STABILITY OF MICROCHANNEL-FLOW UNDER THE ELECTRIC DOUBLE LAYER EFFECT

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ABSTRACT

The effect of the electric double layer (EDL) on the linear stability of Poiseuille planar channel flow is reported. It is shown that the EDL destabilises the linear modes, and that the critical Reynolds number decreases significantly when the thickness of the double layer becomes comparable with the height of the channel. The planar macro scale Poiseuille flow is metastable, and the inflexional EDL instability may further decrease the macro-transitional Reynolds number. There is a good correspondence between the estimated transitional Reynolds numbers and some experiments, showing that early transition is plausible in microchannels under some conditions.

INTRODUCTION

The characteristics of gas flows in microchannels can adequately be modelled by slip velocity and temperature jump related to first or second order models as a function of the Knudsen number, at least in the rarefied regime. Such molecular effects are difficult to model in micro-liquid flows. Yet, estimations obtained from some molecular models indicate that the discontinuous boundary conditions could only hold for microchannels of hydraulic diameters smaller than few microns (Gad-el-Hak, 1999, Tardu, 2003). The interfacial effects (wall/liquid) are presumably mostly responsible for the deviations observed from the classical macro-theory, in larger microchannels. One of the micro-effects that may play an important role is the electric double layer (EDL) at the solid/liquid interface. The electrostatic charges present on the solid surface attract the counterions to establish an electrical field.

In the compact layer next to the wall the ions are immobile. In the diffuse EDL layer however, the ions are less affected by the electrical field and can move. Under the effect of an imposed pressure gradient, the accumulation of the mobile ions downstream sets up an electrical field that induces a streamwise external force. In macroscale flows, these effects are negligible, as well as the thickness of the EDL is very small compared to the height of the channel. In micro-flows, in return, the EDL play a rather significant role. Well-controlled recent experiments have clearly confirmed that it can explain the behaviour of the Poiseuille number in laminar regime, providing that the liquid contains a very small amount of ions (Ren et al. 2001). Kulinsky et al. (1999) reported an increase of 70% of the friction factor under the EDL effect in planar channels of 4 μm heights with distilled water. Large thickness of the diffuse EDL layer of about 1 μm or more have been reported in these experiments.

There is a curious phenomena encountered in some experiments showing that there is an early transition in microchannel flows. Weng and Peng (1994) noticed that the transition occurred at $Re<150$ in microchannels 0.2-0.8 mm wide and 0.7 mm deep, which is significantly smaller than 400 corresponding to macro scale flows (The Reynolds number through this paper is based on the centreline velocity and half height of the channel. Thus 400 correspond to a Reynolds number based on hydraulic diameter and cross section average velocity of 2000). A transitional number of about 250 has also been reported in the experiments conducted in 1mm wide trapezoidal microchannels with channel depths ranging between 79 and 325 μm (Rahman and Gui, 1993 a and b, Gui and Scaringe 1995). The micro tubes experiments of Mala and Li (1999) indicate that there is an early transition from laminar to turbulent flows for $Re>56\text{-}170$. These effects may be attributed to the roughness that may both influence the transition in the entry region, or directly affect the flow behaviour through an implied roughness-viscosity (Weilin et al., 2000). Yet, direct interfacial effects may also play a role in presumed early transition.

The stability mechanism in planar channel flows is mainly non-linear and the secondary instabilities cause the flow to bifurcate before the critical Reynolds number of linear modes. That depends on the behaviour of the time space development of the perturbations near the critical Re and wave numbers. The main aim of this study is to investigate directly the EDL effect on the linear stability of planar channel flow, and estimate indirectly the resulting transitional Reynolds numbers. It is asked whether electrokinetic effects on liquid flows may cause the early transition or not and whether the linear stability EDL flow characteristics may explain the small transitional Reynolds numbers encountered in studies quoted to before.

