

Using Electrical Resistance to Characterize the Flow Parameters of the Solution in Micro/Nano Fluidics Channel

QU Xiao-peng, LIU Jing

(Cryogenics Laboratory, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100080, China)

Abstract: The electrical resistance detection using a hollow cylindrical electrode couple serving as part of the flow channel was proposed to characterize the flow rate, the concentration or the temperature of the electrolytic solution flowing in a micro or even nano channel. A theoretical equations were established to correlate the electrical resistance with these flow parameters and the mechanisms involved were interpreted. Further, several typical conceptual experiments were performed to test the effects of the temperature, the flow rate and the concentration of the solution to the electrical resistance of a column of the flowing electrolyte in mini/micro channel, and strongly supporting results were obtained. How to simultaneously identify multiple parameters incorporating this method with other measurements were discussed.

Key words: micro/nano fluidics; sensor; electrical impedance; Lab-on-a-Chip; μ -TAS

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电阻抗刻画微/纳米流体通道中溶液的流动参数

曲小鹏, 刘 静

(中国科学院理化技术研究所低温中心, 北京 100080)

摘要: 提出了采用一对中空的圆柱形电极作为微/纳米流体通道的一部分, 对流经该处的电解液的电阻值进行测定, 由此刻画溶液的流动速率、浓度及温度等参数的方法。从理论上推导出流体电解液的电阻值与上述参数之间的关系式, 并解释了该方法的测试机理。在此基础上, 完成了几类典型的原理性实验, 对流动于毫/微米通道中的溶液的温度、流速及浓度等对电阻值的影响进行了测试, 结果表明基于电阻测定可以很好地刻画微流体的性能。进一步讨论了将该方法与其他测量措施相结合, 以同时确定多种微流体参数的可行性。

关键词: 微/纳米流体; 传感器; 电阻抗; 芯片实验室; 微全分析系统

1 Introduction

With the development of μ -TAS and Lab-on-a-

Chip system technology^[1,2], the micro fluidics based techniques progress rather rapidly recently and many novel devices have been developed,

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such as the PCR and drug delivery device, etc. In these micro fluidic systems, a precise and timely measurement of physical and chemical parameters of the flowing electrolyte in the microchannel are very important. For example, in a PCR chip, the temperature development generally determine whether a PCR process is successful or not. In other situations, such as in a micromixture chip system, the flow rate is another important parameter, which affects the mixing result significantly. To maintain these chips work appropriately, quite a few new techniques have been developed to measure these different parameters, such as temperature^[3], flow rate^[4], mass flow and fluid density^[5,6], thermal properties (thermal diffusivity and conductivity)^[7,8], chemical concentration^[9,10], etc.

Generally, the temperature, the flow rate and the concentration are among the most basic parameters in the electrolyte critically requested to be measured, from which other parameters can possibly be deduced. While for the temperature measurement, the currently available methods were mainly based on a micro thermoresistor or thermocouple to monitor the changing of the temperature. Due to the highly confined geometry, these methods have to adopt complicated MEMS technique to fabricate the sophisticated micro thermal sensor, which, however, seriously increase the cost. Besides, in a micro fluidics related measurement, the micro thermal sensor must be mounted in the microchannel to accurately measure the temperature there, which not only improves the difficulty of the assembly process but also strongly affects the flow state of the electrolyte. Such drawbacks can in fact be glimpsed from the physical picture (see Fig.1)^[3]. Therefore, it can be concluded that in some fluidic situations, the micro temperature sensor is not appropriate as an ideal measurement method.

Sparks *et al.*, have proposed a single MEMS chip to measure several parameters of electrolyte^[11].

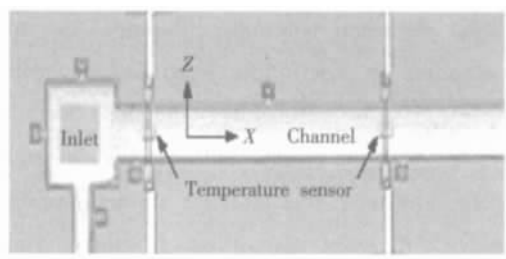


Fig.1 The temperature sensor in the microchannel

图1 微流道内的温度传感器

However, in most cases, multiple sensors generally have to be applied to simultaneously detect these parameters. Such strategy is rather difficult to be assembled in a microfluidic system especially nano channel due to its extremely limited space. Therefore, sensors which is capable of measuring anyone of these three parameters and can easily be integrated with the chip without inducing side effect to the solution are very important in developing a Lab-on-a-Chip and μ -TAS.

