



## Flexible Fluid Pump Using Magnetic Composite Gels

Tetsu Mitsumata,<sup>1\*</sup> Yuki Horikoshi,<sup>1</sup> Jun-Ichi Takimoto<sup>1,2</sup>

<sup>1\*</sup>Department of Polymer Science and Engineering, Graduate School of Engineering, Yamagata University, Yonezawa 992-8510, Japan; fax +81(0).238.26.3101; email: [tetsu@yz.yamagata-u.ac.jp](mailto:tetsu@yz.yamagata-u.ac.jp)

<sup>2</sup>Japan Science and Technology Agency, CREST, Japan.

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**Abstract:** A flexible fluid pump made of magnetic composite gels has been developed. The magnetic composite gel was sodium alginate or poly(vinyl alcohol) gel in which barium ferrite particles were dispersed. The pump consisted of a magnetic gel-rotor and a driving magnet. The rotor demonstrated the rotational motion in response to a rotational magnetic field generated by the driving magnet. The gel-rotor had the shape of a screw so as to deliver water efficiently. The diameter of the rotor was varied from 3.8 to 5.5 mm and the diameter of the tube for delivering water was kept as 6 mm. Flow rates pumped by the rotor increased with increasing the diameter, length, and rotation speeds of the rotor. The maximum flow rate of 34 ml/min was achieved when the rotation speeds was 5000 rpm. The rotor with a diameter of 300  $\mu\text{m}$  was also successfully synthesized, however the rotor did not deliver water in a microtube with a diameter of 500  $\mu\text{m}$ . Processing of the gel-rotor, and the efficiency of the water delivery are presented.

### Introduction

The polymer gel has been positively used to fabricate biomimetic actuators or machines in the next generation. For example, poly[2-(acrylamido)-2-methylpropanesulfonic acid] gel of an ionic polymer gel undergoes bending motion under AC electric fields. The gel is called gel-looper [1] or gel-eel [2], and has been predicted as a material for artificial muscles. Amphiphilic polymer gel swollen by an organic solvent shows a motion on water surface by spreading the organic solvent. A tetrahydrofuran-swollen gel equipped with a spouting hole shows a controlled translational motion with a velocity of 77 mm/s or rotational motion with a maximum speed of 400 rpm and a torque of  $10^{-9}$  Nm on the water surface [3, 4]. Polypyrrole film demonstrates rapid and intensive bending in the solid state induced by the reversible and anisotropic adsorption [5]. Photo reactive polymer shows a deformation by UV light. Liquid crystalline gel film containing freestanding azobenzene undergoes a significant and anisotropic bending toward the irradiation direction due to isomerization [6]. Recently, it has been reported that the long-range locomotion in air of the gel actuator consists of nonionic polymer gels and dielectric liquid pattern [7]. The driving force of the gel originates from the electrohydrodynamic flow of the dielectric solvent inside and outside the gel induced by the ion-dragging mechanism under the electric field. Thus, the actuators made of polymer gels have an advantage of showing biomimetic motion because the gels have a characteristic structure, which is similar to a living body.

In polymer gels, magnetic gel has attracted many researchers to fabricate biomimetic actuators in particular. It is because that the magnetic gel has advantages of fast

