Selection of a Liquid for Swelling the Gel Coating of Electrodes for Electrohydrodynamic Pumping

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ABSTRACT: We investigated an electrohydrodynamic pump that used a gel-coated electrode. An organic liquid that swelled the gel was also used as the working fluid. In this study, we examined 12 liquids. The following properties were found to be preferable for the pumping: (1) the liquid can significantly swell the gel, and (2) it can flow at a high velocity when the electrode is not coated. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 98: 1996–2002, 2005

Key words: gels; silicones; swelling

INTRODUCTION

Electrohydrodynamics has been widely studied over the past decades.[1,2] An example of its applications is an ion-drag pump. The simple arrangement of this pump consists of a pair of electrodes immersed in a dielectric fluid. A high direct-current (dc) voltage applied to these electrodes makes the fluid flow. Early studies in this field were reported by Stuetzer,[3] Pickard,[4] and Jorgenson and Will.[5] Later, there were many studies of this pumping technique that included the geometry of the electrode,[6–8] the selection of effective fluids,[9] and applications for micropumps.[10–12]

The fluids used in such electrohydrodynamic (EHD) pumps have mainly been organic liquids. However, water is a common solvent in biotechnologies. An obstacle to pumping water is its electrolysis. For example, Richter et al.[10] tried to pump water with their ion-drag pump. However, they failed to pump the water because gases were generated by the electrolysis of the water.

There can be similar problems that arise during liquid–liquid extraction, which is aided by EHD phenomena.[13,14] When the extraction system has a continuous water phase, the high conductivity of the water can cause various problems. Therefore, such an extraction has been limited to systems that have a continuous organic phase with dispersed water drops.

The electrolysis of water occurs on the electrodes. Therefore, it can be suppressed by a solid material that separates the water from the electrode. For example, plastics and rubbers can be used for this purpose. However, they also prevent ion-drag pumping because the electrode (emitter) needs to be in contact with the fluid to supply ions to the fluid.[3]

Therefore, we tried gels as a new class of separating materials. We coated an electrode with a gel and discovered that the EHD pumping was not prevented by the gel coating.[15] Because the gel contained a large amount (>95 wt %) of an organic liquid, its electrical properties were expected to be similar to those of the liquid. We concluded that the EHD pumping was, therefore, not prevented. In this study, we examined various organic liquids and found that some properties improved the flow velocity of EHD pumping through a gel-coated electrode.

EXPERIMENTAL

Experimental setup

The experimental setup is shown in Figure 1. It had a circular channel in which a pair of electrodes were placed on the bottom. The channel was made of Teflon plates. The electrode was a thin gold film (ca. 27 nm thick) deposited onto a Teflon plate by ion sputtering. The application of a dc high voltage to the electrode was carried out with a Glassman ER6R50 high-voltage instrument (High Bridge, NJ).

