Three-Dimensional Morphological Chirality Induction Using High Magnetic Fields in Membrane Tubes Prepared by a Silicate Garden Reaction

Wenyong Duan, Shu Kitamura, Ichiro Uechi, Akio Katsuki,† and Yoshifumi Tanimoto*

Graduate School of Science, Hiroshima University, Higashi-Hiroshima 739-8526, Japan, and Faculty of Education, Shinshu University, Nagano 380-8544, Japan

Received: January 26, 2005; In Final Form: April 13, 2005

Using a vertical superconducting magnet (max. 15 T), we studied magnetic field effects on membrane tube morphology prepared by a silicate garden reaction. At zero field, semipermeable membrane tubes grew upward when metal salts were added to a sodium silicate aqueous solution. In the presence of a magnetic field (15 T, downward) *right-handed* helical membrane tubes grew along a glass vessel's inner surface when magnesium chloride and copper sulfate were added. Referring to membrane tubes by the names of metal cations used in their preparation, in the case of Mg(II) and Zn(II) membrane tubes, the *left-handed* helical tubes grew when the field direction was reversed upward. The *left-handed* helical Mg(II) membrane tubes grew in the magnetic field when a glass rod was placed in a vessel. Mg(II) and Zn(II) tubes, separate from a vessel wall, grew in a twisted shape in the magnetic field. In situ observation of the solution's motion during the reaction revealed that the Lorentz force on the outflow from the opened top of the hollow membrane tube induced convection of the solution near the tube exit, engendering chiral growth of the membrane tubes. Relative orientation of the outflow and a boundary (a vessel wall or glass rod surface) helped to determine the convection's direction.

1. Introduction

High magnetic fields (>1 T) are well-known as convenient tools to control various chemical and physical processes.[1] For example, in the photoinduced electron-transfer reaction of a phenothiazine-viologen chain-linked compound, the lifetime of a photogenerated triplet biradical changes from 0.14 μs to ca. 7 μs with an increased magnetic field from 0 to 1 T; it then decreases to 2 μs with a further increase in the field to 13 T.[2] Silver dendrite patterns and yields, as generated by the solid/liquid redox reaction, are influenced markedly by magnetic fields.[3,4] A high magnetic field of 15 T affects the chemical equilibrium of hydrogen-ferromagnetic materials.[5] In electrochemical reactions, corrosion on a gold anode surface was dissolved by application of a 1.7 T magnetic field.[6] In addition, magnetic orientation of crystals[7] and polymers[8] has been reported. Although numerous processes can be influenced by magnetic fields, one process that is interesting to chemists is chirality induction using a magnetic field. Nevertheless, it is extremely difficult to induce magnetically molecular chirality, as reported by Rikken and Raupach.[9] Another interesting chirality is morphological chirality because materials having morphological chirality could be useful in creating a chirality-inducing environment in a chemical reaction. Two-dimensional morphological chirality induction in a silver dendrite was reported by Mogi et al.[3] They showed that a two-dimensional pattern of the dendrite was controlled by application of a magnetic field of ca. 5 T. Recently we demonstrated, for the first time, that the three-dimensional morphological chirality of membrane tubes produced by the reaction of sodium silicate aqueous solution and zinc sulfate crystal is controllable simply through the use of a high magnetic field.[10] At zero field, the tube grew almost straight upward. In a magnetic field of 3−15 T, the tube near the vessel wall grew *helically*, with *right-handed chirality*, whereas it grew helically with *left-handed chirality* on the outer surface of a glass rod when the rod was placed vertically in the vessel. All results were interpreted in terms of a boundary-assisted magnetohydrodynamics (MHD) mechanism, where the Lorentz force on solute ions induces circular convection of the solution whose direction is determined by a boundary and the magnetic field direction. Therefore, it is urgent to elucidate (1) whether this phenomenon is general in silicate garden reactions, (2) how a magnetic field affects a tube grown apart from the vessel wall, and (3) details of the chiral growth mechanism of membrane tubes in a magnetic field.