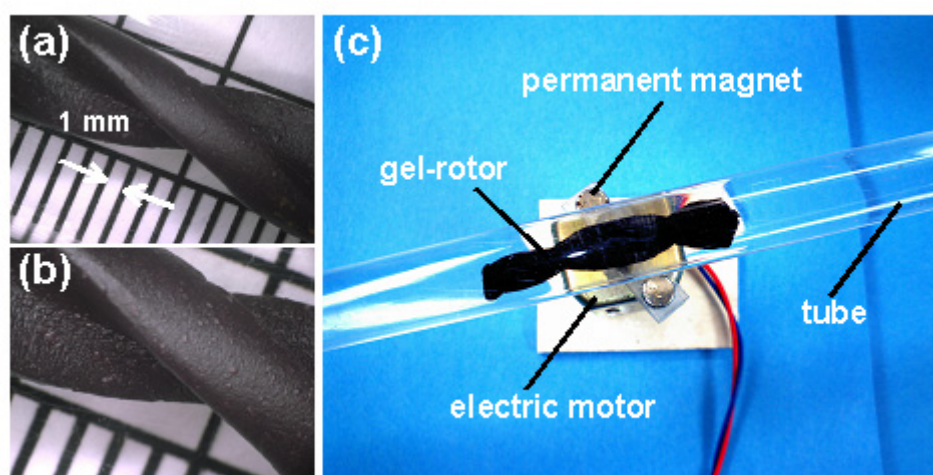
response, large deformation, and large stress generating by the large deformation. The magnetic gel consisted of poly(vinyl alcohol) (PVA) and magnetic fluids largely elongates under non-uniform magnetic fields [8, 9]. The elastic modulus of magnetic soft materials such as PVA-magnetic fluids [10], PVA-barium ferrite [11-13]  $\kappa$ -carrageenan-barium ferrite [14, 15] gels, and polyurethane-barium ferrite elastomers [16] demonstrates large modulus change due to magnetization. These actuations occur without any physical contact such as electrical leads of generators; this is also a great advantage of the magnetic gel.

As the magnetic gel has the advantages mentioned above, we have tried to fabricate a fluid pump using magnetic gels. Pumps made of polymer gels are rich in flexibility; therefore, it is appropriate to use in a winding watercourse. The pump that is not necessary for the physical contact would be useful for microfluidic applications. There are some papers demonstrating a pump made of polymer gels for microfluidic systems. The pump is made from fluid-responsive polymer particles of Poly(acrylic acid) [17]. Low-voltage electroosmosis pumps are developed using a gel salt bridge [18]. However, these pumps need a physical contact such as electrical leads to drive them.

In the present study, the processing technique of gel-molding was introduced to fabricate the fluid pump. Processability of magnetic gels, effects of the rotor size and rotation speed on the flow rates has been investigated.

## Results and discussion

Photographs of the gel-rotor and the driving magnet generating rotational magnetic fields are shown in Fig. 1. As seen in the photograph, the gel has a screw shape.

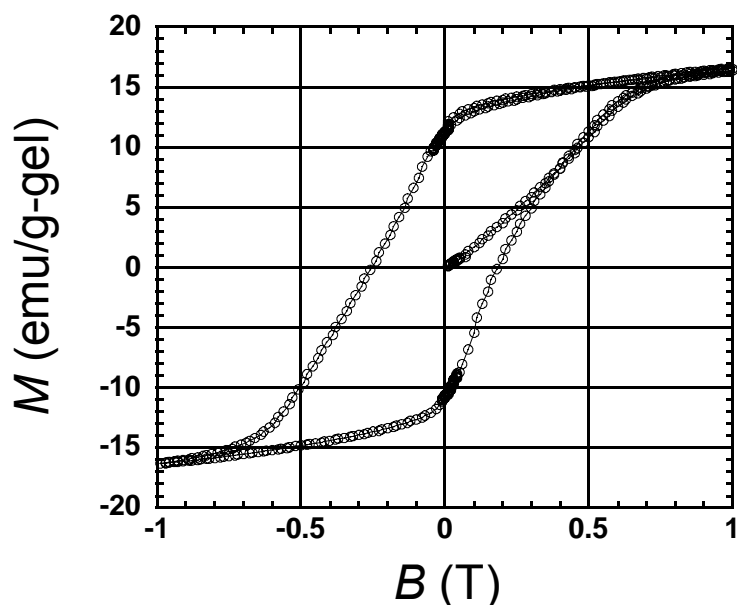


**Fig. 1.** Photographs of the gel-rotor (a, b) and the pump developed in this study (c). The driving magnet consists of a pair of permanent magnet and an electric motor.

Surfaces of the mold made of carrageenan gel were firmly imprinted on the gel-rotor. It is important for fabrication of the pump to process gel-rotors precisely. The gel-rotor demonstrated rotational motion in the tube in response to magnetic fields generated by the rotation of permanent magnets.