Preparation of the gel coating

The electrodes were coated with a gel, which was prepared by a hydrosilylation method. The raw materials were vinyl-terminated polydimethylsiloxane
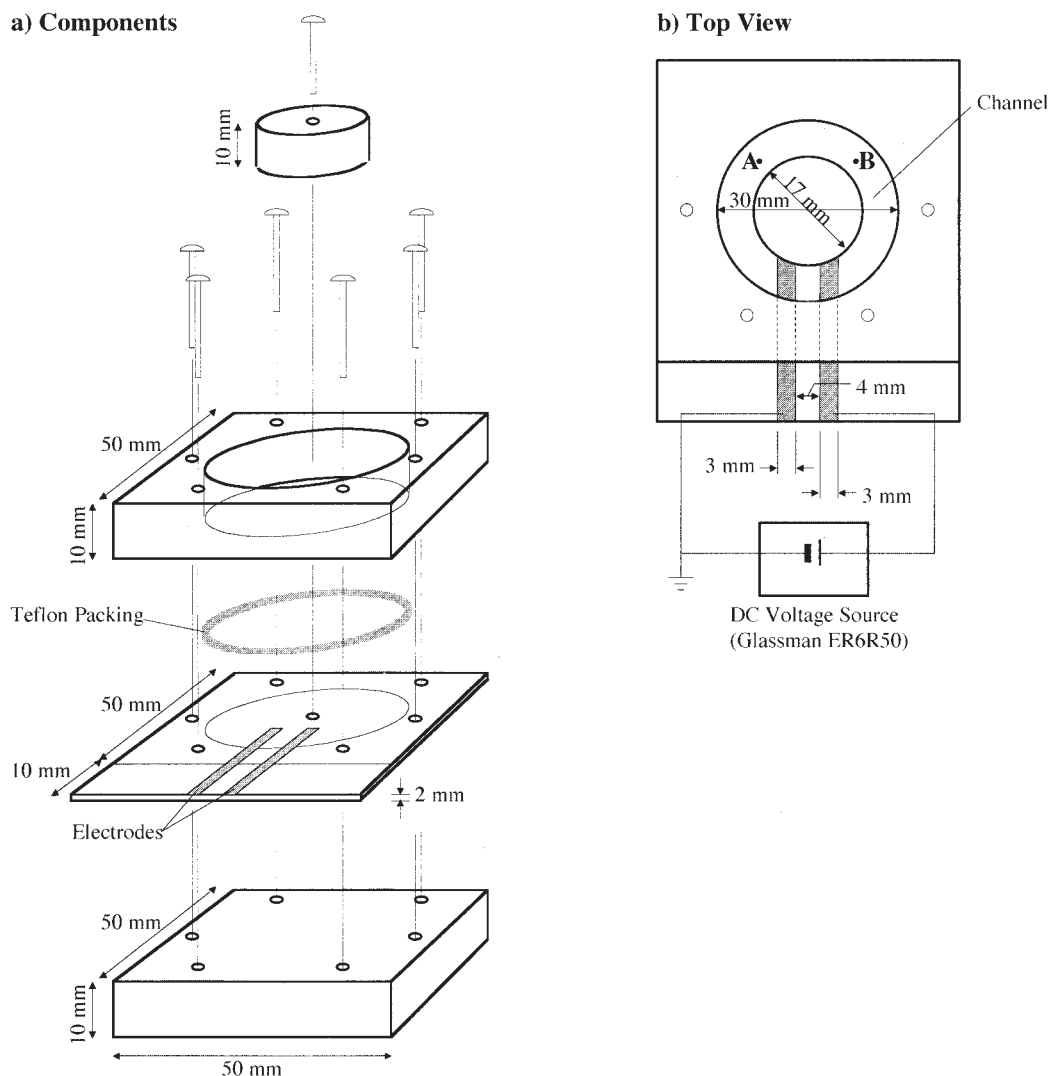
a) **Components**

b) **Top View**

Figure 1 Experimental setup for observing the electrohydrodynamic flow: (a) components and (b) top view of the setup.

(0.30 g, molecular weight $\approx$ 28,000; DMS-V31, Gelest, Inc., Morrisville, PA), a 10 wt % xylene solution of a methylsiloxane–dimethylsiloxane copolymer (0.14 g; HMS-301, Gelest), a 1.0 wt % isopropyl alcohol solution of hydrogen hexachloroplatinate(IV) hexahydrate (0.04 g; Wako Pure Chemical Industries, Osaka, Japan), and a low-viscosity silicone oil (5.00 g; KF96-30, Shin-Etsu Chemical, Tokyo, Japan). A mixture (0.48 g) of these materials reacted in the channel (Fig. 1) at 75°C for 3 h and formed a silicone resin softened by the low-viscosity silicone oil. The channel was then filled with benzotrifluoride (4.0 g) for 48 h, which was replaced with fresh benzotrifluoride every 12 h. The silicone oil in the resin was replaced with benzotrifluoride by this treatment. The obtained gel swollen with the benzotrifluoride weighed 0.930 g. The benzotrifluoride concentration in the gel was 97.0 wt %. The thickness of the gel coating was estimated to be 1.6 mm on the basis of the weight and the density of the gel.

