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Experiments of micro-flow control and micropump conducted by applying electric fields on the flow through a porous membrane have clarified that only NaCl solution in water is effective for the control of micro-flow among the liquids examined but KCl solution and a colloidal dispersion of polystyrene latex particles in water as well as NaCl solution are good as working fluids for the micropump. Although the applied voltages are less than 10 V, performance of the present micropump is found belonging to the highest class in the past methods. Effects of electro-osmosis, electrophoresis and absorption were discussed about whether or not they may become causes of the present phenomena and it is concluded that absorption is most possible as a cause. The colloidal dispersion of polystyrene latex particles is thought to be a good working liquid, because it provides the flow rate proportional to the applied voltage without generating any bubble of by-product.

Key Words: Micro-Fluid, Micropump, Electroosmosis, Electrophoresis, Membrane, Pore

1. Introduction

Control of micro-flow rates or micropump is a key technology in micro-fluid mechanics and microtechniques, and not a few methods for driving liquids are invented and utilized for medical treatment, process in making medicine, and experiment in bio-laboratories⁽¹⁾. In particular, the method making use of electric effects has recently attracted much attention, because it gives smooth flow by the structure of no movable mechanical part and makes easy control of the flow rate by changing voltages applied. Electroosmosis has been most widely utilized among the electric effects: Electroosmotic flow micropumps were fabricated with base on capillary systems packed with silica particles⁽²⁾, a porous core packed with sintered glass beads⁽³⁾, a densely packed particle structure between the polymer frits^{(4), (5)}, a donut channel constructed by a gap between two parallel donut-shaped glass plates⁽⁶⁾. However, relatively high voltages and complicated structures are required for those methods used so far. In this paper, we present a new and simple method that uses the flow generated through a porous membrane by applying voltages less than 10 V on both sides of the membrane, and provide some results obtained by this method.

2. Experimental Method and Apparatus

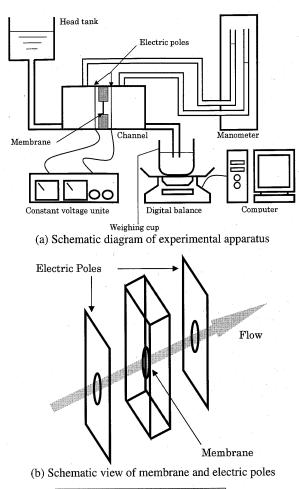
Figure 1 (a) gives a schematic diagram of the experimental apparatus: we set a porous membrane vertically to the flow direction in the channel in which electrolyte solution or colloidal dispersion in water is filled, and applied a voltage on the both sides of the membrane by two electric poles. The left hand side of the membrane is upstream and connected to a head tank, and the right hand side is downstream and connected to a weighing cup set on a digital balance. In the experiment of flow control, a liquid is made beforehand to flow from the head tank to the weighing cup through the channel and membrane, a voltage is applied at a time (usually 300 seconds after the inception of the flow), and the flow rate is measured with a digital balance and recorded with time in a personal computer. For the case of micropump, both heads of the head tank and the weighing cup are adjusted and made to be equal in advance (that is, no flow condition is realized), and then a voltage is applied and the generated flow rate is measured with time. For the two cases of flow control and micro-

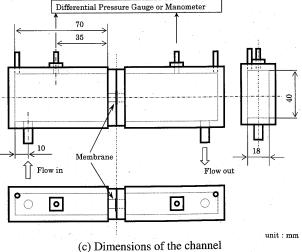
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pump, the voltage is supplied by a constant voltage generator and the pressure differential between the upstream and downstream positions is measured with a manometer or a pressure head of differential type.

Two kinds of membrane were used: one is a metal membrane of nickel whose thickness is $11 \,\mu\text{m}$ and in which 55600 pores of the diameter of nearly $5 \,\mu\text{m}$ are regularly opened in a circle of the diameter of 8 mm.

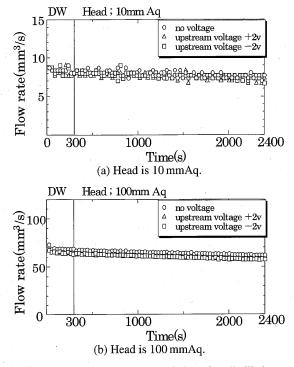


Fig. 2 Flow rate as a function of time for distilled water. Electric poles are stainless plates. Nickel membrane is used and the diameter of the pore is $6.28 \,\mu m$.