PHYSICAL ASPECTS OF ELECTRIC DOUBLE LAYER

Most solid surfaces have an electric surface potential when brought with an electrolyte. The most common mechanism for the charging of surface layers in microfluidics is the depronotonation of surface groups on surfaces such as silica, glass, acrylic and polyester (Hunter, 1981, Sharp et al., 2002). The electrostatic charges present on the solid surface attract the counterions to establish an electrical field . In the compact layer next to the wall and less than 1 nm thick the ions are immobile. In the diffuse EDL layer however, the ions are less affected by the electrical field and can move. The counterion concentration near the wall is larger than in the bulk of the fluid. That results in a net charge density in a unit volume resulting from the concentration difference between cations and anions, according to the Boltzmann equation. The electrostatic potential at any point near the surface, provided that it is small compared to the energy of ions, may be obtained by a linear approximation of the Poisson-Boltzmann equation. Its value at the wall can be related to the Zeta potential between the compact layer and diffuse layer, when the EDLs near the opposite walls do not overlap. The Zeta potential is a property of the solid-liquid pair and can be determined experimentally. The imposed pressure gradient accumulates the mobile ions downstream and sets up an electrical field whose potential is called the streaming or electrokinetic potential. The streaming potential and the net charge density induces a streamwise external force. In the steady state, the streaming current due to the transport of charges is in equilibrium with the conduction current in the opposite direction. That allows the determination of the streaming potential and of the velocity profiles under the EDL effect.

EQUATIONS GOVERNING 2D CHANNEL FLOW UNDER THE EDL EFFECT

We suppose constant properties (viscosity and permittivity). The effects of finite ion size and gradients of the dielectric strength and of the viscosity are neglected. These hypotheses are not contradictory with the fact that we mainly deal with very dilute solutions for which the equilibrium Boltzmann distribution is applicable. The velocity profile under the electrokinetic EDL effect obtained by Mala et al (1997) can be put in non-dimensional form as:

$$u=1-y^2 - 4\frac{I_1-I_2}{\dfrac{\kappa^2 \sinh\kappa}{\bar{\zeta}^2 G}+4\left(I_3-\dfrac{I_4}{\sinh\kappa}\right)}\left\{1-\left|\frac{\sinh\kappa y}{\sinh\kappa}\right|\right\} \quad (1)$$

where, the scaling velocity is the centerline velocity of the Poiseuille component, i.e. $-\frac{a^2\, dp/dx}{2\mu}$ and the scaling length is the half channel height a. There are several parameters in this equation, for instance $G=\frac{(n_0 zea)^2}{\lambda_0 \mu}$, with n_0 standing for the ionic number concentration, z for the valence of positive or negative ions, e for the electron charge λ_0 the electric conductivity of the fluid, and μ for its dynamic viscosity. One of the most important quantities involving in (1) is the non dimensional Debye-Huckle parameter

$$\kappa=ak=a\left(2n_0e^2/\varepsilon\varepsilon_0 k_b T\right)^{1/2} \quad (2)$$

with ε and ε_0 being respectively the dielectric constant of the medium and the permittivity of vacuum, k_b the Boltzmann constant and T the absolute temperature. The characteristic EDL thickness is $1/k$. The non-dimensional Zeta potential reads for $\bar{\zeta}=\frac{ze\zeta}{k_b T}$. The quantities I in (1) are given by:

$$I_1=I_3=\frac{\cosh\kappa-1}{\kappa},\ I_4=\frac{\sinh\kappa\cosh\kappa}{2\kappa}-\frac{1}{2}$$
$$I_2=\left(\frac{1}{\kappa}+\frac{2}{\kappa^3}\right)\cosh\kappa-\frac{2}{\kappa^2}\sinh\kappa-\frac{2}{\kappa^3}, \quad (3)$$

It is seen that the velocity profile can be decomposed into a macro Poiseuille component plus an EDL effect component. A closer inspection of (1) allows expressing the later in another close form as:

$$u_{EDL}= - 4\frac{I_1-I_2}{\dfrac{\overline{\lambda_0\mu}\sinh\kappa}{\bar{\zeta}^2}+4\left(I_3-\dfrac{I_4}{\sinh\kappa}\right)}\left\{1-\left|\frac{\sinh\kappa y}{\sinh\kappa}\right|\right\} \quad 4$$