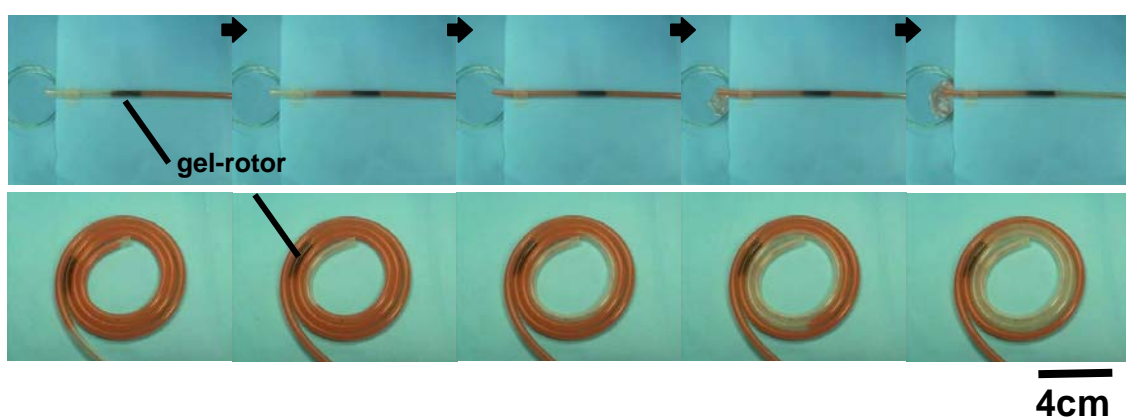
Fig. 2 shows the magnetization curve of the gel containing 30 wt.% of barium ferrite that is the same ferrite concentration of the rotor. The magnetization showed a large

hysteresis indicating the magnetic properties of ferromagnetic substances. The magnetization  $M$  increased as increasing the magnetic field  $B$ , and saturated to approximately 16 emu/g-gel at 1T. When the magnetic field was reduced to zero field, the magnetization had a remanent magnetization of 12 emu/g-gel. Due to the magnetic force between the remanent magnetization and the magnetic field generated by the permanent magnet, the rotor rotates in response to the rotational magnetic fields.



**Fig. 2.** Magnetic hysteresis curve of the gel containing barium ferrite particles (30 wt%).

Fig. 3 displays the photographs representing water-delivering motion of the gel-rotor. Two types of delivering motion in straight and winding tubes are presented.

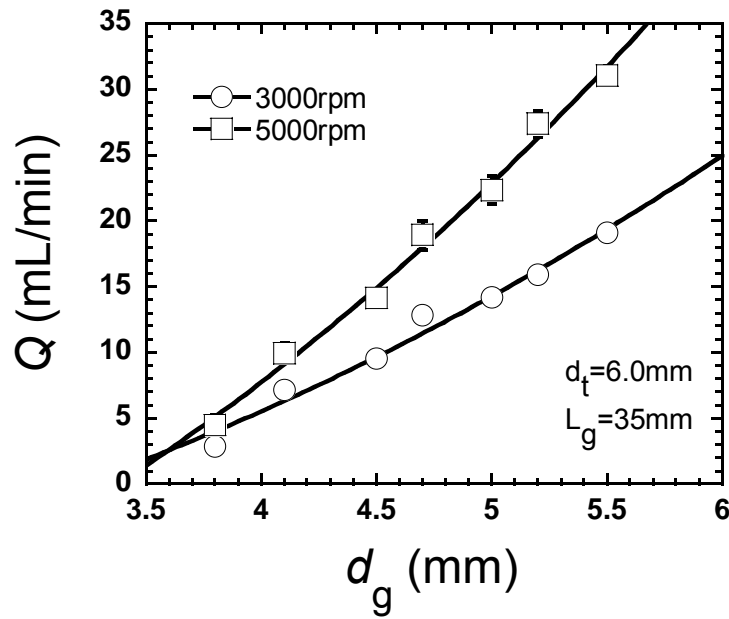


**Fig. 3.** Photographs representing water flow by the gel rotors in a straight tube (top photos) and in a spiral tube (bottom photos).