The other membrane is made of polycarbonate and 10 µm thick, and 320 000 pores of nearly 5 µm in diameter are randomly opened in a circle of the diameter of 10 mm. A silver plate and a zinc plate were used for the plus pole and the minus pole respectively in order to prevent bubble generation. Sometimes, however, stainless plates were used for both poles and in this case some bubbles were generated. A schematic view for setting of membrane and poles is shown in Fig. 1 (b). The membrane is pasted over a hole of 10 mm drilled on the base plate and the holes of the same size are opened in the plates of electric poles for smooth pass of flows. Figure 1 (c) gives dimensions of the channel used in the experiment. NaCl solution of 0.15 mol/L in water was mainly used in the experiment, because it is like a physiological solution, and KCl solution of the same concentration in water was also used for comparison. Furthermore, colloidal dispersion in water of polystyrene latex particles of the diameter of 0.12 µm was adopted in the experiment of micropump.

3. Experimental Results

3.1 Control of flow rates

Figure 2 (a) and (b) show the results of the flow rate for water through a membrane. The water was obtained by treating city water with a series of process; passing through a reverse osmosis membrane, distillation and again passing through ion exchange resin (this water is referred to as distilled water (DW)). Figure 2 (a) and (b) represent the case that the applied upstream heads

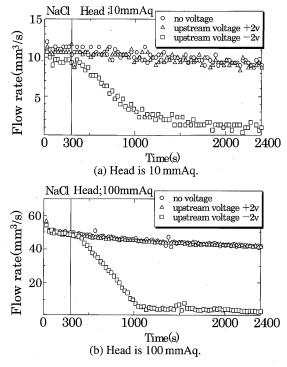


Fig. 3 Flow rate as a function of time for NaCl solution in water. The experimental condition is the same as in Fig. 2.

are 10 mmAq and 100 mmAq respectively, and this is the same in Figs. 3 and 4. In these figures, the ordinate is the flow rate through the membrane and the abscissa is the elapse time in seconds. A normal solid line drown at the time of 300 seconds indicates the inception of applying voltage after the beginning of the flow at the time of zero second. Figure 2(a) and (b) include the three cases; no voltage, +2V and -2V at the upstream, under the condition that the membrane is a nickel film and the poles are stainless sheets. We see from these figures that application of voltages to distilled water does not give any measurable effect on the flow rate. Figure 3 (a) and (b) give the results for a NaCl solution under the same experimental condition as in Fig. 2, and we see that the flow rate gradually decreases after applying a negative voltage in the upstream and becomes almost zero about 1 000 seconds after, but a positive voltage does not give any effect on the flow rate. Figure 4 (a) and (b) show the result for KCl solution in water under the same condition as Fig. 3 except that the applied voltage is 4 V. It is seen that +4 V and -4 V in the upstream region give a slight increase and decrease in flow rates respectively, which is very different from the case of NaCl solution. In Fig. 4, the flow rate is gradually decreasing with time even for no application of voltage. The reason is not clear, but it is thought that some absorption of ions to the wall of pores may pack the pores partially (see Discussion). Polycarbonate membrane was used instead of the nickel membrane and the result is given in Fig. 5. In this case also we see the decrease in flow

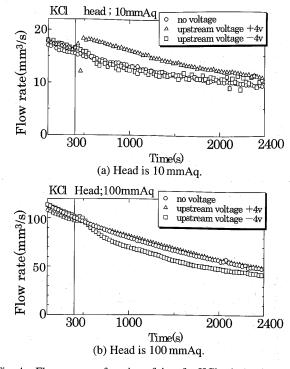


Fig. 4 Flow rate as a function of time for KCl solution in water. Electric poles are stainless plates. Nickel membrane is used and the diameter of the pore is 5.83 µm.

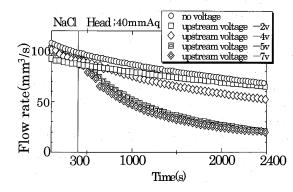


Fig. 5 Flow rate as a function of time for NaCl solution in water. Electric poles are stainless plates. Polycarbonate membrane is used and the diameter of the pore is $5.0 \,\mu m$. Head is 40 mmAq.

rates occurring, but higher voltages are needed in comparison with the nickel membrane. Figure 6 shows the result obtained by using a zinc plate as a negative pole and a silver plate as a positive pole instead of stainless plates. It is seen that this combination of poles makes the flow rate zero faster than for the stainless plates. Next, the NaCl solution is replaced by KCl solution with other conditions unchanged and the result is shown in Fig. 7. The flow rate is decreased to the zero level but after a while it gradually increases again. This suggests that the block mechanism presumably caused with absorption and an electric effect (see Discussion), even if it is formed, is weak in KCl solution.