This study examines magnetic field effects on the silicate garden reaction using several metal salts to answer those questions. Demonstrably, chirality induction using a magnetic field is commonly observed in silicate garden reactions. The tubes grown apart from the vessel wall are also affected by a magnetic field: they grow in a twisted shape whose chirality is opposite to that grown near the wall. In situ observation of the motion of the solution during the reaction in a magnetic field reveals clearly that the Lorentz force indeed induces circular convection of the solution. Chiral growth of tubes near and apart from a vessel wall is induced by the convection whose direction is determined by the boundary.

2. Experimental Section

Magnetic fields were applied using a superconducting magnet (JMTD-LH15T40; JASTEC). The inner diameter of the room-temperature bore tube was 40 mm. The maximum field was 15 T and the direction of the field was vertical and downward unless otherwise noted. The magnetic field in the bore tube was inhomogeneous as described elsewhere.[11]

* To whom correspondence should be addressed at Hiroshima University. E-mail: tanimoto@sci.hiroshima-u.ac.jp.
† Shinshu University.

10.1021/jp050469m CCC: $30.25
Published on Web 06/25/2005

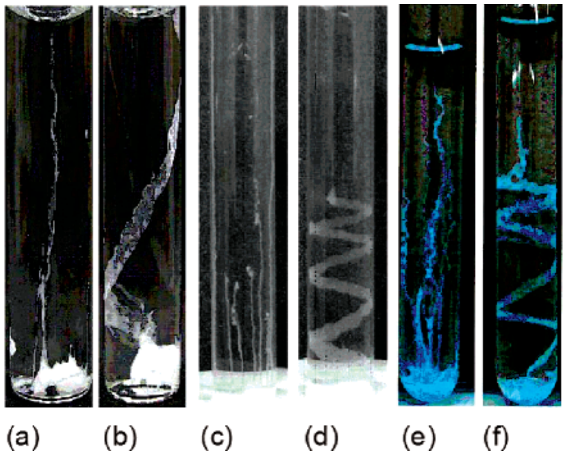

Figure 1. Magnetic field effects on the membrane tubes grown near the inner surface of the vessel wall. Mg(II) tubes at (a) 0 T and (b) 15 T, Zn(II) tubes at (c) 0 T and (d) 15 T, and Cu(II) tubes at (e) 0 T and (f) 15 T.

Sodium silicate aqueous solution (mole ratio, SiO_2/Na_2O = 2.06−2.31; 52−56 wt %) and magnesium chloride (<1 mm; amorphous), zinc sulfate (<1 mm granular), and copper sulfate (<1 mm; granular) were purchased from Wako Pure Chemical Industries, Ltd. and used as supplied. Distilled and deionized water was used. Typically, a sodium silicate aqueous solution (relative density, 1.06; pH ca. 12) was poured slowly in a cylindrical glass vessel (8 mm diameter × 30 mm length) containing a small amount of metal salt (ca. 0.1 g). The vessels were placed in the bore tube of the magnet and outside of the magnet (leak field, ca. 2 mT). For simplicity, the leak field of ca. 2 mT was considered as zero field. After the reaction proceeded for 30−60 min in the bore tube, the vessels were removed from the tube and the membrane tubes' respective morphologies were recorded using a conventional digital camera. All experiments were conducted at room temperature. Chemical composition of membrane tubes was estimated from X-ray fluorescence measurement using a scanning electron microscope (S-4000; Hitachi) equipped with an analyzer (EX-400; Horiba).

In situ observation of the motion of the solution during the reaction in a magnetic field was also carried out with a CCD camera and a digital video recorder. A sodium silicate aqueous solution (2 mL), to which plastic microparticles (polyethylene, ca. 180μm; Aldrich) were added as a tracer, was slowly poured into a cylindrical glass vessel (16 mm diameter × 25 mm length) containing magnesium chloride crystal, and then the motion of the solution (tracer) was observed by a CCD camera from the top.

3. Results

3.1. Membrane Tubes Grown Near the Inner Surface of a Vessel Wall. Figure 1 shows membrane tubes prepared by reactions of sodium silicate aqueous solution and magnesium chloride (diamagnetic), zinc sulfate (diamagnetic), and copper sulfate (paramagnetic).