The water was dyed by a pigment with a color of orange. In the series of top photos, the gel-rotor located at the center showed a rotational motion in the straight tube. By the rotation, the water flowed to the laboratory dish in the left end. The bottom photos

indicate the gel-rotor also delivered water in a winding tube. The elastic modulus of the gel-rotor was approximately  $10^4$  Pa, therefore, the rotor can be bent in the tube. The photographs indicate the bent rotor can rotate in a winding tube.

The relationship between flow rate  $Q$  and diameter of the gel rotor  $d_g$  is shown in Fig. 4. The flow rate was calculated by the weight of water that was delivered during 10 sec. The rotation speed of the driving magnet  $\omega$  was calculated using the relationship between driving currents and rotation speed of the magnet. We assume that the rotation speed of the gel-rotor equals to that of the driving magnet placed under the rotor. The flow rate increased with increasing the diameter of the rotor as  $Q \sim d_g^2$ . The flow rate showed a threshold of the diameter that the rotor could not deliver water below the threshold. The threshold did not depend on the rotation speed and lied approximately  $d_g \sim 3.5$  mm corresponding to 58% of the diameter of the tube  $d_t$ . The ratio of the cross-section area of the gel-rotor to the tube  $(d_g/d_t)^2$  was 0.34 (=34%). Less than this ratio of diameter, it is considered that the shear force generated by the rotation of the rotor does not act enough on the water. This may be caused by the leakage from the narrow gap between the rotor and tube.

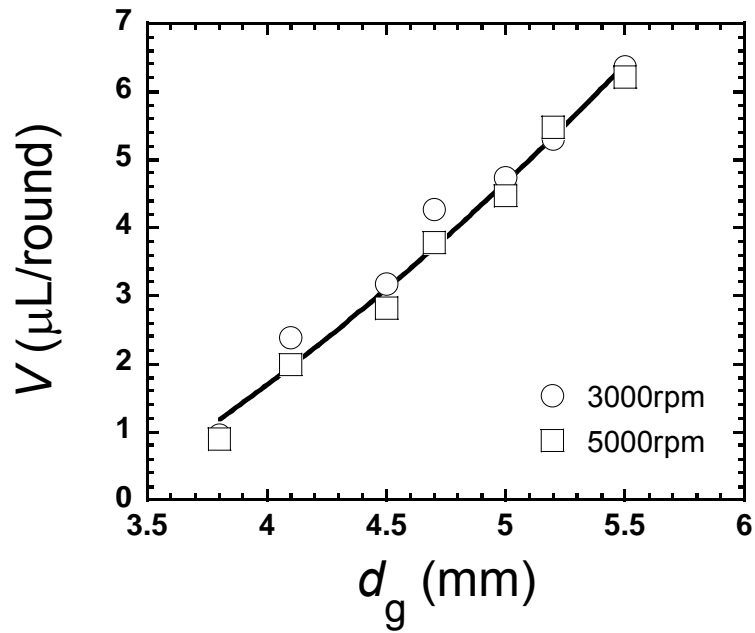


**Fig. 4.** Flow rate as a function of the diameter of rotor at rotational speeds of 3000 and 5000 rpm ( $d_t=6$  mm,  $L_g=35$  mm).

Fig. 5 shows the relationship between the volume of water delivered by one rotation  $V$  and the diameter of the gel-rotor  $d_g$ . The volume of water  $V$  was calculated by the following equation,