The parameter $\overline{\lambda_0\mu}$ is the non-dimensional electrical conductivity times the dynamic viscosity of the fluid. It can be seen that the scaling parameter for $\lambda_0\mu$ is the square of the scaling charge density times the scaling length, i.e. $(\rho_S/k)^2$ leading to:

$$\overline{\lambda_0\mu}=\frac{\lambda_0\mu}{(n_0\,\varepsilon\varepsilon_0\,k_b\,T/2)} \quad (5)$$

There are three parameters governing the EDL effect, namely

$$u_{EDL}=u_{EDL}\left(\kappa,\bar{\zeta},\overline{\lambda_0\mu}\right) \quad (6)$$

Gradients of the dielectric strength, viscosity and conductivity should be incorporated in more realistic models. Yet, these effects are presumably negligible in significantly dilute solutions, such as deionized ultra filtered water (DIUFW) and pure organic liquids.

The main EDL effects may be summarized as follows:

- An increase of the friction constant and apparent viscosity.
- A decrease of the Nusselt number.

These effects are persistent yet not significantly important at least for large values of κ. The apparent viscosity increases for example by a factor of nearly 3 at $\kappa=2$, but the EDL effect on the wall shear stress disappears quickly when $\kappa \geq 10$ (Mala et al., 1997). The Nusselt number decreases by nearly 40% at $\kappa=5$, and less than 5 % at $\kappa=50$ (Tardu, 2003).

INFLEXIONAL INVISCID INSTABILITY

The EDL effect is undoubtedly significant for small values of κ for the liquids containing a very small number of due for example to impurities. In such situations the thickness of the diffuse layer may reach several micrometers. Fig. 2a compares the velocity profile of an " EDL flow" with the conventional Poiseuille flow, when $\kappa=41, G=12720$ and $\bar{\zeta}=2.1254$. This case corresponds to the flow of an *infinitely* diluted aqueous solution ($n_0=3.764\times 10^{19}\ m^{-3}$) through a microchannel of height 100 μm subjected to a Zeta potential of 50 mV. The Debye length is 1.2 μm, which is close to the value corresponding to the de-ionized ultra-filtered water experiments of Ren et al. (2002). The first impression one has from Fig.2a is the close similarity between the EDL and Poiseuille profiles with a decrease of the centreline velocity typical to the EDL flows. The increase of the friction constant is only 16 % in this situation. The important difference however is the presence of an inflexional point at $y \approx \frac{1}{\kappa} \operatorname{arcsin} h\left\{-\frac{2}{r\kappa^2}\sinh(\kappa)\right\}$ in the EDL profile where r is the ratio of the EDL and Poiseuille flow centreline velocities. This makes the flow inviscidly unstable, according to the Fjortoft's criteria (Fig. 2b). The inviscid instability does not imply instability directly in wall flows and an Orr-Sommerfeld analysis is necessary.

STABILITY ANALYSIS

The linear hydrodynamic stability under the EDL effect is studied through classical methods. The Orr-Sommerfeld equation is solved by a Galerkin-like procedure (Von Kerczek, 1982). The normal mode solutions of the disturbance equation are:

$$(u,v,p)=R(\hat{u},\hat{v},\hat{p})\exp(i\alpha x) \quad (7)$$

R is the real part, α is the dimensionless wave-number of the disturbance and x is the streamwise coordinate. Introducing the stream function:

$$\psi(x,y,t)=\phi(y,t)\exp(i\alpha x) \quad (8)$$

the Orr-Sommerfeld equation takes the form:

$$\frac{\partial}{\partial t}L\phi=\frac{1}{Re}L^2\phi-i\alpha\left(uL\phi-\frac{d^2u}{dy^2}\phi\right) \quad (9)$$

with the boundary conditions $\phi=\frac{\partial\phi}{\partial y}=0$ at $y=\pm 1$. The operator L is $L\equiv\frac{\partial^2}{\partial y^2}-\alpha^2$. The stream function is expanded in a Chebyshev polynomial series:

$$\phi(y,t)=\sum_{n=1}^{N} a_n(t)T_{2n-2}(y) \quad (10)$$

where $T_m(y)=\cos\left(m\cos^{-1}y\right)$ denotes the Chebyshev polynomials of the first kind. We took $N=256$ through this study. Eq. 9 takes the form:

$$\mathbf{Q}\frac{d\mathbf{a}}{dt}=(\mathbf{P}-i\alpha\mathbf{J})\cdot\mathbf{a} \quad (11)$$

and the matrices are determined by making use of the τ-method described by Orszag (1971). The last equation can be written as:

$$\frac{d\mathbf{b}}{dt}=\mathbf{D}\cdot\mathbf{b} \quad (12)$$

by introducing $\mathbf{B}$ the matrix that diagonalizes $\mathbf{Q}^{-1}\cdot(\mathbf{P}-i\alpha\mathbf{J})$, $\mathbf{b}=\mathbf{B}^{-1}\cdot\mathbf{a}$ and the diagonal matrix $\mathbf{D}=\lambda_i\delta_{ij}=\mathbf{B}^{-1}\cdot\mathbf{Q}^{-1}\cdot(\mathbf{P}-i\alpha\mathbf{J})\cdot\mathbf{B}$. The eigenvalues are denoted by λ_i and the flow is unstable when $R(\lambda_i)>0$. The disturbances with symmetric streamfunctions are considered only. The method gave very close results to Grosch and Salwen (1968) who used different sets of expansion functions.

RESULTS

Fig.2 shows the neutral curve corresponding to the microchannel-flow with the parameters given below, together with a "macro" Poiseuille flow (i.e. $\bar{\zeta}=0$ or $\kappa\to\infty$). It is clearly seen that the critical Reynolds number decreases by *a factor nearly equal to 2* under the effect of EDL: the critical wave and Reynolds numbers of the microflow are respectively $\alpha_c=1.10$ and $Re_c=3190$ to be compared with $\alpha_c=1.02$ and $Re_c=5772$ of the conventional Poiseuille flow. Due to its inviscid inflexional instability (unstable for $Re\to\infty$ for a given α), the band of unstable wave numbers of the EDL-microflow is significantly larger compared with the Poiseuille-macroflow. The destabilizing EDL effect disappears quickly when the height of the channel is increased by a factor of 4 (400 μm) (triangle in Fig. 2 corresponding to $\kappa=163$). This goes in the same line as previous experimental results showing the lack of micro-effects for the microchannels of height larger than typically 100 μm. For smaller values of κ, in return, the effect of the interfacial effects caused by EDL on the transition may be much more severe. For instance, the critical Reynolds number decreases up to $Re_c = 1042$ for a channel with a 40 μm separation distance subject to the same conditions (the square in Fig. 2 with $\kappa = 16$), and to a value as small as $Re_c = 496$ when $\kappa = 8$. It is clear that one of the most significant effect of EDL is the decrease of the critical Reynolds number, rather than the increase in friction coefficient or the apparent viscosity. For $\kappa = 40$, the friction factor increases by some 7%, but the critical Reynolds number DECREASES by 100%.

Fig. 3 shows the spectrum of eigenvalues as a function of Re for a single fixed wave-number $\alpha = 1$ and $\kappa = 16$. Only the first three eigenvalues corresponding to the complex wave speed $c = -\lambda / i\alpha$ are shown for the sake of clarity. The modes are ordered in such a way that ascending values correspond to descending values of Im $\{c\}$, i.e. of $R\{\lambda\}$. Remind that the flow is stable (unstable) for $R\{\lambda\}$<0 (>0). The Re number was varied from 300 to 20.10^3. The curves corresponding to Poiseuille and EDL-flows are denoted respectively by P-i and E-i. It is seen in Fig.3 that only the first mode has a different Im $\{c\}$ under the EDL effect and that the imaginary parts of higher modes collapse entirely with those of the macro scale flow. Note however that Im $\{c_{1-EDL}\}$is much more positive than Im $\{c_{1-Poiseuille}\}$showing the strong destabilizing effect of the EDL. The EDL modes are slower than the Poiseuille ones, in the sense that $R\{\lambda_{EDL}\}$is smaller than $2/3$ which is the average velocity of the channel, while the modes 2 and 3 of macro scale flow are clearly fast modes (their phase speed is close to the centerline velocity).