Gels swollen with other organic liquids were also prepared. For example, a silicone gel swollen with *n*-decane was prepared as follows. First, the gel swollen with benzotrifluoride was prepared in the channel by the previously described procedure. The benzotrifluoride was replaced by *n*-decane, which filled the channel for 36 h, although the *n*-decane was replaced with fresh *n*-decane every 12 h. The obtained gel weighed 0.632 g and contained 95.7 wt % *n*-decane.

Measurements of the flow velocity

The organic liquid as the working fluid was poured into the channel. Its volume was 1.44 mL, which corresponded to a 3.0-mm depth (Fig. 2). We added a few pieces of human hair (ca. 0.1 mm long) as tracers to visualize the flow. The tracers floated on the surface of the working fluid. The flow velocity was measured during the application of a dc voltage to the electrode.
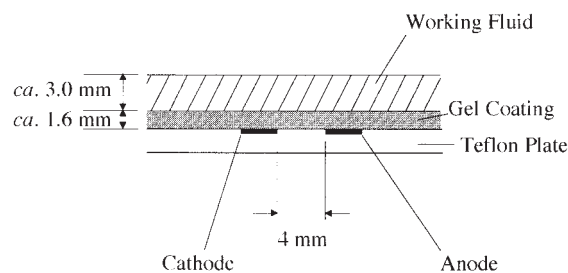
Figure 2 Side view of the gel-coated electrode.

However, the electric field around the electrode could affect the movement of the tracers. One should minimize this effect by measuring the velocity in the region farthest from the electrodes. Therefore, we measured the time taken for the tracer to move between points A and B, as shown in Figure 1(b). Although the velocity measured by this method did not represent the average velocity of the entire fluid layer, we could compare the velocities of the fluids because we used a constant volume of each fluid.

RESULTS AND DISCUSSION

A typical arrangement used in this study included a pair of electrodes, a gel coating, and a working fluid (Fig. 2). The gel coating was made of silicone gel swollen with an organic liquid. The working fluid was the same liquid used for swelling the gel. A high dc voltage applied to the electrode caused an EHD flow of the working fluid. We have already reported an example of such a flow using xylene.[15] The xylene was the liquid for swelling the gel coating and was also the working fluid. In this study, we tried to use various organic liquids for this fluid flow.

First, we compared nitrobenzene with benzotrifluoride. Nitrobenzene poorly swelled the gel. The nitrobenzene concentration in the gel was only 11.3 wt %. However, the benzotrifluoride significantly swelled the gel; it was 97.0 wt % of the gel. Figure 3(a) shows the flow velocity of nitrobenzene when the electrode was coated with the gel. The nitrobenzene did not flow even with the application of a voltage (400–1000 V). However, when the electrode was not coated, the nitrobenzene flowed, as shown in Figure 3(b). Thus, the gel coating totally prevented the flow of nitrobenzene. However, the velocity of benzotrifluoride was not prevented by the gel coating, as shown in Figure 3(c,d). (However, the benzotrifluoride flow was not stable. It was also not durable.) Thus, the gel that was quite swollen did not prevent the flow. Similar results were also obtained with other gels that were quite swollen: a silicone gel swollen by xylene[15] and a poly-(vinyl alcohol) gel swollen by dimethyl sulfoxide. Therefore, a high swelling ratio can be a preferable property for a gel coating.

As mentioned previously, the benzotrifluoride gel did not prevent the fluid flow, although the nitrobenzene gel prevented it. The following is a possible mechanism to explain this difference. For a very swollen gel such as the gel swollen with benzotrifluoride, charges can be injected from the electrode into the gel and move along the lines of electric force (Fig. 4) because the gel contains a large amount (97 wt %) of liquid and its electrical properties can be similar to those of the liquid. Although they move through the gel, some of them then go into the working fluid, and this causes the fluid to flow. We proposed this mechanism in an earlier article.[15] However, the gel swollen with nitrobenzene contained only 11.3 wt % nitrobenzene. The major component of the gel was silicone, which is a typical insulating material. Therefore, the mobility of the injected charge was expected to be very low. That is, the gel prevented the charge from going into the working fluid. As a result, the fluid did not flow. In addition, because the poorly swollen gel will have a much lower permittivity than the working fluid, the electric field in the fluid layer can be very small. Therefore, the fluid flow will be very slow if the charges go into the fluid layer.