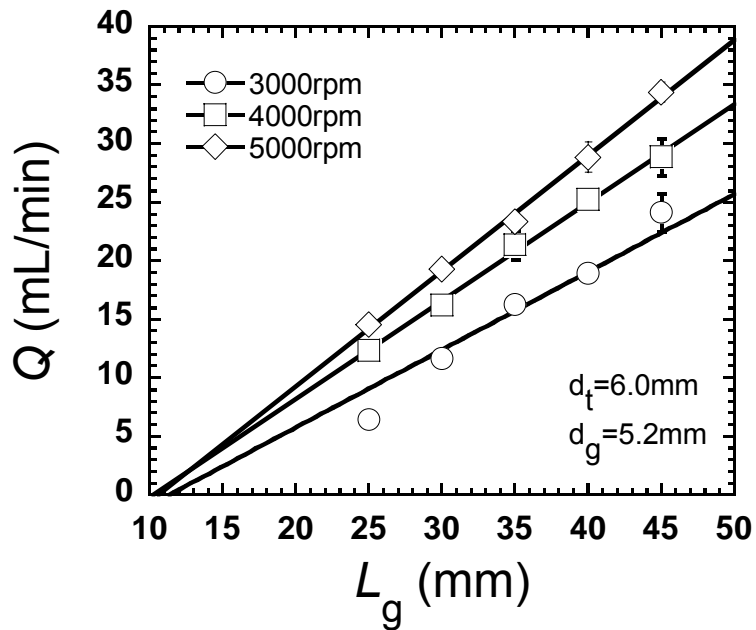
$$V = \frac{Q}{\omega} \quad (1)$$

Here,  $\omega$  is the rotation speed of the gel-rotor. The volume of water delivered by one rotation increased proportionally to the squares of the diameter of the gel-rotor, independently of the rotation speed. This strongly suggests that the efficiency for water delivery is invariable even in the high rotation speed such as 5000 rpm.



**Fig. 5.** Volume of water delivered by one rotation as a function of the diameter of rotor at 3000 and 5000 rpm ( $d_t=6$  mm,  $L_g=35$  mm).

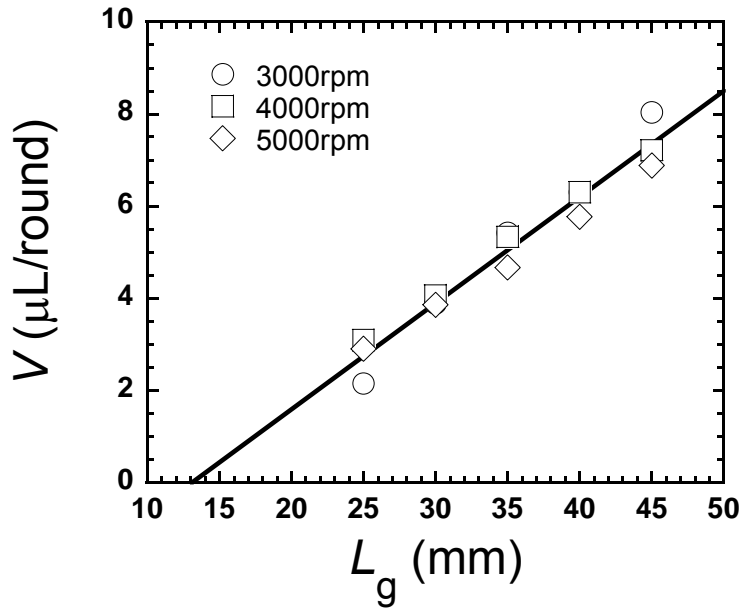
Fig. 6 shows the flow rate  $Q$  as a function of the length of the gel-rotor  $L_g$ . The flow rate increased in proportional to the length of the gel-rotor. The flow rate appeared a threshold not only in a diameter but also in a length of the gel-rotor. It was found that the threshold was independently of the rotation speed. The value of the threshold was estimated to be 11 mm, 10 mm and 11 mm when the rotation speed was 3000 rpm, 4000 rpm, and 5000 rpm, respectively.



**Fig. 6.** Flow rate as a function of the length of rotor at rotational speeds of 3000, 4000 and 5000 rpm ( $d_t=6.0$  mm,  $d_g=5.2$  mm).

This strongly suggests that the minimum length (~11 mm) is needed for the rotor to make water delivery. The periodic length of screw printed on the rotor was approximately 13 mm, which is nearly equal to the minimum length. This means that the periodic length of a ditch printed on the rotor is demanded for the water delivery.

Fig. 7 shows the relationship between the volume of water delivered by one rotation  $V$  and the length of the gel rotor  $L_g$ . The volume of water  $V$  was calculated by eq. (1). The volume of water delivered by one rotation increased proportionally with the length of the gel-rotor, independently of the rotation speed. As well as the diameter of the gel-rotor, the efficiency for water delivery was invariable under the rotation speeds of 3000 to 5000 rpm.



