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ABSTRACT

Artificial cilia are promising as a next generation microfluidic device. In this study, we experimentally report that fibrous carbon wires produced by the self-organization process show cilium motions due to induced charge electro-osmosis (ICEO) under AC electric fields in water. In particular, we experimentally demonstrate that the carbon ICEO cilium with a branching wire structure shows asymmetric beating motion. We believe that our findings will contribute to the research of artificial cilia having asymmetrical motions, which would provide net propulsion in a low Reynolds number regime.

I. INTRODUCTION

The use of fibers and high-aspect-ratio structures is essential for functional structures in biological creatures.[1] In particular, artificial cilia using a hair-like structure are promising as a next generation microfluidic device because of their multi-functionality, such as pumping, mixing, valving, sensing, etc.[2,3] For example, Hanasoge *et al.*[4] reported that a net flow can be generated in a circular channel by magnetic cilia. Chen *et al.*[5] reported mixed performance due to magnetic cilia. Alfadhel and Kosel[6] reported a tactile sensor using magnetic nanocomposite cilia with a giant magneto-impedance sensor. Here, one main strategy for cilium fabrication is a top-down approach using photolithography technology, e.g., Pokroy *et al.*[1] reported the soft-lithography replication method to allow the fabrication of a biomimic array of stable high-aspect-ratio features. The other main strategy is a bottom-up approach using self-organization phenomena, e.g., Vilfan *et al.*[7] demonstrated self-assembled artificial cilia using magnetic beads and suggested a self-repairing function of their cilia.

Although Vilfan's approach is highly challenging, it requires large magnetic equipment for fabrication, driving, and repairment, and thus, it may not be suitable for total miniaturization of systems. To realize total miniaturization of systems, electrostatic cilia driven by an electric field have an advantage, e.g., Toonder *et al.*[8] reported that electrostatic cilia with a curled beam structure can generate fluid velocities not in water but in silicone oil. In this context, we have proposed artificial cilia using induced charge electro-osmosis (ICEO) so far; a valve,[9] a pump,[10] and a catcher[11] were examined theoretically, and the basic beating motions were examined experimentally.[12,13] Note that the ICEO cilia work in water can be miniaturized in the future. Thus, ICEO cilia are promising for biomedical applications. However, ICEO cilia that use the self-organization process have not been explored yet.

Therefore, in this study, we experimentally report that fibrous carbon wires produced by the self-organization process[14] show cilium motions due to ICEO under AC electric fields in water. In particular, we experimentally demonstrate that the carbon ICEO cilium with a branching wire structure shows asymmetric beating motion. Furthermore, through the discussion of carbon ICEO cilia, we show the possibility of the metachronal motion due to the difference in the wire length for the carbon ICEO cilium with a simple wire structure.

II. METHOD

Figure 1(a) shows an experimental setup of our artificial carbon cilium using ICEO. First, to fabricate a carbon wire by the self-organization process,[14] we initially turned off SW_1 and put a graphite rod (Pilot Co., neox graphite 0.3 2B) of diameter ϕ

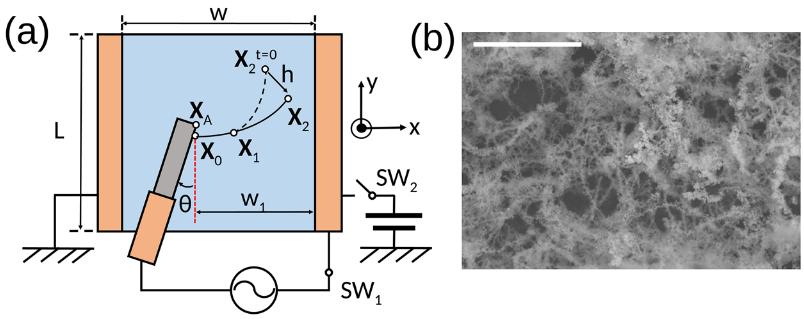

FIG. 1. Experimental setup of the artificial carbon cilium using ICEO: (a) a schematic of our experiment and (b) the SEM photograph of the carbon cilia with a fibrous network structure. Here, w = 5 mm and L = 15 mm. In (b), the scale bar shows 10 μm.