Hereafter, membrane tubes are named respectively according to the metal cation used in their preparation, i.e., Mg(II) membrane tube, Zn(II) membrane tube, and Cu(II) membrane tube, because their compositions are unknown (see below). At zero field, all tubes grew upward with a slightly meandering shape. In the presence of a magnetic field of 15 T, the tubes (or a bundle of tubes) were imposed on the inner surface of the vessel wall, growing helically along the surface. The tubes initially prepared within a distance shorter than ca. 1 mm from the wall grew along the wall in magnetic fields. The pitch of Mg(II) helical tubes was larger than those of others, mainly because the Mg(II) tube's growth rate at zero field was slower than those of others. The pitch of each tube varied slightly from place to place, since magnetic field was inhomogeneous in vertical direction, as described elsewhere.[11] The helix direction was exclusively *right-handed*. Similar helical growth was also observed when nickel sulfate and cobalt sulfate were used as metal salts. Right-handed helical membrane tubes also grew when potassium silicate aqueous solution was used in place of a sodium silicate one. These facts indicate that helical growth of membrane tubes is common in a silicate garden reaction. Furthermore, in the case of the Cu(II) membrane tube prepared by adding *paramagnetic* copper sulfate, the tube did not grow when an average magnetic force of 900 $T^2\ m^{-1}$ was applied downward. The helical tube was too elongated when an average magnetic force of 600 $T^2\ m^{-1}$ was applied upward. These facts indicate that paramagnetism of the metal ion of metal salts does not contribute positively to the helical growth of membrane tubes, because the growth of diamagnetic Zn(II) membrane tubes is independent from the magnetic force.[10]

Figure 2. Mg(II) membrane tubes (a) and Zn(II) membrane tubes (b) grown under the reversed magnetic field (12 T) (see text).

The influence of the magnetic field direction was examined. When the field direction was reversed to the opposite direction, i.e., upward, *left-handed* helical tubes were obtained when magnesium chloride and zinc sulfate were used, as shown in Figure 2.

This fact implies that the chirality of tubes is also determined by the field direction.

3.2. Membrane Tubes Grown Near a Glass Rod in a Vessel. In a previous paper,[10] we reported that Zn(II) tubes grew outer surfaces of a ϕ 2 mm glass rod, a 2 mm × 2 mm square plastic rod, and a 2 mm × 2 mm × 2.8 mm triangle plastic rod in a magnetic field.[10] Therefore, whether a Mg(II) tube prepared using magnesium chloride crystals grew similar to a Zn(II) tube was examined. Figure 3a shows Mg(II) membrane tubes grown near an ordinary glass rod of ϕ 2 mm placed vertically in the vessel at 15 T.

A left-handed helical tube grew along the outer surface of the rod in a magnetic field, though no helical growth was observed at zero field. This fact indicates that left-handed helical tube growth at the outer surface of a rod is not special for the Zn(II) tube growth.

Furthermore, how small a helical tube was prepared using a magnetic field was examined for the Zn(II) tube. The result is shown in Figure 3b. Here, to match the helical growth rate of the Zn(II) tube to the lead diameter of the pencil, a magnetic

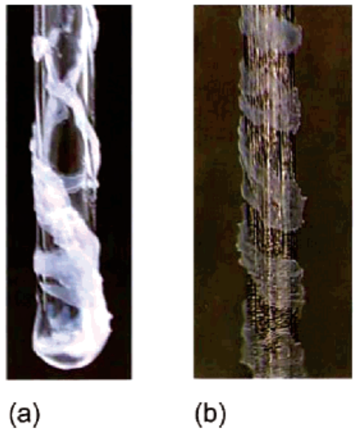

Figure 3. (a) Mg(II) membrane tubes grown along the outer surface of a ϕ 2 glass rod at 15 T. (b) Zn(II) membrane tubes grown along the outer surface of the 300 μm lead of a lead pencil at 5 T. The field was adjusted to 5 T so that the tubes wound helically on the outer surface of the extra-fine lead of a lead pencil.