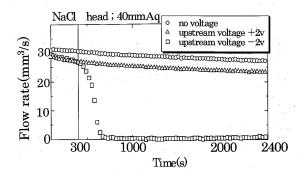


Fig. 6 Flow rate as a function of time for NaCl solution in water. Electric poles are a silver plate as plus pole and a zinc plate as minus pole. Nickel membrane is used and the diameter of the pore is $5.01 \,\mu$ m. Head is $40 \,m$ mAq.

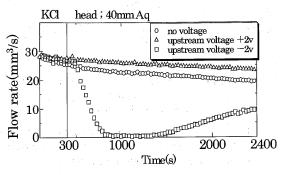


Fig. 7 Flow rate as a function of time for KCl solution in water. The experimental condition is the same as in Fig. 6.

3.2 Micropump

The set of experiment consists of three parts; Fluid drive part, Liquid separation part and Flow rate measurement part, as indicated in Fig. 8. The fluid drive part and the flow rate measurement part are the same as in the experiment of flow control. But we added the liquid separation part in this experiment which replaces the liquid for getting driving forces by the liquid which we want to transport. That is, the driving liquid (Polystyrene latex colloidal dispersion or NaCl and KCl solutions in water) is led from the fluid drive part to a cylindrical vessel of the liquid separation part, in which silicon oil is filled in the upper region and connected to another cylindrical vessel. In the lower region of the another vessel a transporting liquid (in this case water) is filled and led to the weighing cup on the digital balance. After adjusting the liquid levels in head tank and in weighing cup to the same level and attaining the condition of zero flow rate, we applied a voltage difference between the both sides of the membrane and measured the generated flow rate and the pressure differential. Figure 9 (a) and (b) show the result obtained by NaCl and KCl solutions with the voltage of 4V. Application of voltage begins at the time of 300 seconds, and we see the case of no membrane gives no effect, but the membranes of pore size of 5.8 and 3.9 µm generate the flow directing to the minus pole and the flow rate around 2 ml/s. It is seen that the flow rate is slightly greater in

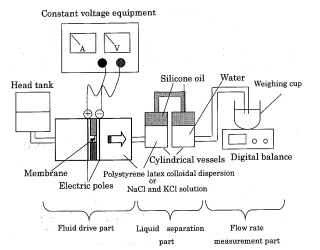


Fig. 8 Fabrication for the experiment of micropump

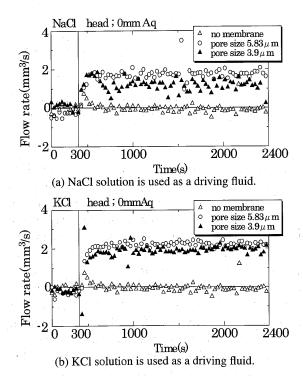


Fig. 9 Flow rate as the pump output plotted against time. The voltage of 4 V is applied after the time of 300 sec. Electric poles are stainless plates. Nickel membrane is used and the diameter of the pore is 5.83 μm or 3.9 μm.

KCl solution than in NaCl solution, but for these solutions bubbles were generated.

Also we used as a driving liquid the polystyrene latex colloidal dispersion in water of 0.1% in volumetric concentration, where the diameter of the latex particle is $0.12 \mu m$. Voltages used are four degrees ranging from 3.0 to 10.0 V. The results are shown in Fig. 10: Each voltage applied at the time of 300 seconds generates the flow directing to the minus pole and increasing with voltage. We notice some fluctuation in the data of flow rate with time, but this is considered not to be characteristic of the flow

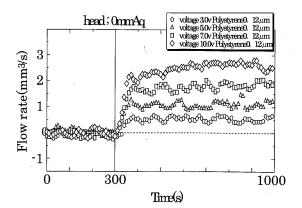


Fig. 10 Flow r We us colloid concen is 0 12

Flow rate as the pump output plotted against time. We used as a driving liquid the polystyrene latex colloidal dispersion in water of 0.1% in volumetric concentration, where the diameter of the latex particle is $0.12 \,\mu$ m. Voltages from 3 V to 10 V are applied after the time of 300 sec. Electric poles are stainless plates. Nickel membrane is used and the diameter of the pore is $5.01 \,\mu$ m.