(=0.3 mm) between parallel Cu electrodes in deionized water (Milli-Q, 18.2 MΩ cm). Here, the gap distance w and width L of the electrodes are 5 mm and 15 mm, respectively. Furthermore, $\boldsymbol{X}_A$ (X_A, Y_A) denotes the edge position, w_1 denotes the gap distance between $\boldsymbol{X}_A$ and the right electrode, and θ denotes the tilt angle of the graphite rod, where $l_0 \equiv |\boldsymbol{X}_A - \boldsymbol{X}_0|$. Then, by applying a DC electric voltage V_0 between the electrodes during a time t_{DC} (typically, 30–120 s; SW_2 on), we obtained a carbon wire through a self-wiring phenomenon at the position $\boldsymbol{X}_0$ [$=(X_0, Y_0)$] on the right-side surface of the graphite rod. Here, we need not prepare any carbon suspension, and the carbon wire has a fibrous network structure microscopically, as shown in Fig. 1(b). In detail, the wire is considered to be an intercalational compound from the composition analysis by energy dispersive x-ray spectroscopy (EDS).[14] Second, to observe beating motions of the carbon wire, we turned off SW_2. Then, by applying a rectangular AC voltage of peak value V_0 and frequency f between the right electrode and the graphite rod during a time t_{AC} (SW_1 on), we observed that the tip position $\boldsymbol{X}_2$ [$=(X_2, Y_2)$] of the carbon wire moved toward the right electrode. Here, we define the displacement h at t as $h(t) \equiv |\boldsymbol{X}_2(t) - \boldsymbol{X}_2(0)|$. Furthermore, we define the provisional wire lengths l_1 and l_2 as $l_1 \equiv |\boldsymbol{X}_1 - \boldsymbol{X}_0|$ and $l_2 \equiv |\boldsymbol{X}_2(t_{AC}) - \boldsymbol{X}_1|$, where $\boldsymbol{X}_1$ is a joint position that is considered not to move, and $l_t(=l_1 + l_2)$ denotes a total provisional wire length. Furthermore, we determined $\boldsymbol{X}_2(t)$ and $h(t)$ by using the recorded video data of size 1280 × 720 with a frame rate of 25–27 fps.

III. RESULTS

A. Beating motions of the carbon ICEO cilium with a simple structure

Figures 2(a)–2(c) (Multimedia view) show the motions for the carbon ICEO cilium (Sample S_1) at t = 0, 0.33, and 1 s under an AC electric field, whereas Figs. 2(d)–2(f) (Multimedia view) show the