Such method introduces only a couple of electrodes along the channel to measure the electrical resistance of the electrolyte between them. If the electrode was particularly made as a hollow cylindrical tube, which also serves as part of the flow channel, then it will cause no effect to the flow. Besides, such simple configuration makes it easy to fabricate and assemble the sensors. Therefore, it would found significant applications in a micro or even nano fluidics system.

2 Theoretical model

The equivalent conductance Λ for the electrolyte can be defined as

$$\Lambda = \frac{\sigma}{vN} \quad (1)$$

where, σ is the specific electrical conductivity of the electrolytic solution; v the valency of ion in the solution; N is Avogadro's constant. The value of Λ depends only upon the ionic velocities and the degree of ionization^[12].

If an electrical potential difference was applied on a column of electrolytic solution, according to the basic laws of electricity, the ions within the electrolyte will be subjected to an accelerating force. In addition, the medium through which the ions move will exert upon them a viscous drag, or retarding force, whose value depends upon the size of the ion, the velocity and the coefficient of viscosity of the solvent (the temperature is one of the most important factors that affect the viscosity of the solvents). Thus, the ion will accelerate until the two opposing forces become equilibrium. After that, it continues to travel with a steady velocity and conductance of the electrolyte remains at a new level constantly [12].

Experimental observations have shown that electrolytic conductance depends on the thermodynamic variables such as concentrations, temperature, pressure, dielectric constant and viscosity of solvent, valence type of the electrolyte, field strength and frequency used in measurement, etc [13]. If some of these parameters were kept unchangeable, it is possible to determine the changing of one or more parameters by measuring the equivalent conductance.

The modern theory for strong electrolyte conductance was first developed by Debye and Hückel [14] and then developed fully by Onsager, Fuoss, *et al* [13]. However, the above theories were established mainly for analyzing the steady state electrolyte but not the flowing one. Therefore, they can not be directly used in the presently studied flowing electrolyte.

When an electrolytic solution is under equilibrium state, the central kation ion is surrounded with anionic atmosphere; at the same time the central anion is surrounded by cationic atmosphere. If an electrical potential difference is applied on the solution, the anion and kation would move to the electrodes in opposite direction; the kation

moves to the cathode, while the anion to the anode.

Supposed that the velocity of the kation with charge of e_1 is v_1^0 under the electrostatic field (E) while the velocity of the same kation under the unit electrostatic field is u_1^0 . The force to drive the kation to move can be written as

$$F = E e_1 \varepsilon \quad (2)$$

Further, one has

$$u_1^0 = \frac{v_1^0}{E e_1 \varepsilon} \quad (3)$$

i.e.

$$v_1^0 = E e_1 \varepsilon u_1^0 \quad (4)$$

When the electrostatic field is applied on the electrolyte, the kation and anion move to cathode and anode separately, so the dissymmetry comes into being and the effect of relaxation is created. The variation of the electrostatic field that creates the relaxation effect is supposed to be ΔE , and the electrical potential at the distance of r away from the central ion is

$$\psi = \frac{e_1 \varepsilon}{D r} e^{-kr} \quad (5)$$

where, D is the dielectric constant; ε is the charge of an electron; k is a constant in Debye and Hückel's theory.

According to reference [15] one has

$$\Delta E = - \frac{|e_1 e_2| \varepsilon^2}{3DKT} \cdot \frac{q}{1 + \sqrt{q}} kE \quad (6)$$

where, $q = \frac{|e_1 e_2|}{|e_1| + |e_2|} \cdot \frac{l_1^0 + l_2^0}{|e_2| l_1^0 + |e_1| l_2^0}$; l_1^0 , l_2^0 are the equivalent conductance of the kation and anion in the infinitely diluted electrolyte separately; K is the Boltzmann's constant.

For the electrolyte with flow rate of v , when the electrostatic field is applied, the ion in the electrolyte is pushed both by the relaxation force and the electrophoretic force.

The variation of the ionic velocity under the effect of relaxation is supposed to be v_1' and can be obtained as

$$v_1' = (E + \Delta E) e_1 \varepsilon u_1^0 = E e_1 \varepsilon u_1^0 \left(1 + \frac{\Delta E}{E} \right) = v_1^0 \left(1 + \frac{\Delta E}{E} \right) \quad (7)$$

The variation of the velocity of the ion under the electrophoretic force is supposed to be Δv_1 which can be obtained as

$$\Delta v_1 = -\frac{e_1 \varepsilon E}{6\pi\eta} \cdot \frac{k}{1+ka} \cdot \left(1 + \frac{\Delta E}{E}\right) \quad (8)$$

where, π is the electrode potential; η is the coefficient of viscosity; a is the mean diameter of ions.