DISCUSSION AND CONCLUSION

The macroscale Poiseuille flow is ***metastable***; i.e. the corresponding stability is ***subcritical.*** Non linear analysis shows that the instability may occur with finite amplitude when all infinitesimal disturbances are stable (Drazin and Reid W.H.,1981). There is a significantly lower critical value of the Reynolds number Re_G compared to Re_c, above which the flow is unstable and below which there is no bifurcation. The exact theoretical determination of Re_G is still a matter of research. Experiments show that $\frac{\mathrm{Re}_G}{\mathrm{Re}_c} \approx \frac{1000}{5772} \approx \frac{1}{6}$. The transitional Reynolds number depends on the shape and shape factors of the channels.

The Poiseuille flow is monotonically stable only for $\mathrm{Re} < 100$ to be compared with 5772. The transitional Reynolds number Re_t of channel flows is about 400 according to experiments (2000 based on the hydraulic diameter and channel averaged velocity). Thus, the ratio $\frac{\mathrm{Re}_t}{\mathrm{Re}_c} < \frac{1}{15}$ in macroscale Poiseuille flows.

The main non-linear stability mechanism in *Macro Poiseuille flow* is due to a secondary instability with the development *of spanwise inflexion with strong shear* (Itoh, 1974). In the EDL flow there *is already a streamwise inflexion point in the base flow.* Thus the Reynolds numbers Re_G and Re_t should be *much lower under the EDL effect.* A non-linear analysis is necessary to check out this point but the first stage is then the determination of the marginal curve which is done here. Some arguments on the reinforcing effect of the EDL on the subcritical nature of the macro Poiseuille flow may however already be given. The square of the amplitude of a finite disturbance is given by:

$$\frac{d|A_1^2|}{dt} = 2\alpha c_i |A_1|^2 + (k_1 + k_2 + k_3)|A_1|^4 \qquad (13)$$

according to Stuart (1960). The flow reaches a subcritical equilibrium state when $k_1 + k_2 + k_3 > 0$. The coefficient k_1 represents the distortion of the mean motion: it is related to the eigenfunctions of the linear stability problem, and it is negative. The coefficient k_2 is linked to the generation of the harmonic of the fundamental and is also likely negative. The wall normal distortion of the fundamental (k_3) must " be positive and outweigh the combined negative effect of k_1 and k_2 to reach a subcritical state". Now, k_1 is proportional to $\mathrm{Re}_c \alpha_c^2$ (Eq. 6.3 in Stuart). It has therefore a significantly smaller negative contribution to $k_1 + k_2 + k_3$ under the EDL effect. Furthermore, part of the terms involving in the coefficient k_3 is inversely proportional to Re_c according to the equation 6.5 of Stuart (1960) and the EDL presumably reinforces the positive character of k_3 in the subcritical state.

The experimental verification of the linear stability results is difficult because of the subcritical character of the stability mechanism, and the difficult control of the level of turbulence at the inlet (Nishioka et al., 1975). That also rises the question of the EDL effect on the stability in the developing region. It is also somewhat difficult to reach such high Reynolds numbers in microchannels, even though one may consider the possibility of Debye lengths larger than $1\,\mu m$, thus higher channels compared with the numerical example given before. Well-controlled experiments are certainly more difficult in microchannels. Yet, it should be possible to detect the EDL effects on the transitional Reynolds number by classical Re-pressure gradient curves. Table 1 shows the expected transitional

Reynolds numbers under the EDL effect as a function of κ. The ratio $\frac{\mathrm{Re}_t}{\mathrm{Re}_c}=\frac{1}{15}$ is taken same as in macro channels, although, it is more than likely that $\frac{\mathrm{Re}_t}{\mathrm{Re}_c}$ is significantly smaller under the EDL effect according to the arguments given above. We will show in the presentation that the expected transitional Reynolds number is within the range of experimental possibilities. For larger channel of typical height 100 μm or so, the critical Reynolds number should also be measured, at least in principle. The experiments are limited to small Reynolds numbers for microchannels of height smaller than 20 μm, because very high pressure is then necessary and the channels will then break.