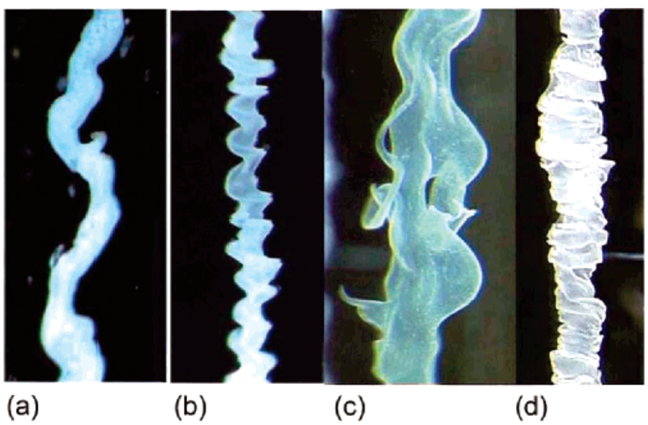

Figure 4. A Mg(II) membrane tube grown apart from the vessel wall at (a) 0 T and (b) 15 T. A Zn(II) membrane tube grown apart from the vessel wall at (c) 0 T and (d)15 T. The magnification factor is ×175.

field was adjusted to 5 T so that the tube wound on the lead. A left-handed Zn(II) helical tube grew on the outer surface of a ultrafine lead of a lead pencil (ϕ 300 μm), indicating that the tube grew very accurately along the surface.

The chirality of tubes grown on the outer surface was opposite that of the tubes grown along the inner surface of the vessel wall. The boundary caused by a vessel wall or a surface of a glass rod serves an important role in determining the helixes' chirality.

3.3. Membrane Tubes Grown Apart from a Vessel Wall. As explained in the above section, a magnetic field affects the tube shape near the vessel wall and a glass rod. It is therefore expected that the field affects tubes grown apart from a vessel wall, though the tubes, prepared at a distance longer than ca. 1 mm from the wall, grow almost upward in a magnetic field. We examined the morphology of tubes grown apart from a vessel wall in magnetic fields to examine this possibility. Figure 4 shows Mg(II) and Zn(II) membrane tubes grown apart from the vessel wall.

In a magnetic field of 15 T, they grew in a twisted form, while at zero field they meandered upward. The twist direction was exclusively *left-handed*, which was opposite that of the tubes grown near the vessel inner surface.

3.4. In Situ Observation of the Motion of the Solution in a Magnetic Field. A previous study[10] suggested that a magnetic field might induce circular convection of a solution whose

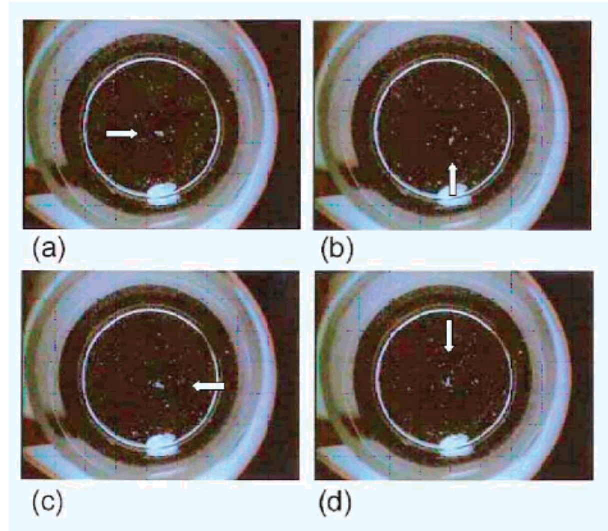

Figure 5. Sequential photos of the magnetic field-induced convection during Mg(II) tube formation reaction in a cylindrical glass vessel at 15 T (top view). They are taken at (a) 35, (b) 40, (c) 46, and (d) 52 s delays after the reaction start. The diameter of the glass vessel was 16 mm. The white lump at the lower side of the photos is a magnesium chloride crystal. The motion of a small aggregate of tracers at the center of the vessel is indicated by an arrow.

direction is determined by a boundary: In the case of Zn(II) helical membrane tubes, the direction is right-handed at the inner surface of the vessel wall, whereas it is left-handed at the outer surface of a glass rod placed in the vessel. Therefore we observed motion of the sodium silicate aqueous solution during the reaction in a magnetic field by adding polyethylene microparticles as a tracer to the solution to obtain experimental evidence. No convection of the solution was observed when a sodium silicate aqueous solution was placed by itself in a magnetic field of 15 T. At zero field, no detectable convection was observed when a small amount of magnesium chloride crystal was added to the solution. Figure 5 shows that convection of the solution was observed when a magnetic field of 15 T was applied to the solution during the reaction.