Therefore, under the application of the electrostatic field, the variation of the fluid v_1 can be obtained as

$$v_1 = v_1' + \Delta v_1 + \nu = \left(v_1^0 - \frac{e_1 \varepsilon E}{6\pi\eta} \cdot \frac{k}{1+ka}\right) \cdot \left(1 + \frac{\Delta E}{E}\right) + \nu \quad (9)$$

Then one has

$$\frac{v_1}{v_1^0} = \left(1 - \frac{e_1 \varepsilon E}{6\pi\eta v_1^0} \cdot \frac{k}{1+ka}\right) \cdot \left(1 + \frac{\Delta E}{E}\right) + \frac{\nu}{v_1^0} \quad (10)$$

The velocity of the ion is proportional to the equivalent conductance of the fluid, i.e. $\frac{v_1}{v_1^0} = \frac{l_1}{l_1^0}$.

Besides, u_1^0 has such relation with l_1^0 as

$$u_1^0 = \frac{N l_1^0}{|e_1| F \times 10^7} \quad (11)$$

where, F is Faraday's constant.

From equation (4) and (11), one can get

$$\frac{E}{v_1^0} = \frac{1}{|e_1| \varepsilon u_1^0} = \frac{F^2 \times 10^7}{N \varepsilon l_1^0} \quad (12)$$

From equation (10) and (12), one has

$$\frac{l_1}{l_1^0} = \left(1 - \frac{|e_1| F^2 \times 10^7}{6\pi\eta N l_1^0} \cdot \frac{k}{1+ka}\right) \cdot \left(1 + \frac{\Delta E}{E}\right) + \frac{F^2 \times 10^7}{N E \varepsilon l_1^0} \cdot \nu \quad (13)$$

From this equation, the electrical resistance equation can be written in the symbolical form of $R=R(T, c, \nu)$, which indicates that the electrical resistance of a column of flowing electrolyte depends on its temperature, concentration and flow rate etc. Therefore, it can be concluded that if any two of these three parameters were known then the third parameter can be calculated by performing a measurement on the electrical resistance.

which leads to

$$l_1 = \left(l_1^0 - \frac{|e_1| F^2 \times 10^7}{6\pi\eta N} \cdot \frac{k}{1+ka}\right) \cdot \left(1 + \frac{\Delta E}{E}\right) + \frac{F^2 \times 10^7}{N E \varepsilon} \cdot \nu \quad (14)$$

Similarly, l_2 can be obtained as

$$l_2 = \left(l_2^0 - \frac{|e_2| F^2 \times 10^7}{6\pi\eta N} \cdot \frac{k}{1+ka}\right) \cdot \left(1 + \frac{\Delta E}{E}\right) + \frac{F^2 \times 10^7}{N E \varepsilon} \cdot \nu \quad (15)$$

Because the equivalent conductance of the electrolyte has such relation as

$$\begin{aligned} \Lambda &= l_1 + l_2 \\ \Lambda_0 &= l_1^0 + l_2^0 \end{aligned} \quad (16)$$

Then, we get the equivalent conductance of the flowing non-associated diluted electrolytic solution as

$$\begin{aligned} \Lambda &= \left[\Lambda_0 - \frac{(|e_1| + |e_2|) F^2 \times 10^7}{6\pi\eta N} \cdot \frac{k}{1+ka}\right] \cdot \\ &\quad \left(1 - \frac{|e_1 e_2| \varepsilon^2}{3DKT} \cdot \frac{q}{1 + \sqrt{q}} k\right) + \frac{2F^2 \times 10^7}{N E \varepsilon} \cdot \nu \end{aligned} \quad (17)$$

The electrical resistance equation of the electrolyte can be defined as

$$R = \frac{1000}{c} \cdot \frac{l}{s} \cdot \Lambda = \frac{C_0 \Lambda}{c} \quad (18)$$

where, l is the length of the channel and s is the cross sectional area of the channel. The $\frac{1000}{s} \cdot l$ was defined as a tube's constant C_0 which is determined only by the channel shape and size.