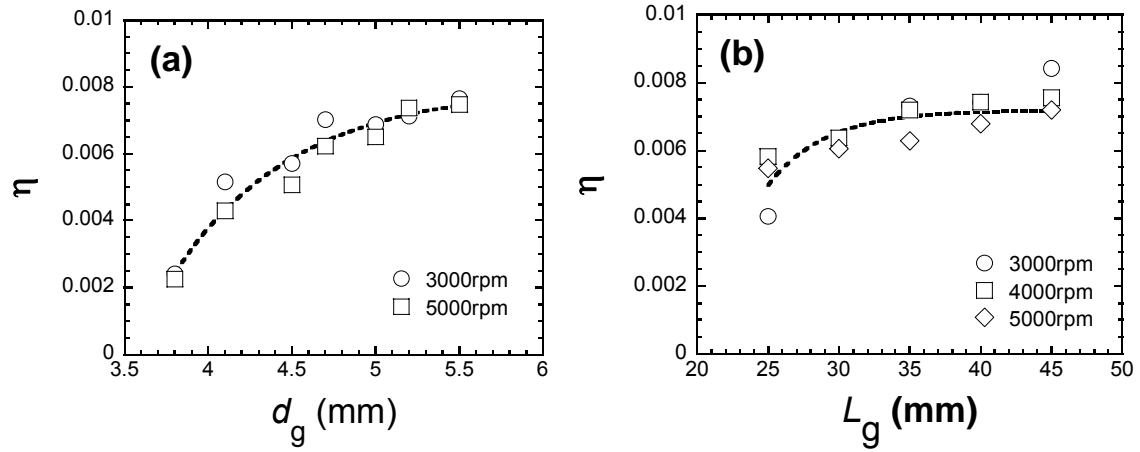
**Fig. 7.** Volume of water delivered by one rotation as a function of the length of rotor at 3000, 4000, and 5000 rpm ( $d_t=6.0$  mm,  $d_g=5.2$  mm).

The relationship between the efficiency of water delivery and the diameter or the length of the rotor is shown in Fig. 8. The efficiency stands for the volume of water delivered one rotation by a unit cross-section area and unit length of the rotor; therefore the efficiency  $\eta$  was defined by the following equation,

$$\eta = \frac{V}{SL_g} = \frac{4V}{\pi d_g^2 L_g} \quad (2)$$

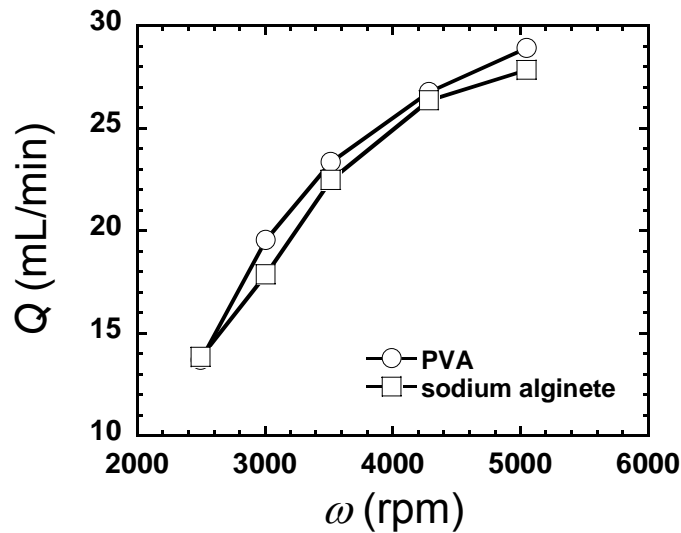
Here,  $S$  and  $L_g$  represent the area of cross-section and the length of the rotor, respectively.  $d_g$  is the diameter of the rotor. The efficiency increased and was saturated to a maximum value of ~0.008 on increasing the diameter or the length of the rotor. The efficiency was strongly affected by the diameter rather than the length of the rotor. The maximum value, which was seen at the diameter of  $d_g > 4.7$  mm or the length of  $L_g > 35$  mm, would be dominated by the shape of the rotor. The maximum efficiency is considered to be a good parameter to evaluate the water delivery in the present system. The pitch of the ditch printed on the rotor was constant as 0.08 turns/mm in the present study. The flow rate should be dependent on the pitch. The data is not shown in this paper because the pitch of the ditch is not

variable easily. The mold for the drill used in the present study was imprinted by commercial drills for woodwork. This is the reason why the pitch is constant. The pitch of the ditch will be a key factor especially in a micro device. The experiment using custom-made drills is already being planned and the data will be presented in a subsequent paper.