Rigorous convection of the solution occurred immediately after adding the salt into the solution in a magnetic field. Its direction was considerably irregular, as it was dependent on the amount and location of the added salt. Figure 5 shows that the solution near the wall underwent a right-handed steady convection in a short time when a small amount of the salt was placed on the bottom of the vessel near the vessel wall. On the other hand, the solution near the salt underwent left-handed steady convection in a short time when a small amount of the salt was placed on the bottom of the vessel near the center of the vessel. Typically, the rate of convection was 0.3−2 mm s^{-1} in the present experimental condition. These in situ observations implied that a magnetic field induces convection of the solution. Its direction is right-handed when the salt is placed near the vessel wall, whereas it is left-handed when it is placed apart from the wall. The direction of helical tubes and twisted tubes is parallel to that of the convection of the solution. The temperatures of the solution before and after the reaction are almost the same (<0.1 °C) regardless of the magnetic field.

4. Discussion

4.1. Formation of Membrane Tubes. The silicate garden reaction is a well-known reaction that is often shown to students

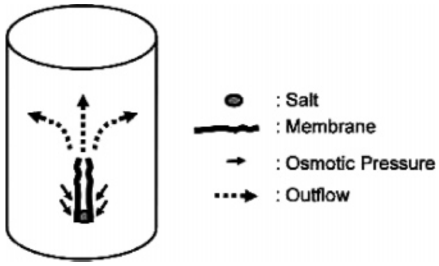

Figure 6. Mechanism of membrane tube formation (side view).

as a chemical demonstration.[12] It is a reaction of sodium silicate aqueous solution and metal salts that forms insoluble membrane tubes. However, the reaction is highly complex, as reported elsewhere,[13−18] because the composition of water-soluble silica is not well defined. At pH 11.5, the main water-soluble silica is $SiO(OH)_3^-$ when the molar concentration of Si is within 0.005−0.02 mol dm^{-3} and in concentrated basic solutions at pH 11.5 and Si concentration of $>$0.02 mol dm^{-3}; it is suggested that Si aqueous species polymerize.[16] The Si concentration and pH of the present solution are roughly estimated respectively as 0.8 mol dm^{-3} and 12, indicating that the silica is dissolved in several different forms. Therefore the silicate garden reaction would be given, *symbolically*, by two parallel reactions, i.e., the reaction of water-soluble silica ions and H^+, precipitating water-insoluble silica, and that of metal ion and hydroxyl ion, precipitating metal hydroxide. The products' compositions are dependent on experimental conditions: concentration, reaction time, stirring condition, and so on. Therefore, the product is a mixture of metal hydroxide and silica with no well-defined composition.[13,15] Previous speculation—that metal silicate membrane tubes would be formed from the reaction of sodium silicate solution and zinc sulfate crystal—was incorrect. In the case of Mg(II) and Zn(II) membrane tubes, the atomic ratios of Si/Mg and Si/Zn were estimated as ca. 2.5 and 2−2.4, respectively, from a preliminary X-ray fluorescence analysis.

The mechanism of membrane tube growth is also complicated.[14−18] Figure 6 shows growth of a membrane tube schematically.

A colloidal semipermeable membrane composed of water-insoluble silica and metal hydroxide is formed on the crystal surface when metal salt crystals are added into a sodium silicate aqueous solution. Water and OH^- ion diffuse by osmotic pressure into the space between the membrane and the crystal surface, dissolving the crystal. Their osmotic inflow ruptures the membrane and the solution, rich with metal salt ions, flows out. Subsequently, the metal ions in the outflow react with water-soluble silicate ions at the opened-exit of the membrane tube, forming hollow tubes. Because of the difference in the aqueous solutions outside and inside the tube, hollow tubes grow upward. In situ observation of the tube growth shows that a hollow membrane tube has an opened exit at its top.