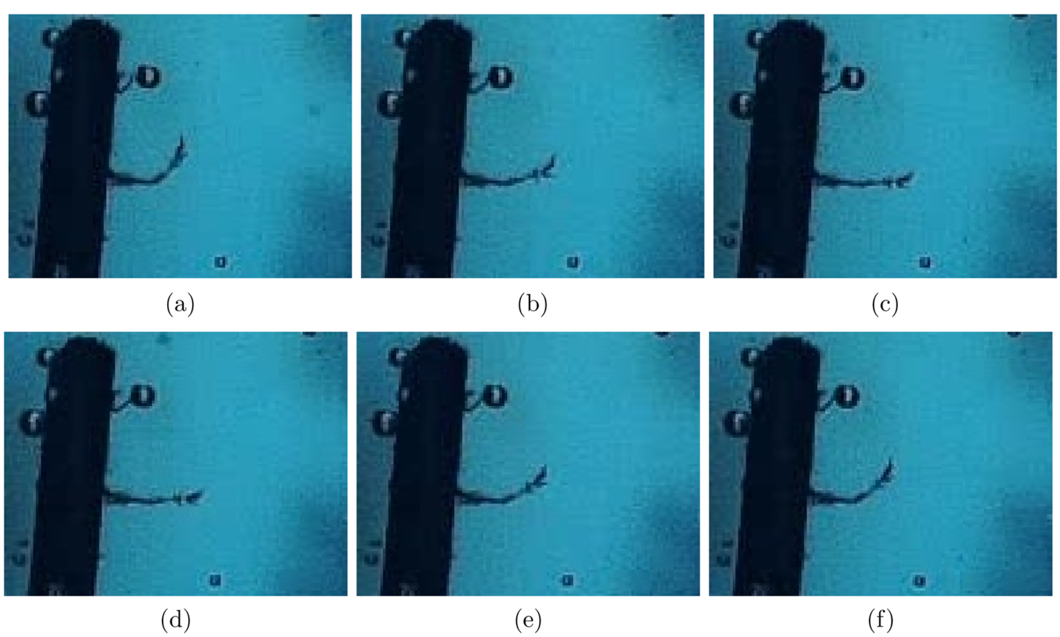

FIG. 2. Beating motions of the carbon ICEO cilium with a simple structure (V_0 = 30 V, f = 100 Hz): (a) t = 0 s (initial state), (b) t = 0.33 s (on-state), (c) t = 1 s (on-state), (d) t = 2 s (switch off), (e) t = 3 s (off-state), and (f) t = 4 s (off-state). Here, θ = 5°, w_1 = 2.41 mm, l_0 = 0.72 mm, l_1 = 0.2 mm, l_2 = 0.3 mm, and l_t = 0.5 mm. Multimedia view: https://doi.org/10.1063/1.5143700.1

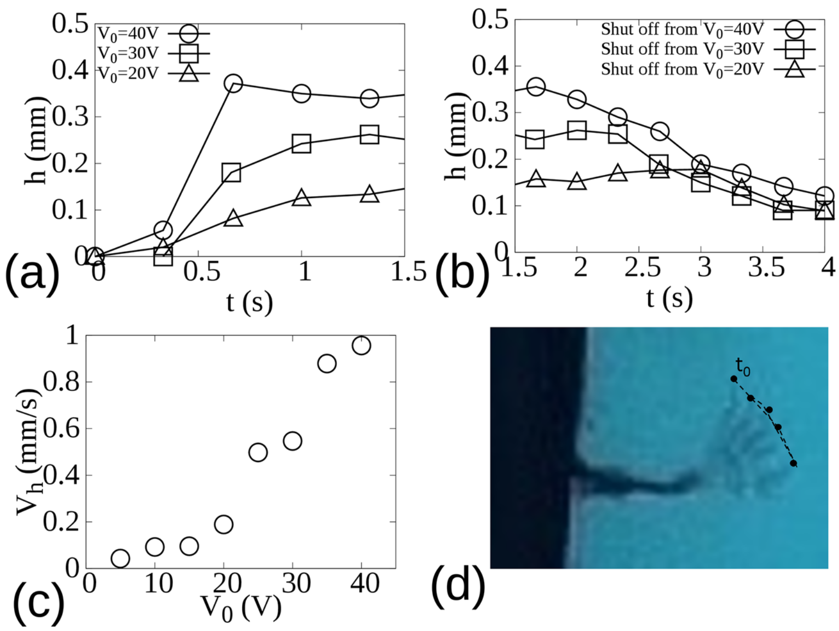

FIG. 3. Characteristics of the carbon ICEO cilium with a simple structure: (a) dependence of h on t when we applied V_0, (b) dependence of h on t when we shut off V_0, (c) dependence of V_h on V_0 (f = 100 Hz), and (d) the trajectory of the cilium (V_0 = 30 V, f = 100 Hz). In (a) and (b), V_0 = 30 V and f = 100 Hz. Here, $\theta = 5°$, w_1 = 2.41 mm, l_0 = 0.72 mm, l_1 = 0.2 mm, l_2 = 0.3 mm, and l_t = 0.5 mm.

returning motions at t = 2, 3, and 4 s without the AC electric field. As shown in Fig. 2 (Multimedia view), we observed a large deformation of the beam for the carbon cilia with a simple structure.