Next, we compared benzotrifluoride with *n*-decane. Both liquids significantly swelled the gel. The concentrations of these liquids in the gel were 97.0 wt % for benzotrifluoride and 95.7 wt % for *n*-decane. However, the velocities were quite different. That is, benzotrifluoride flowed because of the application of voltages ranging from 400 to 1000 V, although the electrode was coated with the gel [Fig. 3(c)]. However, *n*-decane did not flow under voltages of the same range [Fig. 3(e)]. Thus, *n*-decane was not preferable for swelling the gel coating despite the high swelling ratio. We also measured the velocity with the non-coated electrode. As a result, benzotrifluoride showed a significant velocity [Fig. 3(d)], but *n*-decane showed a very low one [Fig. 3(f)]. These results were similar to those measured when the electrode was coated with the gel. Therefore, another preferable property of the gel coating can be as follows: the liquid, which is used as both the working fluid and the swelling solvent of the gel, should show a high velocity even when the electrode is not coated.

As stated previously, *n*-decane did not flow by the application of a voltage despite the high swelling ratio of the gel. The reason can be explained as follows. Generally, during ion-drag pumping, charge injection from the electrode into the working fluid is necessary to cause the working fluid to flow.[3] This injection is also needed for the gel-coated electrode, as previously mentioned (see Fig. 4). However, *n*-decane did not flow even when the electrode was the noncoated one. This means that no charge (or a very small amount) was injected into the fluid, and this suggests that almost no charge was also injected into the gel when
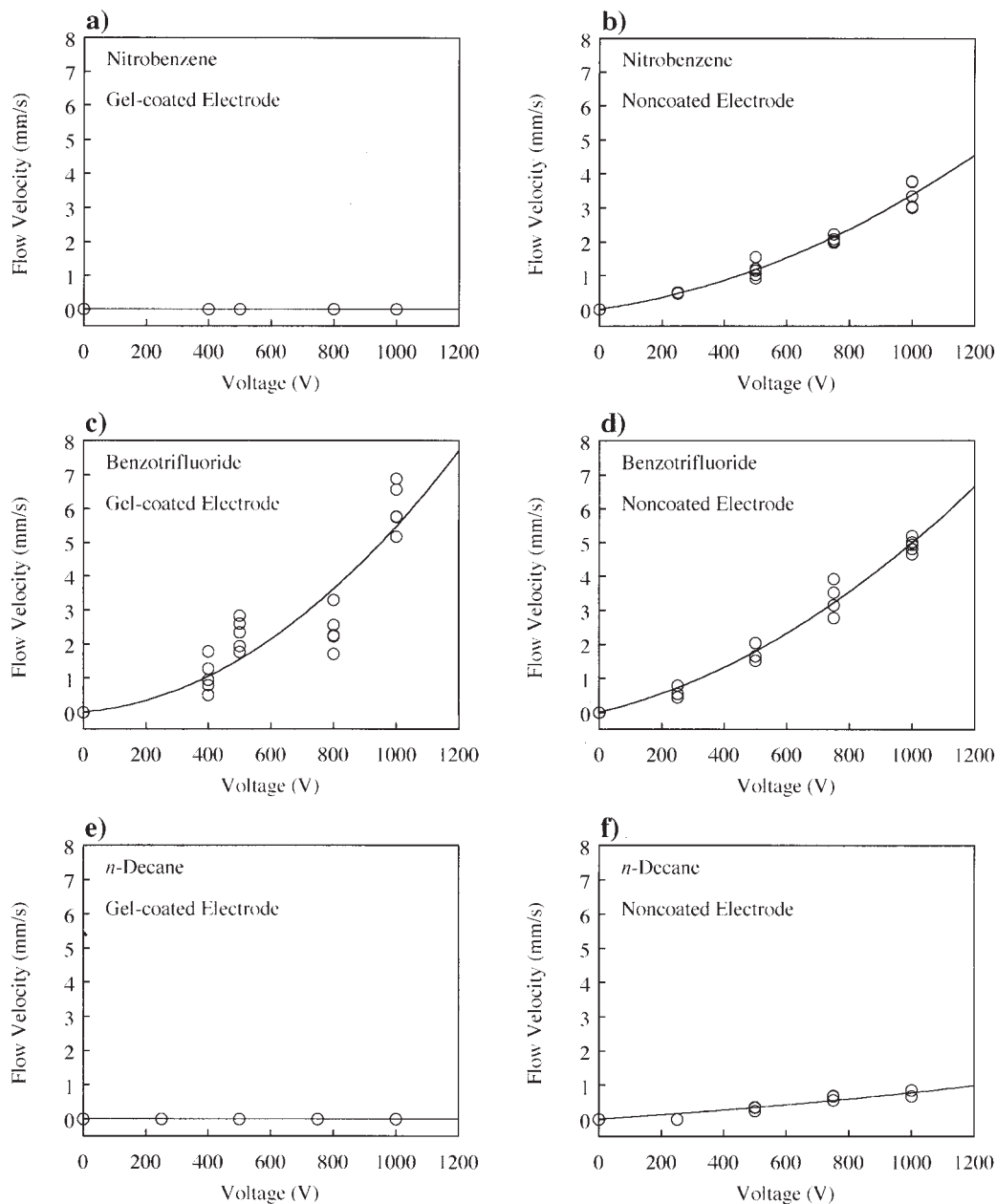
Figure 3 Flow velocities of nitrobenzene, benzotrifluoride, and *n*-decane as the working fluids. The electrode was (a,c,e) coated with the gel or (b,d,f) not coated.

