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Thermoelectric Effect on Electroosmotic Flow in Microchannel

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Abstract. The present work reports the thermoelectric effect on electroosmotic flow (EOF) in microchannel. The thermoelectric (TE) effect on EOF was analyzed and the switch process of fluids controlled by TE unit was numerically simulated. The mathematical model was established that is composed with electrokinetics fluid, liquid-solid phase transition, and heat transfer. The microfluidic chip with the integrated TE unit was fabricated that includes the microchannel layer, heat conducting layer and refrigerating layer. As the transition from the liquid phases to the solid phase can significantly decrease electrical conductivity of electrolyte, the electrical current is available to make sure the status of phase transition. From the results, the controlling integrated TE unit is effective to close/open the EOF in microchannel. The freezing phenomena were described by results from numerical simulation and experiment.

1. Introduction

The development of lab-on-a-chip requires efficient fluids controlling for preventing contamination, adjusting flow velocity and direction, tunable sample supplying. The tunable flux and open-close switch of fluid are often required. EOF has been the most useful method to drive the fluid, due to the simplicity and easy-control advantages. But there are few reports about microvalves for EOF.

Various fluid switching methodologies have been developed for microfluidic devices. An interesting microvalve is the ice valve or phase changing (PC) valve. The PC valve is opened and closed by thermally-actuated phase changing (freezing or melting) of the local working fluid (ice plug). The thermal actuation is commonly realized by using the Peltier effect. Comparing to other valves, it is relatively noninvasive, reusable, and leakage-free. The principle of PC valve was firstly reported by Bevan etc (1995)^[1]. Later Gui and Liu (2004)^[2] tested cooling & heating the liquid in a cylindrical tube *via* a TE unit. More recently Welle (2005)^[3] integrated some mini TE units with a more complicated microfluidic chip.

The present work mainly deals with the thermoelectric effect on EOF in microchannel. In the following sections, the developed methodology is described, including the mathematical formulation, the numerical simulation, the prototype fabrication, and the evaluation of results.

2. Mathematical formulation

Consider an ideal 2D rectangular microchannel in which is filled with incompressible Newtonian electrolyte^{[4][5]}.

2.1. Electrical field

In the bulk liquid, thin electrical double layers (EDL) and zero net charge density are assumed. The external electric potential ϕ in the liquid can be described by the Laplace equation for electrostatics

$$\nabla \cdot \sigma \nabla \phi = 0 \quad (1)$$

Here σ is the electrical conductivity.

For the thin EDL, Smoluchowski equation $\vec{u}_{eo} = -\varepsilon \varepsilon_0 \zeta \vec{E} / \mu$ can be safely used as the slip boundary of velocity. where \vec{u}_{eo} is the slip velocity vector, ε is the relative permittivity, ε_0 is the free space permittivity, ζ is the zeta potential, μ is the dynamic viscosity of the electrolyte solution, \vec{E} is the electric field strength defined as $\vec{E} = -\nabla \phi$.

2.2. Flow field, Heat Transfer and Phase Change

The EOF equations in microchannels are the modified incompressible NS equation and the continuity equation

$$\frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \vec{u} = -\frac{1}{\rho} \nabla p + \mu \nabla^2 \vec{u} + \rho_e \vec{E} \quad (2)$$

$$\nabla \cdot \vec{u} = 0 \quad (3)$$

As the thermal capacity and thermal conductivity often change evidently following phase change, the different variables are induced. The energy equations for heat transfer are given respectively by

$$\rho \cdot C_p^L \left(\frac{\partial T}{\partial t} + \vec{u} \cdot \nabla T \right) = k^L \nabla^2 T + \sigma^L \vec{E}^2 \quad (4)$$

$$\rho \cdot C_p^S \frac{\partial T}{\partial t} = k^S \nabla^2 T + \sigma^S \vec{E}^2 \quad (5)$$

where superscript L and S are the abbreviate of liquid and solid respectively, k is the thermal conductivity of electrolyte, C_p is the thermal capacity of electrolyte, T is the temperature, and the source term of $\sigma \vec{E}^2$ expresses the influence of Joule heating in EOF.