The transitional Reynolds numbers reported by some authors indicating early transition in microchannel liquid flows agree also qualitatively with the estimation given here. All of these experiments have been conducted with DIUF-W but the channel wall material may be different from one experiment to the other. In lack of experimental details it is hard to give definitive conclusions. Yet, there is a satisfactory agreement between the estimated theoretical Re_t and the measurements, in particular with those of Weilin et al. (2000) as it will be shown in the oral presentation. These authors indicated that "the range of Re_t values varies somewhat, depending on the hydraulic diameter and the material of the wall" pointing at a plausible EDL effect.

To conclude, the EDL destabilizes the linear modes of the Poiseuille channel flow and early transition in microchannels is plausible. This effect can be experimentally checked, provided that the liquid contains a very small amount of ions and the channel height is sufficiently small. In practice that would require the use of DIUF-W or organic liquids, and channel heights larger than 20 μm (but smaller than typically 100 μm) for the feasibility problems (microchannel failures at high pressures). There is no effect on stability for liquids with high ionic concentration and/or mini channels. The non-linear stability analysis of the EDL flow is necessary, although physical considerations indicate a much more rapid transition compared to macro flows. Direct Numerical Simulations can also be helpful to this end. Controlled experiments in a way similar to those reported by Ren et al. (2002) have to be conducted, by keeping the same channel with the same roughness distribution, and changing the ionic concentration of the liquid. The fact that the channels cross-section shape, in particular the corners have important contribution to the EDL field have also to be considered (Yang et al., 1997, 1998).

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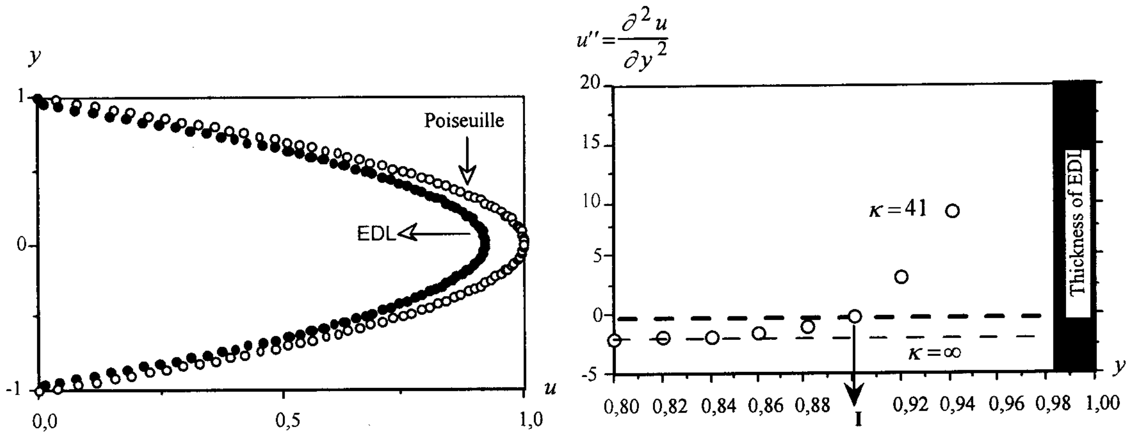

Figure 1 EDL and Poiseuille velocity profiles (a), and the inflexional instability of Fjortoft type (b). The parameters of the EDL flow are given in the text. The broken line in (b) corresponds to Poiseuille flow with $\kappa = \infty$ and the circles to the $\kappa = 41$ EDL flow. I. shows the inflexion point The inviscid instability is of Fjortoft type.