Figure 3(a) shows the dependence of h on t under the AC electric field at V_0 = 20–40 V and at f = 100 Hz, whereas Fig. 3(b) shows the dependence without the AC electric field. As shown in these figures, the forward and recovery response times were $\tau_f \simeq 1$ s and $\tau_r \simeq 1.5$ s, respectively, although there was an overshoot in a forward stroke at V_0 = 40 V. Figure 3(c) shows the dependence of the maximum velocity $V_h \equiv \frac{dh}{dt}$ on V_0. From Fig. 3(c), we found that there is a nonlinearity in V_h corresponding to the characteristic of ICEO. Note that the carbon fiber has a conductivity,[14] and thus, the tip of the surface is polarized under the electric field E_0. Then, an electric double layer (consisting of polarized surface charge and

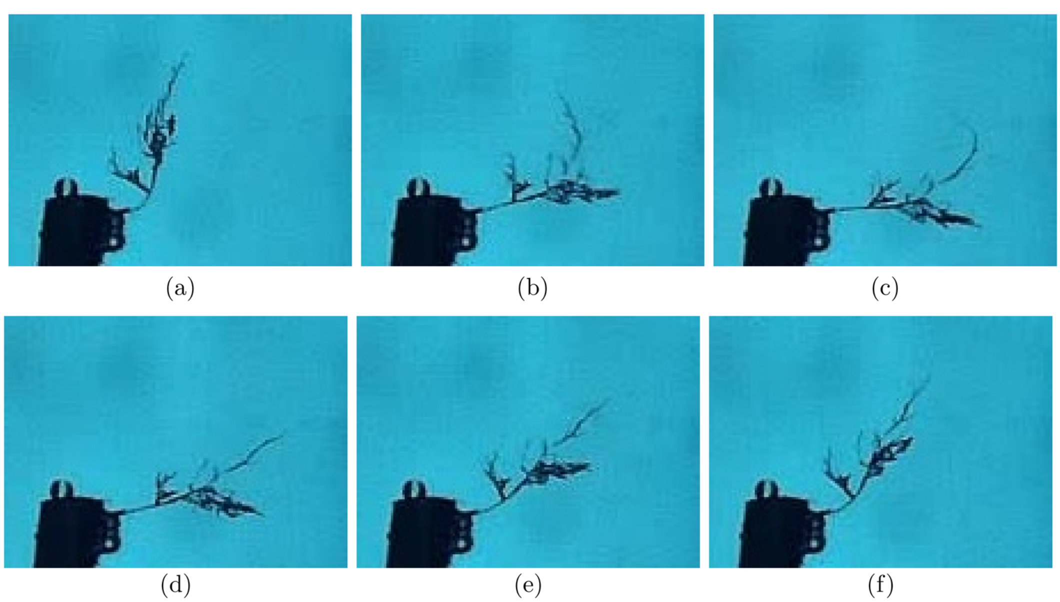

FIG. 4. Beating motions of the carbon ICEO cilium with a branching structure (V_0 = 30 V, f = 100 Hz): (a) t = 0 s (initial state), (b) t = 0.33 s (on-state), (c) t = 0.67 s (on-state), (d) t = 3 s (switch-off), (e) t = 5 s (off-state), and (f) t = 7 s (off-state). Here, $\theta = 7°$, l_0 = 0.056 mm, and w_1 = 3.20 mm. Multimedia view: https://doi.org/10.1063/1.5143700.2