the gel-coated electrode was used. Therefore, *n*-decane did not flow when a voltage was applied to the gel-coated electrode despite the high swelling ratio of the gel.

On the basis of these results, we examined organic liquids of various chemical structures that included an aliphatic hydrocarbon, aromatic hydrocarbon, alcohol, ether, ketone, and ester, as shown in Table I. We selected these liquids, which had boiling points higher than 100°C, because they were easily handled without significant losses due to evaporation.

We first examined these liquids in terms of the swelling ratio. We immersed a strip of silicone rubber
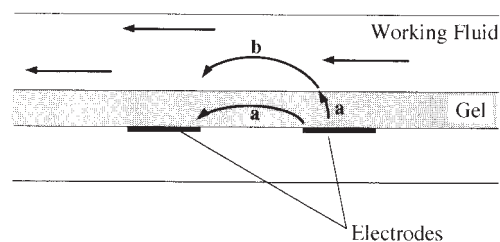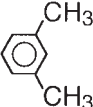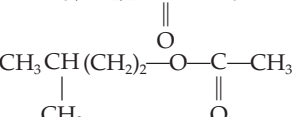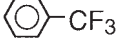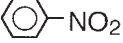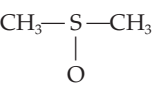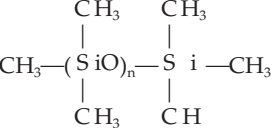

Figure 4 Possible mechanism of fluid flow caused by the application of a voltage with a gel-coated electrode. Arrow a indicates charges moving through the gel; arrow b indicates charges moving through the working fluid.