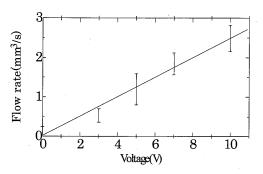
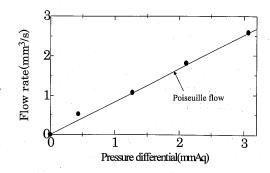


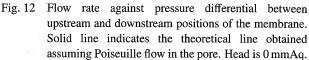
Fig. 11 Flow rate as the pump output plotted against the voltage.

but due to some noise given to the balance used, because it is seen to exist before applying voltages. Figure 11 gives a correlation between the voltage and the flow rate for the same data as in Fig. 10 in the steady state after a long time has passed since applying voltages, and we see a linear relationship between the two. Only the data of one concentration (0.1%) is shown here, but we confirmed by experiments that variation of concentration hardly gives influence on the characteristic of flow rate. Because no bubble is generated and there is a linear relationship between the flow rate and the voltage, polystyrene latex colloidal dispersion in water is thought to be a promising driving liquid.

Figure12 shows the flow rate against the pressure differential measured between upstream and downstream positions in the channel together with a theoretical line which was obtained by assuming Poiseuille flow for the flow through the pores in the membrane. It is seen that the experimental data almost agree with the theoretical line, and this tells us that the pressure differential generated by the applied voltage is consumed exclusively for the flow

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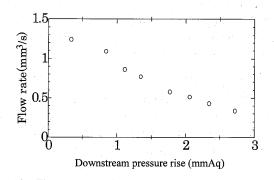


Fig. 13 Flow rate against the pressure applied at the downstream position. This corresponds to the pump performance.

through the pores in the membrane, because the water levels in the upstream head tank and in the downstream cup on the balance are nearly the same in the present experimental condition.

Furthermore, flow rates were measured under the condition of rising the pressure in the downstream region, and the result is shown in Fig. 13, which corresponds to a pump performance. We see that the flow rate decreases monotonously with increasing the pressure and the absolute value of pressure is low compared with those of past experiments⁽²⁾⁻⁽⁵⁾. But the downstream pressure can be made higher by increasing the voltage applied.

4. Discussion

4.1 Electrophoresis and electro-osmosis

If the present micropump effect is generated mainly by electrophoresis, mobility of a charged particle in an electric field m is given by dividing the ionic equivalent conductivity L by Farady constant $F^{(7)}$. And the mobility of Na⁺ is given by

$$m=L/F$$

=(50.1 cm² · Ω⁻¹)/(9.65×10⁴ C)
=5.19×10⁻⁴ cm² · s⁻¹ · V⁻¹

Now the applied voltage is 4 V in one example of the present experiments, and the electric field is approxi-

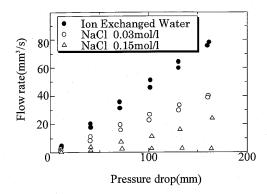
mately given by $2.0 \text{ V} \cdot \text{cm}^{-1}$. Therefore we have a velocity of Na⁺ particle as 1.04×10^{-2} mm \cdot s⁻¹ from the above equation. On the other hand, the velocity obtained by the present experiment of micropump is about $1.4 \text{ mm} \cdot \text{s}^{-1}$. This great difference between both the velocities shows that the pump effect seems not to be accounted for by the electrophoresis theory. On the other hand, the measure of thickness of electric double layer, Debye length λ_D , is given by the relationship for NaCl solutions⁽⁸⁾; $\lambda_D = 0.304 / \sqrt{[\text{NaCl}]}$ nm, where [NaCl] presents the concentration (mol $\cdot l^{-1}$). In the present experiment, [NaCl] is given as $0.154 \text{ mol} \cdot l^{-1}$ and we have $\lambda_D = 1.97 \text{ nm}$. This predicted length is very thin and it is questionable for the electric double layer and therefore for the electro-osmosis to play a main role in the flow through the hole of the diameter of $5\,\mu\text{m}$. Nevertheless if it is a main cause, the velocity v is given by the Helmholtz-Smoluchowski equation⁽⁹⁾ $v = \frac{\varepsilon E\varsigma}{\mu}$, where ε is the dielectric constant, E is the electric field, ς is the zeta potential and μ is the viscosity. At the present stage, ς is unknown and we cannot know the absolute value of v, but according to this equation vis proportional to the applied voltage. This is noticed in Fig. 11.