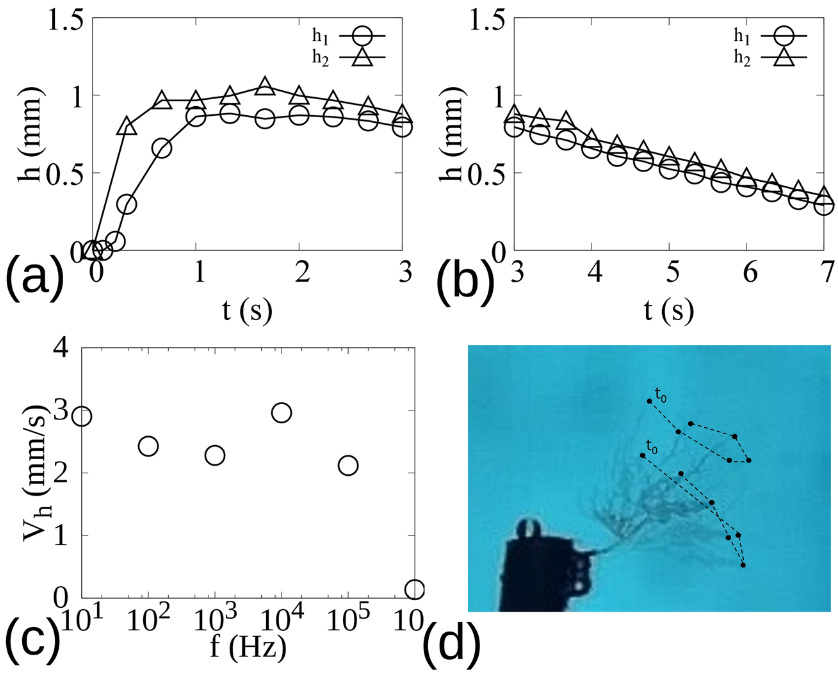

FIG. 5. Characteristics of the carbon ICEO cilium with a branching structure: (a) dependence of h on t when we applied V_0, (b) dependence of h on t when we shut off V_0, (c) dependence of V_h on f (V_0 = 30 V), and (d) t = 0–7 s (V_0 = 30 V, f = 100 Hz). In (a) and (b), V_0 = 30 V and f = 100 Hz. Here, $\theta = 7°$, l_0 = 0.056 mm, and w_1 = 3.20 mm.

counter ions) is formed at the tip, and the ions in the double layer moves along the tangential electric field E_t. As a result, the ICEO flow velocity[15] is proportional to E_0^2, and thus, V_h (due to the ICEO flow) is also proportional to E_0^2. Therefore, the behavior shown in Fig. 3(c) is expected under ICEO, and the underlying physics that drive this behavior are considered to be a hydrodynamic interaction due to ICEO. Figure 3(d) shows a picture composed of the six images in Fig. 2 translucently stacked. From Fig. 3(d), we understood that asymmetric motion cannot be observed for the carbon ICEO cilium with a simple structure.

B. Beating motions of the carbon ICEO cilium with a branching structure

Figure 4 (Multimedia view) shows the beating motions of the carbon ICEO cilium with a branching structure. As shown in Figs. 4(a)–4(c) (Multimedia view), the shorter branch responded faster than the longer branch to the electric field by the ICEO effect. This is because the bundle fiber structure of the shorter branch produces much ICEO flow than the thin fiber structure of the longer branch. Furthermore, as shown in Figs. 4(d)–4(f) (Multimedia view), the recovery of the deformation near the common root of the cilium was observed at t = 5 s, and then, the shorter and longer branches return while attracting each other until t = 7 s through the interaction due to the tangled thin wires. This is why the shorter branch seems to come back late at t = 7 s. Probably, the interaction between branches and the difference in the ICEO driving force are the origin of the asymmetric motion of the carbon ICEO cilium with a branching structure.