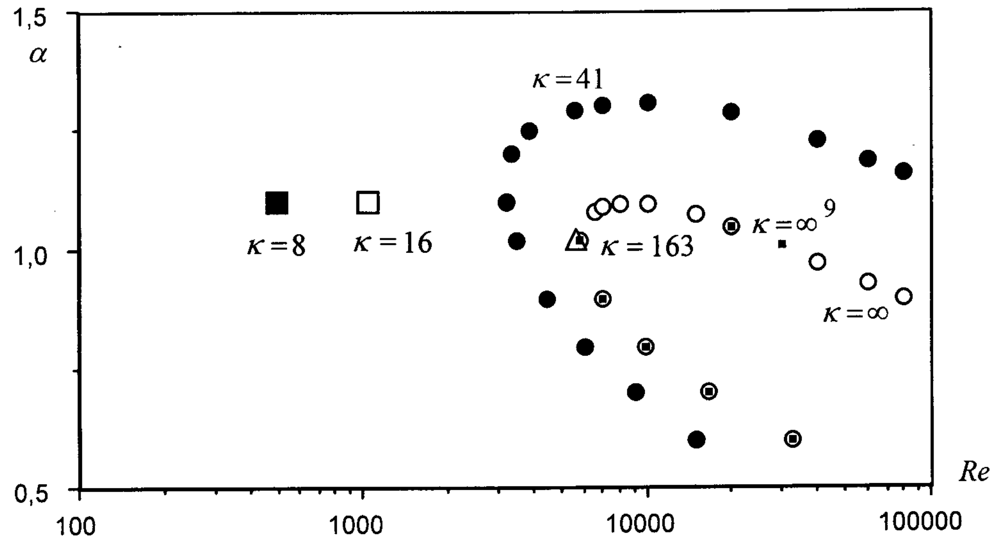

Figure 2 Neutral curves of the EDL flow compared with the Poiseuille flow. The open circles correspond to Poiseuille flow with $\kappa = \infty$. The results of Grosh and Salwen (1968) are shown by small bold squares. Bold circles correspond to $\kappa = 41, G = 12720$ and $\bar{\zeta} = 2.1254$. The rest of the results are obtained by changing the microchannel height and keeping constant the rest of the parameters.

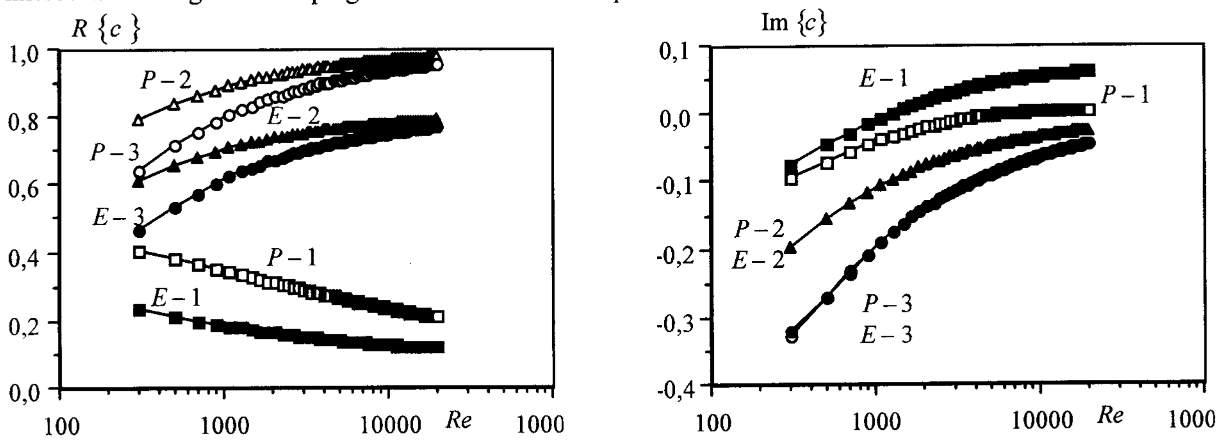

Figure 3 Real (left) and imaginary parts (right) of the first three eigenvalues versus $Re, \alpha = 1$ and $\kappa = 16$.