4.2. Mechanism of Three-Dimensional Morphological Chirality Induction. Experimental results described in the previous sections can be summarized as follows: (1) A magnetic field produces *right-handed* helical tubes grown along the inner surface of a vessel wall regardless of the magnetism of metal salts, whereas *left-handed* ones grow on the outer surface of a glass rod placed vertically in the vessel. (2) The helix chirality is reversed when the magnetic field direction is reversed. (3) *Left-handed* twisted tubes grow in a magnetic field when salts are placed apart from the vessel wall. (4) The aqueous solution near the salt undergoes convection during the reaction in a magnetic field, whose direction is *right-handed* when salt is placed near the vessel wall, and *left-handed* when it is placed apart from the wall. (5) The handedness of helixes is the same as the direction of convection. Further, a previous study showed that (6) pitches of the Zn(II) helical tubes grown on the inner surface of a glass vessel are proportional to the inverse of the magnetic field.[10] These observations suggest that a magnetic field affects mass transportation from bulk solution to the reaction zone (exit of the hollow membrane tube). Although two possible mechanisms of magnetic field effects on mass transportation in solution exist, i.e., MHD and magnetic force, the latter cannot explain the above observations. This is because magnetic force, which is parallel to the magnetic field, cannot induce circular motion of the solution in the plane perpendicular to the field. All the results are explainable only by the boundary-assisted MHD mechanism. In the MHD mechanism, the Lorentz force on moving ions in a solution in a magnetic field results in the convection of the solution (MHD-induced convection) because of collision among solute ions and solution. This mechanism is well-known in electrochemical reactions.[19] The boundary-assisted MHD mechanism is the mechanism in which a boundary posed by a wall or a surface serves an important role in determining the direction of MHD-induced convection.

In the reaction of sodium silicate and magnesium chloride, for example, a possibility exists that the motions of more than six ionic species, i.e., sodium ion, water-soluble silica ions, proton and hydroxyl ion in a bulk solution, and magnesium and chloride ions in the hollow tube, are affected by the Lorentz force. The in situ observation of the solution's motion in a magnetic field indicates that thermal motion of the ions in the bulk solution at room temperature was unaffected by a 15 T magnetic field, though a previous study advanced this possibility.[10] No Lorentz force-induced convection resulting from the thermal motion of ions at room temperature was confirmed through in situ observation of the motion of sodium silicate solution without magnesium chloride salt at 15 T. The thermal motion of the solution induced by the reaction heat is seemingly negligible, since the temperature of the solution before and after the reaction is almost the same regardless of the magnetic field. The most important feature of the silicate garden reaction is that a metal salt solution in a hollow tube flows out from the opened top of the tube and precipitates as membrane tubes at the tube exit. Consequently, a stream of ionic solution is induced by the reaction, as shown schematically in Figure 6.

The stream was unobservable in the tracer experiment at zero field, but it was reported to be observable using a Mach−Zender interferometer.[18] The stream flows in all directions. Therefore, the average speed of the stream might be too moderate for detection using the tracer. The solution in the tube contains both magnesium positive and chloride negative ions. However, the solution flowing out of the tube may be transiently and locally rich with the chloride negative ion because magnesium ion reacts immediately with hydroxyl ion at the hollow tube exit. This electrically nonequilibrium state may be dissolved in a short time by diffusion of sodium positive ion to the outflow and neutralization of electric charge of the solution. Therefore, the cause of the convection may be attributable primarily to the chloride negative ion in the outflow. Figure 6 shows that this outflow spreads outward horizontally and vertically. The horizontal motion of the solution rich with chloride ion, which is perpendicular to the field, is affected by the Lorentz force $\boldsymbol{F}_L$, given by the following equation:

$$\boldsymbol{F}_L = q\boldsymbol{v} \times \boldsymbol{B} \tag{1}$$

where q is the charge of an ion, $\boldsymbol{v}$ is its velocity, and $\boldsymbol{B}$ is the