In this way, the electrical resistance equation of the electrolyte can be defined as

$$R = C_0 \cdot \frac{\left[\Lambda_0 - \frac{(|e_1| + |e_2|) F^2 \times 10^7}{6\pi\eta_0 N} \cdot \frac{k}{1+ka}\right] \cdot \left(1 - \frac{|e_1 e_2| \varepsilon^2}{3DKT} \cdot \frac{q}{1 + \sqrt{q}} k\right) + \frac{2F^2 \times 10^7}{N E \varepsilon} \cdot \nu}{c} \quad (19)$$

3 Experiments

From the above theoretical analysis, it can be found that the electrical resistance of the electrolyte depends on its flow rate, temperature and concentration. This relationship can in fact be easily demonstrated through several conceptual experiments. To obtain a preliminary understanding of this physical picture, a set of equipment was de-

signed to measure the electrical resistance of the electrolyte changes, as schematically illustrated in the Fig.2.

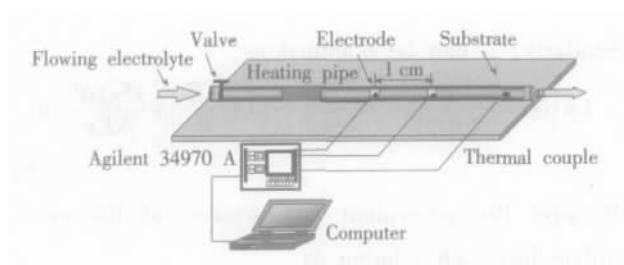


Fig.2 Experimental set up to detect electrical resistance of solution in flow channel

图2 测试微流内流体电阻值的实验台

In this system two microchannels, which were made of polyethylene were filled with sodium chloride solution as the electrolyte. In the temperature experiment the internal diameter of the microchannel is 1.5 mm, while for the concentration and the temperature experiments the diameter of the microchannel is 4 mm to demonstrate the effect of changing the temperature and the concentration on the electrical resistance. A valve was introduced in the microchannel to control the flow rate of the electrolyte. A segment of the microchannel at the up side of the valve is substituted by a metallic pipe which can be selectively heated by an alcohol burner to serve as the heater segment to vary the temperature of the electrolyte running through the tube. At the downriver of the heater, there is a couple of metallic hollow tubes to replace the two segments of the plastic microchannel to serve as a couple of electrodes. The length for each metallic tubes serving as electrode is 1 cm and the distance of the couple of electrodes is 1 cm. There is a thermocouple near the second electrode to measure the temperature of the electrolyte there. The temperature values measured by the thermocouple are compared to the parameters measured by the couple of electrodes. All the signals measured by the thermocouple and electrodes are accuquisted into

the Agilent 34970 A for later analysis.

Clearly, since the metallic tubes with concentric cylindrical structure were adopted to substitute plastic microchannel as heater and electrodes, the influence of them on the flowing state of the electrolyte were minimized and their machining process was significantly simplified.

In the experiment, the DC electricity was imposed on the electrolyte during the whole measurement, some side-effect on the experimental result will be discussed later in this paper.

In testing the temperature effect, the concentration of the sodium chloride solution adopted as the electrolytic solution was 0.11 mol/L and the flow rate was kept at constantly 0.014 m/s in the channel when the valve opens. The temperature pulse was created when the alcohol burner heated the heater segment of the pipe. Electrodes and the thermocouple then measure the electrical resistance and the temperature respectively.

In the flow rate-electrical resistance relation experiment, the flow rate of the electrolyte was adjusted by the valve. In the concentration-electrical resistance relation experiment, the changing of the concentration was realized by switching the micro-channel between beakers with different electrolyte concentrations.

All the experiments were done at 23.6 °C.

4 Results

4.1 Effects of temperature

During the experiment, the temperature of the electrolyte was varied by the heater segment and the transient electrical resistance of the electrolyte thus induced was measured by the couple of electrodes. By analyzing the data acquired from the electrodes and the thermocouple, one can approximately grasp the relationship of the temperature with the electrical resistance of the electrolyte.

As indicated in Fig.3, when a pulse heating

with duration time of 10 s was imposed on the external wall of the tube (i.e. the heater segment), it will raise the temperature of the solution running through it. This in turn results in a sudden decline for the electrical resistance curve of the solution. Clearly, there exists a strong dependence of the electrical resistance of the solution on the temperature.