The theory of phase change can be expressed as

$$H = h + \Delta H \quad (6)$$

$$\Delta H(T) = \begin{cases} L & T \geq T_{PC} \\ 0 & T < T_{PC} \end{cases} \quad (7)$$

$$h = T \cdot C_p \quad (8)$$

where H is the enthalpy, h is the sensible heat, L is the latent heat, T_{PC} is the phase changing (PC) temperature. The domain can be regarded as a porous medium and the porosity λ can be expressed as

$$\lambda = 1 - f_s = \begin{cases} 0 & \text{Solid} \\ 0 \sim 1 & \text{Hybrid} \\ 1 & \text{Liquid} \end{cases} \quad (9)$$

where f_s is the local solid fraction. The fluid velocity \vec{u} in NS equation should be modified as $\lambda \vec{u}_f$, \vec{u}_f is the actual fluid velocity from NS equation.

3. Numerical Simulation

The numerical simulations are performed using a commercial solver of the NS equations. The 2D microchannel is rectangular ($80\mu\text{m} \times 1200\mu\text{m}$). The 10mM KCL solution is selected as the work buffer. In order to simplify the process, we only assigned the independent parameters in solid phase and liquid phase. The summary parameters are shown in table 1.

Table 1. Summary Parameters ^{[6][7]}

	Liquid	Solid		Liquid	Solid
Density (kg/m³)	1000	1000	Dynamic viscosity (kg/m.s)	0.000855	
Zeta potential (V)	-0.1	-0.1	Latent heat (J/kg)	324000	324000
Deby thickness(nm)	10	10	Thermal capacity (J/Kg.K)	4179	1960
σ (S/m)	0.13	0.0013	Thermal conductivity (W/m.K)	0.613	2.400
Relative permittivity	78.5	78.5	PC temperature (K)	273	273

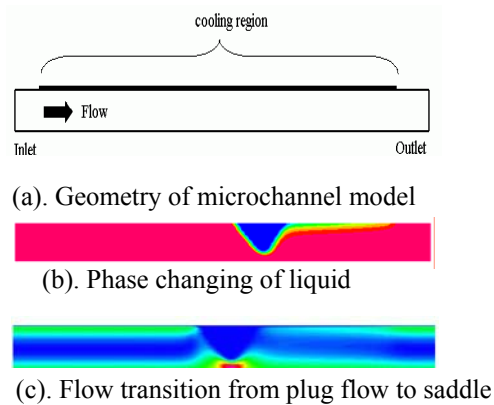


Figure 1. Simulation process of thermo-electric effect on EOF

As shown in figure 1.a, the boundary condition of electrical insulation is applied on all walls. On top of microchannel, the negative heat flux is applied for the endothermic&exothermic process (1000 μm length), except two adiabatic boundaries adjacent to inlet and outlet (100 μm length). On the bottom of microchannel, all boundaries are assumed adiabatic (1200 μm length). A potential drop and zero pressure drop condition are applied between inlet and outlet. The steady simulation was done. In all simulation, the ice cores always appears near the middle of cooling area firstly and then extends until heat balance (figure 1.b). In the freezing process, the distribution of EOF velocity transits from plug flow to saddle flow, due to the pressure gradient induced by phase change (figure 1.c).

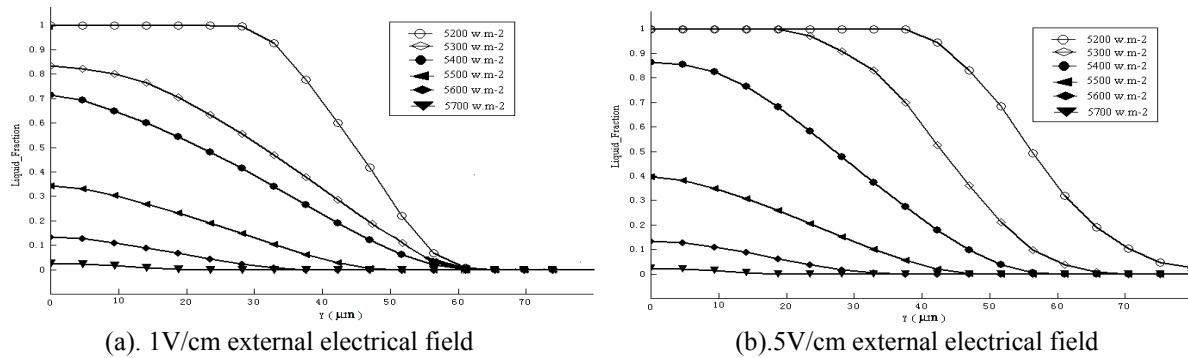


Figure 2. PC ratio with various cooling power density in the section of middle microchannel