Figure 5 shows the characteristics of the ICEO cilium with a branching structure. Specifically, Fig. 5(a) shows the dependence of h on t under the AC electric field, whereas Fig. 5(b) shows the dependence without the AC electric field. In Figs. 5(a) and 5(b), $h_1(t) \equiv |\boldsymbol{X}_{2,1}(t) - \boldsymbol{X}_2(0)|$ and $h_2(t) \equiv |\boldsymbol{X}_{2,2}(t) - \boldsymbol{X}_2(0)|$, where $\boldsymbol{X}_{2,1}$ and $\boldsymbol{X}_{2,2}$ denote the tip positions of the longer and shorter branches, respectively. As shown in these figures, the forward and recovery response times of $\boldsymbol{X}_{2,1}$ were $\tau_f \simeq 1$ s and $\tau_r \simeq 4$ s, respectively, whereas the forward and recovery response times of $\boldsymbol{X}_{2,2}$ were $\tau_f \simeq 0.5$ s and $\tau_r \simeq 4$ s, respectively. Note that although the recovery speed of $\boldsymbol{X}_{2,2}$ was slower than that of $\boldsymbol{X}_{2,1}$ until t = 5 s, their recovery times were almost the same. Figure 5(c) shows the dependence of V_h on f. From Fig. 5(c), we found that V_h of the carbon ICEO cilium is approximately constant at $f \leq 10^5$ Hz although it becomes small at $f > 10^5$ Hz because of its insufficient charging time to the electric double layer. Figure 5(d) shows a picture composed of the six images in Fig. 5 translucently stacked. From Fig. 5(d), we found that asymmetric motion can be observed for the carbon ICEO cilium with a branching structure.

IV. DISCUSSION

A. Possibility of metachronal motion

Figure 6 (Multimedia view) shows the possibility of the metachronal motion. Specifically, Fig. 6(a) (Multimedia view) shows samples (S_1–S_4) having different beam lengths (l_0 = 0.72–4.10 mm) on the same graphite rod, whereas Fig. 6(b) (Multimedia view) shows the dependence of $h/h_{\max}$ on t. As shown in Fig. 6(b) (Multimedia view), there was a tendency that the response time becomes long as l_2 increases. Thus, there is a possibility to realize a metachronal motion of the carbon ICEO cilia by arranging carbon wires of different lengths in the order of length. Furthermore,

