\beta_+)}{(\beta_+)} \right] \tag{27}$$

Practically, knowing the space charge density in the liquid transported by the flow, we have to compute its non dimensional value and determine the space charge densities on the wall which gives this value with the two different models. Then, it is possible to compare the two values obtained.
We can see in Fig. 7 the comparison between the value of the space charge density on the wall obtained with the assumption of weak space charge density (ρ_{wch}) and without

this assumption (ρ_{wreal}). For this comparison the half distance between the two plates (β) has been fixed ten times greater than the diffuse layer thickness (δ_0).

When the non dimensional space charge density on the wall is small compared to one ($\rho_+ \ll 1$ in the whole section), the two different models gives the same result. Obviously, we are in the case of weak space charge density. By cons, when the space charge density on the wall is large the model of weak space charge density gives a wrong value of ρ_w, much smaller than the reality.
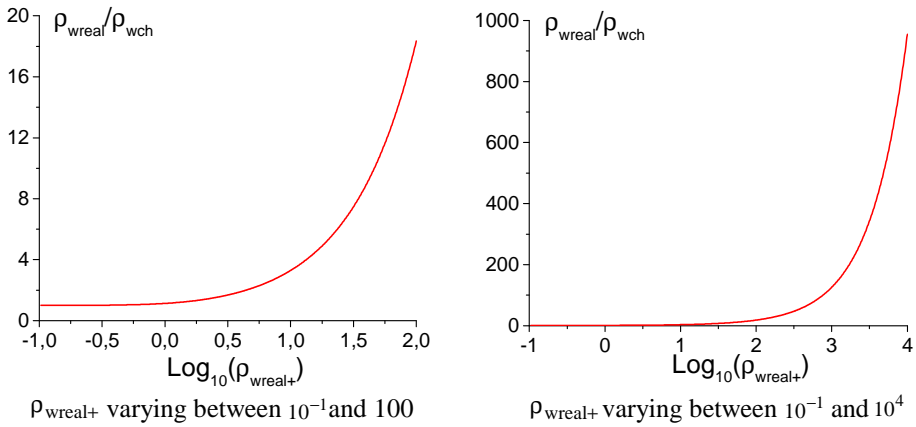

Fig.7. Comparison between the models with and without the assumption of weak space charge density

This large difference between the simple model (weak space charge density assumption) and the more realistic one is physically important as it leads to a wrong interpretation of the intensity of the wall reaction and will give a wrong value of the zeta potential deduced from flow electrification experiments.

This important point needs some more explanations. How weak space charge density assumption underestimates the space charge density magnitude near the wall? With the weak space charge assumption the profile of the space charge density in the diffuse layer is given by equation (3), thus an hyperbolic cosine function, while the real profile must follow a function much more rapidly growing near the wall. Thus, for a given experimental value of the charge transported by the flow if we use the weak space charge assumption, when it is not appropriate, we will obtain a smaller value of the space charge on the wall than that really existing and a smaller value of the zeta potential. In practice this case exists very often. For instance, for dielectric liquid the non dimensional space charge density on the wall computed from flow electrification experiment can be up to 7500 [12] depending on the material and the liquid, this means that the real value of the space charge on the wall and the zeta potential is **much more important** than what is predicted with the weak space charge model. This is always the case with dielectric liquids but also

with water for which the non dimensional space charge density on the wall is often in the range of 10 to 20.

Of course, the expression of the charge transported is much more simple with the weak space charge assumption and computing the space charge density on the wall from an experimental value of the charge transported by the flow using this expression is also much more simple. However, without this assumption an analytical solution also exists thus, even if it is a little more complex it can gives the real values of the space charge on the wall and the zeta potential instead of wrong values.

VI. CONCLUSION

This paper concerns flow electrification in rectangular channels. We know that the space charge density on the wall and the zeta potential is obtained from experiments of charge convection. Thus, to obtain the space charge density on the wall we can use different models. We made a comparison between the simple model, commonly used, making the assumption of weak space charge density and considering that the rectangular channel can be assumed identical to two parallel plates. Firstly, we compared, in the case of weak space charge density, the prediction with a rectangular model and that given for two parallel plates; we found that the more simple model (parallel plates) give results not so different than those obtained with channel of rectangular cross section. Secondly, for $\beta \geq 10\,\delta_0$, we compared, in the case for which the hypothesis of parallel plane can be assumed ($\alpha > 10\,\beta$), the results predicted with and without the assumption of weak space charge density. We found that the assumption of weak space charge density leads to space charge on the wall sometimes much smaller than without this assumption, assumption, thus, this assumption must not be used because it is not realistic.

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