The performance of PC valve is evaluated by the maximum PC ratio in microchannel section. The figure 2 shows PC ratio in the section of middle microchannel with different parameters. With the 1V/cm external electrical field of EOF, when the cooling power density (CPD) increases to 5200 $\text{W}\cdot\text{m}^{-2}$, the ice cores with 18 μm widths appears, and almost full clogging the channel with 5700 $\text{W}\cdot\text{m}^{-2}$ CPD. As the EOF external electrical field increases to 5V/cm, the width of ice cores almost decreases to zero with 5200 $\text{W}\cdot\text{m}^{-2}$ CPD. But following the increasing of CPD, the PC ratio curves approximately equal to the result of 1V/cm external electrical field and the CPD of full clogging is almost 5700 $\text{W}\cdot\text{m}^{-2}$ too. We have calculated the process with the condition of 10V/cm external electrical field too and the CPD of full clogging is still approximately 5700 $\text{W}\cdot\text{m}^{-2}$.

4. Experimental Setup

As shown in figure 3, the designed microfluidic chip is composed of microchannel layer, heat conducting layer, and refrigerating layer from top to down: The microchannel layer is formed from

PDMS (DowCorning Corp) by casting on the SU-8 mold. The size of microchannel is $60\mu\text{m} \times 80\mu\text{m} \times 80\text{mm}$ (W×H×L). The conductive layer is a glass slide with $120\mu\text{m}$ thickness that is irreversible bonded with microchannel layer. The refrigerating layer includes one TE unit and a frame for fixing the TE units. The TE unit is a Peltier cooler ($4\text{mm} \times 4\text{mm} \times 2.4\text{mm}$). The maximum heat flux density is $8.0\text{e}4\text{W}/\text{m}^2$. The frame is formed by a laser cutter from a PMMA piece (2mm thickness). The refrigerating layer is linked to heat conducting layer by glue.

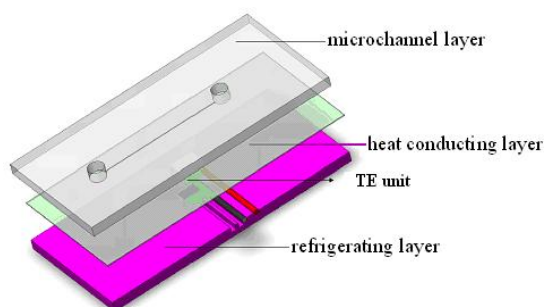


Figure 3. The microfluidic chip

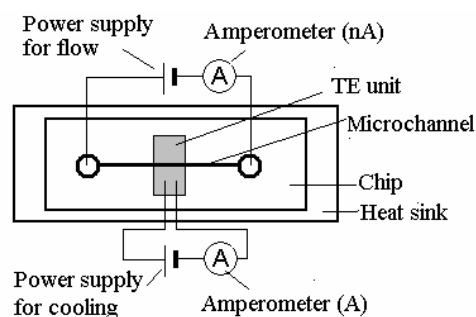


Figure 4. The setup of EOF driving and PC measurement

A $10\text{mM} \cdot \text{L}^{-1}$ KCL electrolyte is selected as buffer solution, which electrical conductivity at 23°C is about $0.13\text{S}/\text{m}$. Although there are still absent of exact information about electrical conductivity of KCL electrolyte in solid phase, mostly it will fall to one orders of magnitude at least^{[6][7]}. So the phase transition is ensured indirectly through the electrical current decreasing. The setup of EOF driving and PC measurement are shown in figure 4. A power supply is used to drive fluid flowing and another is used to drive the TE unit. A precise amperemeter (resolution 1nA) is used to measure the electrical current in microchannel. A steel heatsink is applied to advance the heat dissipation from TE heat tips.