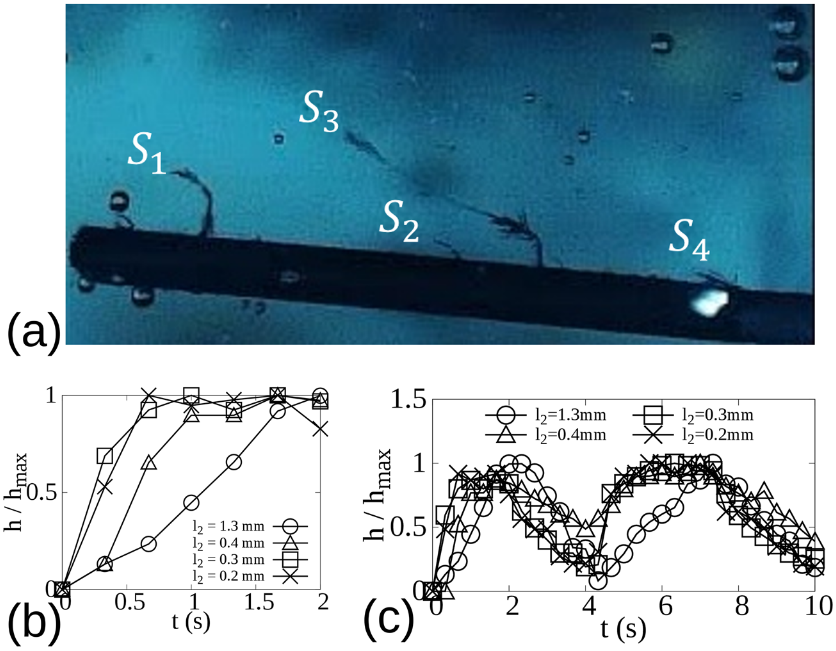

FIG. 6. Possibility of a metachronal motion due to the difference in wire length for the carbon ICEO cilium with a simple wire structure: (a) aamples S_1–S_4 (t = 0), (b) metachronal motions (V_0 = 30 V, f = 100 Hz), and (c) multiple cycles (V_0 = 30 V, f = 100 Hz). In (b), V_0 = 30 V and f = 100 Hz. Here, θ = 5° and w_1 = 2.41 mm and l_0 = 0.72, 2.34, 2.85, and 4.10 mm for S_1, S_2, S_3, and S_4, respectively. Multimedia view: https://doi.org/10.1063/1.5143700.3

Fig. 6(c) (Multimedia view) shows multiple cycles at V_0 = 30 V and f = 100 Hz. From Fig. 6(c) (Multimedia view), we can recognize that the cilium motion of the carbon wire can be repeated. Here, an elastic force of the carbon wire makes the cilium return to the initial state, and the balance between the elastic force and the flow resistance determines the returning velocity. Thus, we may control the returning time by controlling the synthesis parameters.

B. Meaning of our study

In this study, we have reported that fibrous carbon wires produced by the self-organization process show cilium motions due to ICEO under AC electric fields in water although self-assembled cilia using magnetic beads were already reported.[7] This carbon ICEO cilium intrinsically does not require large equipment for fabrication, driving, and repairment. Thus, it is suitable for total miniaturization of systems. In particular, since the carbon cilium can be produced by a simple self-organization process,[14] we may produce a channel covered with the fine cilium (or the cilia) at a low cost in the future.

C. Possibility of miniaturization

Although we have demonstrated the carbon ICEO cilium using a carbon rod,[14] the carbon rod is not always required. In other words, we may use various carbon self-wiring processes[16] for making a channel covered with our carbon ICEO cilium. In fact, a bundle of carbon wires can be produced between Cu electrodes without the carbon rod if graphite material exists in the solution [please see Fig. 5(a) of our previous paper[14]]. Thus, we may develop the path to miniaturization of the carbon cilium in the future. Here, we emphasize that the path to miniaturization offers interesting potential applications like low Re pumps although challenges remain for realization, as outlined in Sec. IV D.

D. Outstanding challenges

Many challenges remain for the self-organization process that produces carbon wires, e.g., the control of (1) thickness, (2) length, (3) direction, (4) position, and (5) pattern. In particular, it is a great challenge to make carbon wires grow in a regular pattern, ordered by length. Potential approaches to overcome these challenges are, e.g., (i) to control the carbon concentration, (ii) to control the growing time, (iii) to control the direction of the electric field, and (iv) to place seeds (e.g., graphite films) in the desired positions. Furthermore, by achieving a periodic pattern of electric fields, we may obtain a regular pattern of carbon wires. For example, by applying a voltage between a rectangular electrode and a wire placed above the edge of the rectangular electrode, we obtain asymmetrical electric fields. Thus, we may obtain a regular pattern by arranging the units since the carbon wire grows in the direction of the electric fields. By these approaches, we will obtain the desired applications, that is, in the future, we will demonstrate actual metachronal waves of a bundle of carbon wires growing in a regular pattern and will observe the net flow due the metachronal motion. Note that a net flow with a hair-like structure such as the structure of carbon wires has not been demonstrated yet although Hanasoge *et al.*[4] showed a net flow with a plate-like cilium structure.

V. CONCLUSION

In conclusion, we have demonstrated that fibrous carbon wires produced by the self-organization process show cilium motions due to ICEO under AC electric fields in water. In particular, we have demonstrated that the carbon ICEO cilium with a branching wire structure shows asymmetric beating motion. Furthermore, through the discussion of carbon ICEO cilia, we have shown the possibility that the carbon ICEO cilium with a simple wire structure shows different response times due to the difference in the wire length, and thus, it can be applied to a metachronal motion.

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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