TABLE I
Examined Organic Liquids

Compound	Structure	Boiling point (°C)
n-Decane	$CH_3(CH_2)_8CH_3$	174.1
m-Xylene	(structure: benzene ring with two CH_3 groups)	139.1
1-Pentanol	$CH_3(CH_2)_4—OH$	138.0
Dibutyl ether	$CH_3(CH_2)_3—O—(CH_2)_3CH_3$	142.4
2-Heptanone	$CH_3(CH_2)_4—C—CH_3$, with $\|$ O below	150.2
Isoamyl acetate	$CH_3CH(CH_2)_2—O—C—CH_3$, with CH_3 and O below	142.0
1,2-Dichlorobenzene	(structure: benzene ring with two Cl groups)	180.5
Benzotrifluoride	(structure: benzene ring with CF_3)	102.1
Nitrobenzene	(structure: benzene ring with NO_2)	210.9
Dibutylamine	$CH_3(CH_2)_3—N—(CH_2)_3CH_3$, with H below	159.6
Dimethyl sulfoxide	$CH_3—S—CH_3$, with O below	189.0
Silicone oil (Shin-Etsu, KF96L-5)	$CH_3—(SiO)_n—Si—CH_3$, with CH_3 and CH_3 above, CH_3 and CH below	130 (10 mmHg)

* Scan r001-r004 at 100%

(20 × 5 mm, 0.5 mm thick) in each liquid and measured the swelling ratio. The ratio was evaluated with the following equation:

$$\text{Swelling ratio}(\%) = [(L^3 - L_0^3)/L_0^3] \times 100$$

where L_0 is the initial length of the strip and L is the length when the swelling reaches its maximum. We used the silicone rubber instead of the silicone gel for experimental convenience. Because the polymer chains (poly(dimethylsiloxane)) of the rubber and the

TABLE II
Properties of Various Organic Liquids

Compound	Swelling ratio (%)	Permittivity[a]	Flow velocity (mm/s)	Dynamic viscosity (mPas)[a]
n-Decane	150	2.0	0.72	0.93 (20°C)
m-Xylene	139	2.4	1.57	0.58 (25°C)
1-Pentanol	16	13.9	0	3.31 (25°C)
Dibutyl ether	199	3.1	0	0.60 (30°C)
2-Heptanone	112	9.8	3.55	0.80 (20°C)
Isoamyl acetate	125	4.6	2.10	0.87 (20°C)
1,2-Dichlorobenzene	50	6.8	2.13	1.32 (25°C)
Benzotrifluoride	146	9.1	4.92	0.89 (20°C)
Nitrobenzene	6	34.8	3.34	2.01 (20°C)
Dibutylamine	201	3.0	0.85	0.95 (20°C)
Dimethyl sulfoxide	5	48.9	—	2.00 (25°C)
Silicone oil (Shin-Etsu, KF96L-5)	104	2.6	0	4.58 (25°C)

[a] These data were extracted from ref. [16]
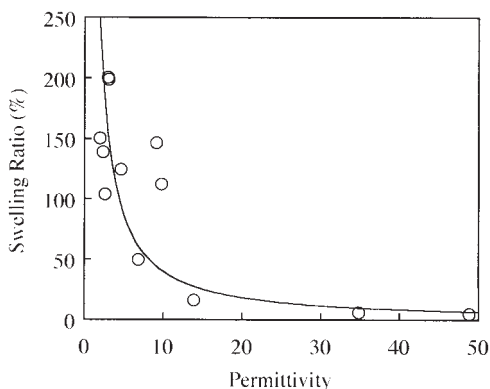
Figure 5 Swelling ratio versus the permittivity.

Figure 7 Swelling ratio versus the flow velocity of various organic liquids.

gel were the same, the use of the silicone rubber can be valid as long as we compare only the swelling ratios among the liquids. The measured swelling ratios are shown in Table II. The permittivity and dynamic viscosity are also shown in this table. Liquids with a low permittivity, such as *n*-decane (2.0) and *m*-xylene (2.4), showed high swelling ratios. However, liquids with a high permittivity, such as nitrobenzene (34.8) and dimethyl sulfoxide (48.9), showed low swelling ratios. Figure 5 shows the swelling ratios versus the permittivity. The swelling ratio roughly correlated with the permittivity. That is, a lower permittivity gave a higher swelling ratio. This was reasonable because the permittivity of the silicone rubber was low ($\approx$3.1).