**Fig. 8.** The efficiency of water delivery as a function of the diameter (a) and the length (b) of gel-rotor.

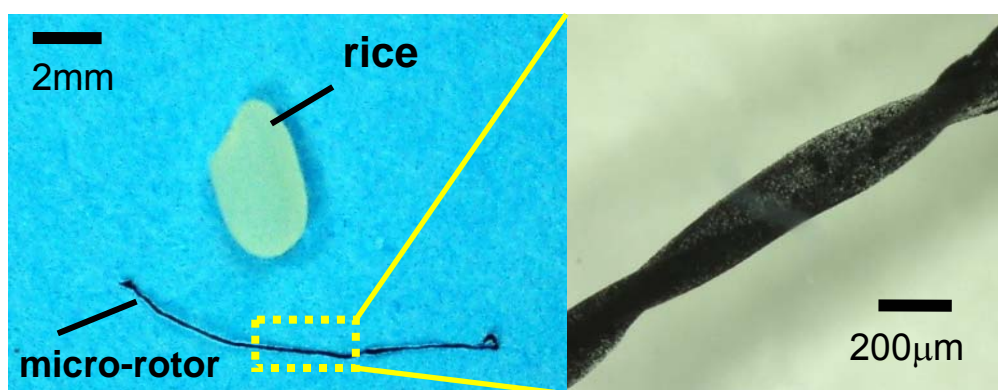
Fig. 9 shows the rotation speed  $\omega$  dependence of the flow rate  $Q$ . The data presents the effect of the material of the rotor on the flow rate. The ratio of the cross-section area of the gel-rotor to the tube  $(d_g/d_t)^2$  was 0.75, which was far larger than the critical value of 0.34. In both rotors (PVA and sodium silicate) the flow rate increased roughly proportional to the rotation speed when it was below 3500 rpm. Above the rotation speed of 3500 rpm, the flow rate tends to saturate to a constant value of approximately 28 mL/min. No apparent difference on the flow rate between PVA and sodium alginate was observed. It was shown that the ability of water delivery was independent of the materials of the gel-rotor.



**Fig. 9.** Rotation speed dependence of the flow rate for magnetic gel-rotors made of sodium alginate and PVA ( $L_g=35$  mm,  $d_t=6.0$  mm,  $d_g=5.2$  mm).



Fig. 10 demonstrates the photograph of the gel-rotor with a diameter of approximately 300  $\mu\text{m}$ . The rotor was synthesized using the mold with a diameter of 1000  $\mu\text{m}$ . The large decrease in the diameter was caused by the shrinkage during post-crosslinking. On decreasing the diameter of rotor, the shrinkage was significantly observed. The micro-rotor did not deliver water even in a capillary tube with a diameter of 500  $\mu\text{m}$ . As described in Fig. 4, the flow rate falls down to zero when the cross-section area of the gel-rotor to the tube  $(d_g/d_t)^2$  is less than the critical value of 0.34. The value  $(d_g/d_t)^2$  for the micro-rotor in the capillary tube was 0.36, which was larger than the critical value; however, the micro-rotor did not deliver water. Although the reason of this phenomenon is unclear, it might be that the shear force by the rotor acting on water is weakened in a microtube where the flow is described by the micro-hydrodynamics.



**Fig. 10.** Photograph of a micro-rotor made of sodium alginate gel containing barium ferrite with a diameter of  $\phi \sim 300 \mu\text{m}$ .