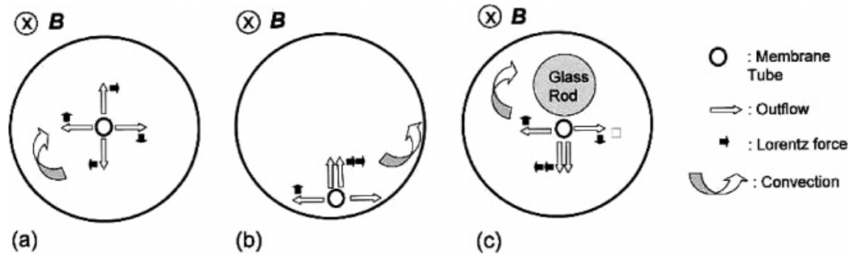

Figure 7. Mechanism of helical and twisted membrane tube formation in a magnetic field (top view): (a) A tube grown apart from the vessel wall; (b) a tube grown near a vessel wall; and (c) a tube grown near a glass rod's outer surface.

magnetic field flux density. A magnetic field does not affect the motion of the outflow parallel to the field. Because the motion of negative ion-rich outflow is the radial direction and the magnetic field is downward, the direction of the Lorentz force is left-handed, as shown in Figure 7a. As the tube apart from the vessel wall grows under the steady influence of the left-handed convection, the tube is twisted to the left-handed direction. Consequently, the tube grown apart from the wall is twisted in the left-handed direction. The motion of the outflow near a wall is influenced by its boundary. Figure 7b shows that the direction of the outflow is limited when a tube grows near the inner surface of a vessel wall. The outflow moving to the outer radial direction is bound on the wall to the inner radial direction. Figure 7b shows that the outflow is thereby imposed on the wall and moves along the wall. Consequently, the outflow moves in a right-handed direction along the wall, thereby inducing right-handed convection. The convection speed is faster than the growth speed of the tube. Therefore, the hollow tube grows to the right-handed direction along the wall. Figure 7c shows that the restriction posed by a wall becomes reversed when a hollow tube grows near the outer surface of a glass rod. The outflow to the outer surface of a rod is bound on the surface in the opposite direction. Because of these motions, the outflow is imposed on the wall and moves in the left-handed direction along the outer surface of the rod, thereby inducing left-handed convection. The Lorentz force initially influences the direction of the ionic aqueous solution in a magnetic field when the solution moves freely. In the presence of a boundary such as a vessel wall or a glass rod, the solution moves along the surface of the boundary with the direction determined by the relative orientation of the tube and the wall (boundary-assisted MHD mechanism). Therefore, the boundary plays a very important role in determining the direction of convection.

During the reaction, motion of water-soluble silica negative ions in the bulk solution might also be affected by the magnetic field because it must diffuse near the exit of the hollow tube to react with magnesium ion. Similarly, the motion of sodium positive ion might be affected by the magnetic field because the ion must diffuse to the outflow to neutralize the solution's electric charge. However, the speed of the diffusive motion could be slower than that of convection. Therefore, the Lorentz force on the motion of these ions may not induce the solution's convection to a measurable degree.

Convection of the solution occurs by the balance of the Lorentz force on the solution and a frictional retarding force from its surroundings. Estimation of the Lorentz force to the outflow of ionic solution is impossible, as the concentration of ions in the solution and the retarding force are unknown. Here, for the purpose of discussion, we will estimate *tentatively* the intensity of the Lorentz force on the outflow transiently containing 10^{-3} mol dm^{-3} of chloride ion in the case of the Mg(II) tube formation reaction. If we further assume that the speed of the outflow is the same as the convection speed of the solution (0.3−2 mm s^{-1} at 15 T), the Lorentz force on the outflow is estimated to be 4.3×10^{-3} to 2.9×10^{-2} N cm^{-3}. These values are more or less comparable to gravity on water (9.8×10^{-3} N cm^{-3}). Since the outflow speed at the exit of the hollow tube could be much faster than the convection speed, the Lorentz force on the outflow could be strong enough to induce convection, even though the concentration of chloride ion in the outflow is lower than 10^{-3} mol dm^{-3}.