Next, we examined the liquids in terms of the velocity arising from the application of a voltage. We applied a voltage (1000 V) to each liquid, using the noncoated electrode, and measured the velocity (Table II). Liquids with a high permittivity, such as nitrobenzene (34.8) and benzotrifluoride (9.1), showed high velocities. [Dimethyl sulfoxide also had a high permittivity (48.9), but it burned when the voltage was applied, and the measurement ended in failure.]

According to a report by Crowley et al.,[9] the velocity (U) of a laminar flow in a wide slot between two planes can be described as follows:
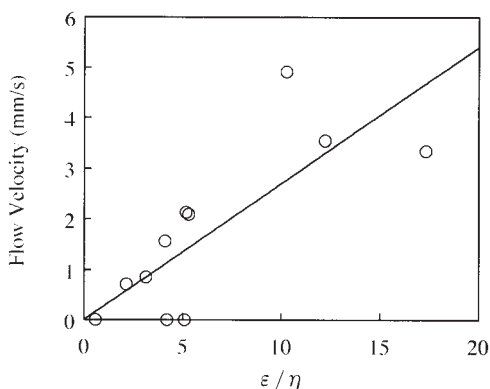
$$U \approx (E^2 d^2 / 12L)(\varepsilon / \eta)$$

where ε is the permittivity, E is the electric field, d is the height of the slot, η is the dynamic viscosity, and L is the length between the electrodes. Because our geometry of the channel and electrode was different from that of Crowley et al.'s report, we could not directly apply this equation to our case. However, we have plotted ε / η versus the velocity in Figure 6. The velocity roughly correlated to ε / η. This result agreed with Crowley et al.'s report, showing that a high permittivity and a low viscosity led to a high flow velocity.

As discussed previously, a liquid that significantly swelled the gel and showed a high flow velocity was preferable for the gel coating. Figure 7 is a scatter graph plotting the swelling ratio versus the velocity. The upper right of this graph is the preferable region for the gel coating. That is, benzotrifluoride, 2-heptanone, isoamyl acetate, and *m*-xylene were preferable liquids for the gel coating.

Thus, benzotrifluoride was a preferable liquid in terms of its swelling ratio and flow velocity. However, it was not durable, as mentioned previously. The flow
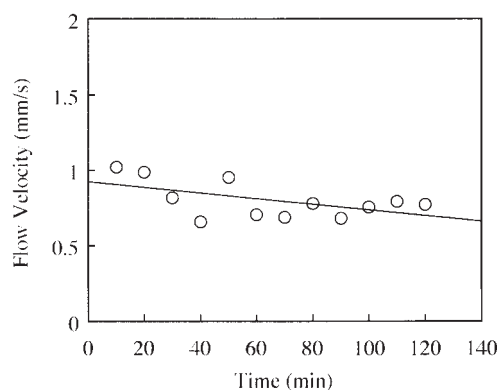

Figure 6 Flow velocity versus ε / η.

Figure 8 Flow velocity of *m*-xylene. A voltage (1000 V) was applied with the gel-coated electrode.

became unstable within several minutes. On the other hand, *m*-xylene was also a preferable liquid that showed a high swelling ratio and a high flow velocity. It could steadily flow for more than 2 h, as shown in Figure 8. It was much better than benzotrifluoride in terms of durability. Other halogenated compounds did not have very good durability. This might come from the electrochemical decomposition of these compounds by the applied voltage.

CONCLUSIONS

The EHD pumping of an organic liquid is possible even when the electrode is coated with a gel. The liquid acts as both the working fluid and the swelling solvent of the gel. The followings properties of the liquid are preferable for obtaining a high flow rate: (1) the liquid can significantly swell the gel coating, and (2) the liquid can flow at a high velocity when the electrode is not coated. When the liquid has both properties, a high flow rate can be obtained.

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