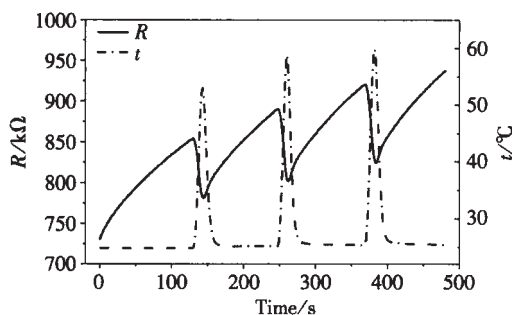


Fig.3 The response of the electrical resistance of the solution to the pulse heating

图3 脉冲加热作用下溶液电阻值的变化情况

Further, one can still observe from Fig.3 that the response of the electrical resistance to the change of the temperature is very quick. However, after the decline of the electrical resistance, it can not resume to its original value even the transient heating has been stopped. The reason can partially be attributed to that the flow rate of the electrolyte tend to decrease with the progress of the experiment. This is because the total volume for the tested solution was initially kept constant and its flow was driven by gravity via elevated the container to a fixed height above the flow channel. Therefore, with the running out of the solution, the flow rate inside the channel will gradually decrease, it results in a gradually increased electric resistance of the solution. The heating effect of the electrode couple to its accurate measurement may also possibly contribute to the above phenomenon. But comprehensively addressing this problem still needs future research. While in a real Lab-on-a-

Chip and μ -TAS, because the sizes of the channel and the electrodes are extremely small, the thermal capacity of the electrodes is very low. In this case, the above lingering effect will become so small that can be neglected totally.

In the experiment, alcohol burner was adopted as the heating device to show the evident effect of the temperature on the solution electrical resistance. Therefore, the changing of the solution temperature appears rapid with a wide fluctuating range. Sometimes, it may lead to generation of certain bubbles in the microchannel due to a strong heating. In fact, this has been reflected by the peak jump in the electrical resistance curve as illustrated in the Fig.4. The relationship between the peak value of the electrical resistance and the formation of the bubbles could be used as a very useful indicator to monitor the bubble behavior in a micro/nano fluidic channel. This may find some applications in developing certain sensor and actuator system.

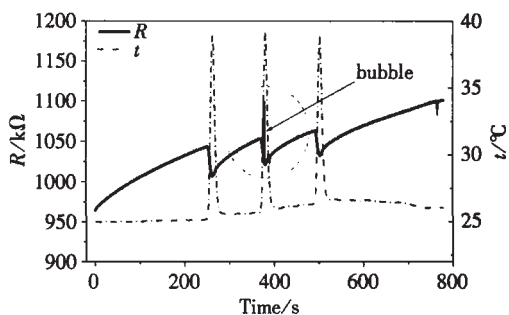


Fig.4 The effect of bubbles on the change of the electrical resistance

图4 气泡对溶液电阻值变化的影响

4.2 Effects of flow rate

When the water flow rate was intentionally adjusted by the valve, the changing of its electrical resistance was recorded by the electrodes. The relationship between them could be evidently observed from Fig.5.

The electrical resistance of the electrolyte

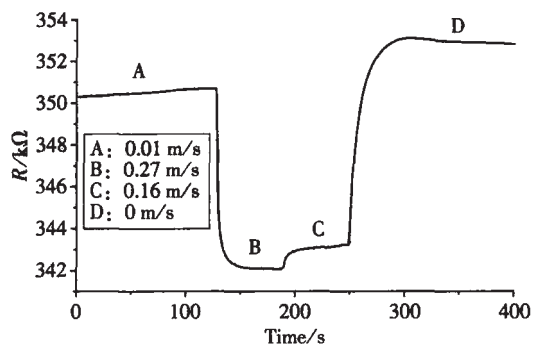


Fig.5 The response of the electrical resistance to the flow rate of the electrolyte

图 5 电阻值对电解液流速变化的响应情况

changed correspondingly with the variation of the flow rate. When the flow rate increased to a higher value, such as from A to B, the electrical resistance declined to a lower value quickly in just several seconds and then maintained at that value. When the flow rate of the electrolyte decreased, such as from B to C or from C to D, the electrical resistance increased to a stable value with a relatively slow speed. This result accords well with the above analysis on the relation between the flow rate and the electric resistance of the solution. The response of the electrical resistance to the changing of the flow rate is relatively slow because the size of the channel used is still large. When diameter the channel becomes very small such as several microns, the response time can be significantly shortened.

When the other parameters were fixed, the flow rate of the solution can be deduced by measuring the electrical resistance of the electrolyte.

4.3 Effects of concentration

In such experiments, when the channel was switched to an electrolyte with different concentration, the electrical resistance of the solution running through the hollow electrode couple also subjected to change quickly. This can be observed in Fig.6 when the solution concentration was changed from A to B and the electrical resistance varied

quickly in several seconds. Clearly, this method is very sensitive even the diameter of the channel is in several millimeters, let alone in the situation where the diameter was diminished to microns.