5. Test Results

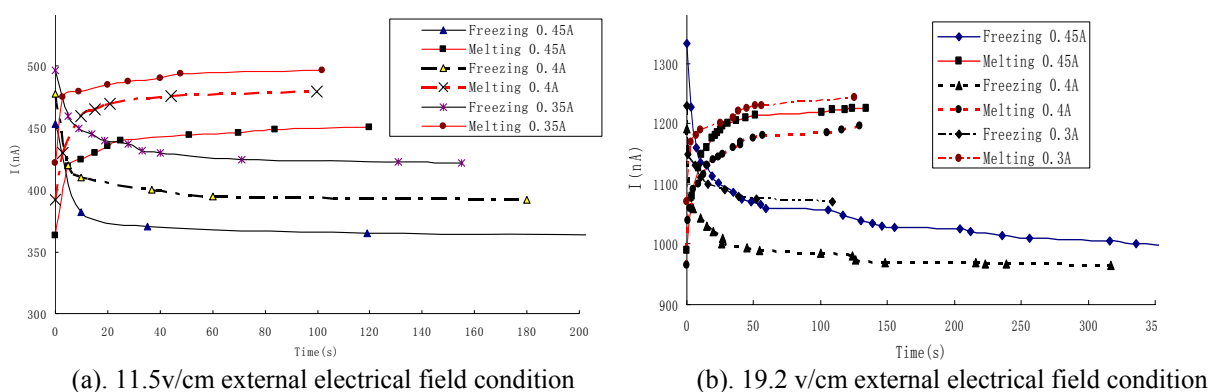


Figure 5. The fluctuation of electrical current in EOF

Previously the electrical conductivities have been measured *via* the precise amperemeter by freezing or melting all electrolytes in microchannel. The electrical conductivity in solid phase is about twentieth to that in liquid phase from the results. The heat flux of TE units is measured *via* a thermographic camera (A40 FLIR) and the typical TE heat flux values with electrical current (in brackets) were ensured such as $1.38\text{e}5\text{W}/\text{m}^2$ (0.45A), $1.21\text{e}5\text{W}/\text{m}^2$ (0.40A), $1.07\text{e}5\text{W}/\text{m}^2$ (0.35A), $0.85\text{e}5\text{W}/\text{m}^2$ (0.3A). Then the fluctuation of electrical current in EOF is dynamically monitored with different heat flux. Figure 5.a shows that the process of fluctuation with $11.5\text{v}/\text{cm}$ for EOF in freezing

and melting. Figure 5.b shows that the process of fluctuation with 19.2 v/cm for EOF in freezing and melting.

We use the following equation $\frac{I_{Start}}{I_{End}} = \frac{(L-x)/\sigma_L + x/\sigma_s}{L/\sigma_L}$ to evaluate the length of ice plug x .

With 11.5v/cm external electric field for EOF, the lengths of ice plug are 340 μm (0.45A), 300 μm (0.4A), and 236 μm (0.35A). With 19.2v/cm external electric field for EOF, the lengths of ice plug are 474 μm (0.45A), 250 μm (0.4A), and 201 μm (0.3A). When the TE heat flux decreases to 0.85e5w/m², no obvious fluctuation of electrical current is observed. When the external electric field for EOF increases to 38.0 V/cm, obvious fluctuation of electrical current isn't observed too. From results, the freezing periods of are 35s (0.45A), 60s (0.4A), 75s (0.35A) with 11.5V/cm external electric field for EOF and 408s (0.45A), 220s (0.4A), 109s (0.35A). Due to the difference of latent heat between liquid phase and solid phase, the melting processes often increases a little faster than freezing process.

6. Conclusions

The thermoelectric control method by cooling the liquid plug for EOF was presented, and it does not require any moving part. A mathematical formulation was given, in which electrical field, flow field, heat transfer, and phase change were considered. Furthermore, we simulated the freezing phenomena in EOF. The flow transition from plug flow to saddle flow was found in the phase change process. PC valve performance was predicted. For 1~10V/cm external electrical field condition of EOF, the cooling power density of full clogging is about 5700w.m⁻².

According to the fluctuation of electrical current, we experimentally found that its response time ranges from 35s to about 408s, and the length of ice plug ranges from 474 μm to 200 μm , depending on the external electric field of EOF and heat flux of TE. Issues of the measurement of the flow velocity distribution will require further research.

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