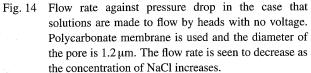
Also it should be noted that the flow rate encountered in the experiment of flow control is one order of magnitude greater than the output flow rate in the experiment of pump, as indicated by Figs. 3 (b) and 9 (a), although the flow control is restricted to several cases, that is, all cases of NaCl solutions examined and one case of KCl solution shown in Fig. 7. This discrepancy in flow rate between the flow control and the micropump is also open to question.

Now the flow rate of the present micropump is compared with those of the several reported micropumps. For comparison, the flow rate has been expressed in the style of the flow rate per unit voltage and unit area performance of the pump; $0.000 6 (ml/min/)V/cm^2$ was reported by Gan et al.⁽³⁾, $0.001 (ml/min/)V/cm^2$ by Zeng et al.⁽⁴⁾ and $0.03 (ml/min/)V/cm^2$ by Yao et al.⁽⁵⁾, while it is 0.015(ml/min/)V/cm² for Polystyrene latex colloid dispersion shown in Fig. 11, and $0.03 (ml/min/)V/cm^2$ for NaCl and KCl solutions of Fig. 9 (a) and (b) in this experiment. Then the present method belongs to the highest class with respect to the flow rate among the methods presented so far.

4.2 Absorption and its block mechanism

Comparing the cases of no voltage in Fig. 2 for DW with those in Figs. 3, 4 and 5 for NaCl and KCl solutions, we notice that gradual decreases in flow rates with elapse time are greater in NaCl and KCl solutions than in DW. This suggests that the flow causes some absorption of ions to the wall of pores, even if any voltage is not applied. Concerning this, we conducted an experiment of the pressure drop against the flow rate through smaller pores for DW and NaCl solution without application of voltage dur-





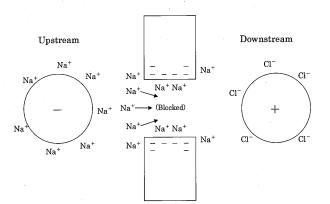


Fig. 15 Block mechanism of Na⁺ ions at the aperture

ing the time of 900 seconds after starting flow. The result is given in Fig. 14, where the membrane used is a sheet of polycarbonate and the diameter of the pore is $1.2 \,\mu\text{m}$. We see NaCl solutions provides higher resistance than DW at the same flow rate, which may be due to absorption.

Usually plus ions in a liquid are attracted to the surface of a plate immersed in the liquid, because the surface of most substances wears minus electric charges. In addition, plus ions and minus ions in the liquid are attracted to the negative and positive poles respectively by applying voltages (see Fig. 15 for a NaCl solution). When the fluid flows, the Na⁺ ions gathered around the minus pole on the upstream side also flow toward the aperture, but there already exist Na⁺ ions around and in the aperture and the coming Na⁺ ions will be blocked by the existing Na⁺ ions and consequently the flow is stopped at the aperture. If the pole on the upstream side is positive, Cl⁻ ions gather around the positive pole and flow to the aperture as Na⁺ ions do in the above mentioned case of the upstream negative pole. But in this case the hydrated radius of Clion is smaller than that of Na⁺ ion as shown in Table 1, and therefore it can pass through the aperture which is made narrow with the attached Na⁺ ions. K⁺ ions also can pass through the aperture because they are smaller than

ion	H_2O^+	Na ⁺	K+	C1-
Bare ion radius(nm)	—	0. 095	0. 133	0. 181
Hydrated radius(nm)	0. 28	0.36	0. 33	0. 33

Na⁺ ions as shown in Table 1. This is an explanation of the present experimental result of flow control. Also the micropump effect is thought to be caused with the plus ions or the plus ion charged colloid particles which proceed to the minus pole from the plus pole separated by the membrane. Electro-osmosis or electrophoresis may be concerned with this process.

5. Concluding Remarks

Micro-flow control and micropump was experimentally investigated by a new and simple method applying an electric field on the flow through a porous membrane. It is clarified that only NaCl solution in water is effective for the control of micro-flow among the liquids examined but KCl solution in water and a colloidal dispersion in water of polystyrene latex particles as well as NaCl solution are good as working fluids of the micropump. Although the applied voltages are less than 10 V, which is one order of magnitude lower than those in current experiments, performance of the present micropump is found belonging to the highest class in the past ones. Effects of electro-osmosis, electrophoresis and absorption were discussed about whether or not they may become causes of the present phenomena and it is concluded that absorption can be the most possible cause. The colloidal dispersion of polystyrene latex particles is a good working liquid of micropump, because it generates the flow rate proportional to the applied voltage without any bubble of by-product.

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