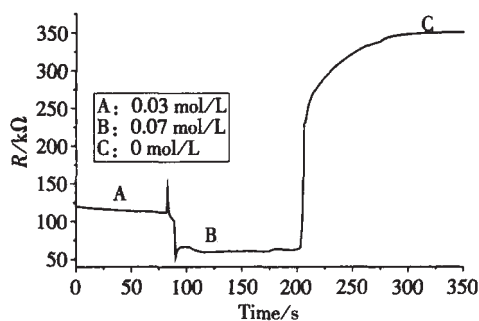


Fig.6 The response of the electrical resistance to the changing of the concentration

图 6 电阻值随电解液浓度变化的响应情况

For a relatively large scale channel, the electrical resistance of the flowing solution usually took a longer time to reach its new stable value, due to the process for the solution concentration to become equilibrium (see B to C in Fig.6).

From the above experiments, it can be seen that when other parameters were fixed the flow rate and the temperature were measured by other sensors, the solution concentration can be deduced by detecting its electrical resistance.

5 Discussion

As indicated by the equation $R = R(T, c, \nu)$ (Eq.19), any variation of T , c or ν would lead to a change of R respectively. At the present stage, the new method can still not be applied to simultaneously detect T , c or ν unless it is incorporated with other sensors to measure two of them. However, this particular feature has made it a very attractive technique in some important microfluidic system, where sensors for the temperature, flow rate and concentration have been mounted simultaneously. In that case, the electrical resistance measurement can be applied as a spare

sensor. When anyone of these three sensors is broken, the electrical resistance sensor would serve for its role without ceasing working of the channel, because the parameter measured by the disabled sensor can be deduced from the electrical resistance equation with the assistance of other two parameters measured by the remained sensors.

In this experiment, the DC has been applied for the electrical resistance measurement which may lead to some side effect. For example, it can be observed from Fig.3 and Fig.4 that the curve increase gradually during the experiment. There are two reasons to account for this phenomena. The first reason is that the gradually reduced flow rate would increase the electric resistance of the electrolyte in the experiment, just as explained in previous section. The second one may lie in that the DC electricity would possibly electrolyze the electrolyte during the test, which in turn changed the electrical characteristic of the electrolyte.

With the decrease of the diameter of the channel, traditional sensors may be invalid due to large size, disturbance to the flow and difficulty in mounting them in an extremely small geometry. However, this is just the situation for the present method to work perfectly. Therefore, although in the above experiments, the internal diameter of the channel is still in a macro scale, the present method will be especially useful in the measurement of micro/nano fluidics. Should be pointed out that, to develop a practical equipment, the electric field applied on the flowing solution should be a kind of pulsant electric field in order to diminish the effect of the electric field to the solution and to accurately measure the electrical resistance. Further, when the size of the flow channel falls into the micro or nano scale, the electric double layer (EDL) inside the flow may plays an important role for the electrical resistance of the solution. For such case, the present theory still needs

modification which should comprise the electrolyte model and the EDL model together. This will be studied in our later research.

6 Conclusion

In this paper, a novel method to characterize the flow parameters of the electrolyte running inside the microchannel by measuring its electrical resistance was proposed and a theoretical model was developed to interpret the mechanisms involved. This method can be possibly applied in micro/nano channel to monitor the temperature, concentration or flow rate of the flowing electrolyte. The proposed hollow cylindrical structure for the sensor would significantly simplify the integration process of the whole system. With some improvement, the electrical resistance measurement could be a useful measurement unit in the μ -TAS and Lab-on-a-Chip device in the near future.

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- Biography:**
- JIA Yun-fang** (1974—), female, received Dr. degree in microelectronics from Nankai University in 2004, her research activities are the application of semiconductor sensors in the biochemical assay;
- FENG Xi-zeng** (1961—), male, professor of chemistry and biochemistry at the Nankai University, he is interested research in advanced materials and nanobiology;
- WANG Li-kai** (1981—), male, MD student, he majors in microfluidic networks in biology.
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- Biography:**
- QU Xiao-peng** (1980—), male, master degree student of the Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, he is interested in micro heat and fluidics research;
- LIU Jing** (1969—), male, professor of the Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, his research interests are in the fundamental and application researches on micro/nano fluidics and thermal devices, E-mail: jliu@cl.cryo.ac.cn.