## Conclusions

A fluid pump using magnetic composite gels has been constructed. The fluid pump mainly consists of a gel-rotor and a driving magnet. The rotor was made from sodium alginate or PVA hydrogel containing the particles of barium ferrite. It was shown that the rotor underwent rotational motion using rotational magnetic fields and delivered water in straight and spiral tubes. Flow rates pumped by the rotor increased with increasing the diameter and the length of the rotor, and the rotation speed. The maximum efficiency of water delivery was estimated to be 0.008 for the rotor obtained in this study. The maximum flow rates of 34 mL/min were achieved when the rotation speed was 5000 rpm. The rotor with a diameter of 300  $\mu\text{m}$  was also successfully synthesized; however the rotor did not deliver water in a microtube with a diameter of 500  $\mu\text{m}$ . Since the driving force is a magnetic field, no electrical leads to the pump are needed. Additionally, the gel rotor is very flexible and the system of the pumping is very simple. This water-delivering system presented here would be useful to a pump working at a winding watercourse, such as  $\mu$ -TAS or a blood vessel in a human body.



## Experimental part

### *Synthesis of Magnetic Gel Rotor*

#### *-Sodium alginate gel*

A Pre-gel solution of the magnetic gel rotor was prepared by mixing the 1 wt.% sodium alginate (I-5, Kimitsu Chemical Industries) aqueous solution and the barium ferrite  $\text{BaFe}_{12}\text{O}_{19}$  (Sigma-Aldrich Co.) in a water bath. The concentration of ferrite was 30 wt%. The mean diameter of the magnetic particle was determined to be 15  $\mu\text{m}$  by using a particle size analyzer (Mastersizer2000, Malvern Instruments). The pre-gel solution was poured into a mold made of carrageenan gel with 3 wt.%  $\text{CaCl}_2$ . The shape of the mold was like a screw. After gelling the surface of the rotor, the mold was melted in hot water at 80 °C for several minutes. Post-crosslinking was carried out by sintering the rotor in a 3 wt.%  $\text{CaCl}_2$  aqueous solution for few days. The diameter and length of the rotor were  $\phi 3.8\text{-}5.5$  mm and 20-45 mm, respectively. The obtained rotor after the post-crosslinking was kept for a day in a large amount of purified water to remove excess ions and to give an equilibrium swelling. The gel rotor was put under a uniform magnetic field of 1 T for 1 min in order to give the rotor a remanent magnetization. The diameter of barium ferrite is much larger than that of the magnetic domain as a result the gel rotor has a remanent magnetization under no magnetic field.

#### *-Poly(vinyl alcohol) gel*

A Pre-gel solution of the magnetic gel rotor was prepared by mixing the 4 wt.% of PVA (Wako Pure Chemical Industries) aqueous solution and the barium ferrite  $\text{BaFe}_{12}\text{O}_{19}$  (Sigma-Aldrich) at room temperature. The concentration of ferrite was 30 wt%. Glutaraldehyde of a crosslinking agent and 1 N hydrochloric aqueous solution were added to the PVA aqueous solution. Crosslinking density was 1 mol%. The PVA solution containing ferrite and crosslinking agent was poured in a mold made of carrageenan gel of screw shape. Gelation took place at room temperature for 1 h. The rotor obtained was immersed in a large amount of purified water for 1 day to remove unreacted agents. Magnetization of the rotor made of PVA gel was also carried out in a similar way to the sodium alginate gel.

### *Magnetic Measurements*

Magnetization measurements up to 1T were carried out by a vibrating sample magnetometer (VSM-P7, Toei Industrial). The sample was a disk of 2 mm in diameter and 1mm in thickness. The magnetic gel was wrapped in a thin film of poly(vinyl chloride) to avoid evaporation of water from the gel.

### *Flow Measurements*

The flow rate was calculated by the weight of water that was delivered during 10 sec. The weight of water was measured by an electronic balance. We assume the rotation speed of the gel rotor equals to that of the driving magnet placed below the rotor. The rotation speed of the magnet  $\omega$  was calculated from the relationship between driving currents and rotation speeds of the magnet.

### Microscope Observations

Microscope observations for gel rotors were carried out using a digital microscope (VH-Z00R, VHX-900, Keyence Co.).

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