The effect of the gradient field on the growth of membrane tubes prepared by the silicate garden reaction was also reported.[20] Paramagnetic salts such as manganese sulfate were added to sodium silicate aqueous solution in a horizontal magnetic field (up to 9 T, 410 T^2 m^{-1}). In magnetic fields, twisted tubes grew in the high-field direction. These tendencies are concurrent with the present observation: the tubes grown separately from a vessel wall (Figure 4) and the results were explainable by the combination of magnetic force and Lorentz force.

The silicate garden reaction is special because the outflow from a hollow membrane tube induces intense convection of the solution in a magnetic field. Usually the rate of diffusion is not rapid, but the Lorentz force might also affect it even though it is very slow. To examine whether this mechanism is also applicable for a diffusion-limited reaction, we also observed the dendrite formation reaction of silver ion and zinc plate in aqueous solution.[21] Silver ion in bulk solution diffuses to the zinc metal surface and silver dendrite is deposited there. In situ observation revealed that many tops of flexible silver dendrites, composed of numerous microcrystals, undergo precession motion in a horizontal magnetic field, indicating that a magnetic field affects even a diffusion-limited reaction where the concentration gradient is rendered steep by the reaction. This finding is important because the diffusion-controlled process of ions between liquid and solid phases is a popular process in chemistry. These results present the possibility of preparing helical or twisted tubes and wires using magnetic fields, provided that the solid product of a reaction is flexible in morphology.

The concept of using three independent macroscopic factors affecting mutual orientation of the reactant molecules to accomplish absolute asymmetric synthesis was discussed by Bielski and Tencer.[22] In the present study we utilized simply Lorentz force to induce macroscale morphological chirality of membrane tubes. It seems that Lorentz force is a simple method for macro- and microscale chirality induction, though it is unclear whether it is applicable to molecular-scale chirality induction.

In conclusion, it is verified that three-dimensional morphological chirality of membrane tubes is commonly inducible in a silicate garden reaction using a magnetic field. Helical tubes and twisted tubes were prepared by applying a magnetic field. Their morphological chirality is controllable using both the magnetic field direction and the boundary condition. A magnetic field induces circular convection of the solution near the tube. All results are interpreted in terms of the boundary-assisted MHD mechanism.

Acknowledgment. We thank Mr. Shizuo Hanafusa of Hiroshima Prefecture Eastern Research and the Development Center for X-ray fluorescence measurement. The work was supported partly by the Grant-in-Aid for Scientific Research (No. 15350016) from JSPS and partly by the Grant-in-Aid for Scientific Research on Priority Area "Innovative utilization of strong magnetic fields" (Area 767, No. 15085208) from MEXT of Japan.

References and Notes

(1) Kitazawa, K. (supervised), Ozeki, S., Tanimoto, Y., Yamaguchi, M., Eds. **Jiki Kagaku (Magneto-Science)**; IPC: Tokyo, Japan, 2002.

(2) Fujiwara, Y.; Aoki, T.; Yoda, K.; Cao, H.; Mukai, M.; Haino, T.; Fukazawa, Y.; Tanimoto, Y.; Yonemura, H.; Matsuo, T.; Okazaki, M. *Chem. Phys. Lett*. **1996**, *259*, 361.

(3) Mogi, I.; Okubo, S.; Nakagawa, Y. *J. Phys. Soc. Jpn*. **1995**, *60*, 3200.

(4) Tanimoto, Y.; Katsuki, A.; Yano, H.; Watanabe, S. *J. Phys. Chem. A* **1997**, *101*, 7359.

(5) Yamaguchi, M.; Yamamoto, I.; Miura, S. *Phys. Lett. A* **1989**, *134*, 504.

(6) Watanabe, T.; Tanimoto, Y.; Nakagaki, R.; Hiramatsu, M.; Sakata, T.; Nagakura, S. *Bull. Chem. Soc. Jpn.* **1987**, *60*, 4163.

(7) Fujiwara, M.; Fukui, M.; Tanimoto, Y. *J. Phys. Chem. B* **1999**, *103*, 2627.

(8) Kimura, T.; Sata, H.; Ito, E. *Polym. J.* **1998**, *30*, 455.

(9) Rikken, G. L. J. A.; Raupach, E. *Nature* **2000**, *405*, 932.

(10) Uechi, I.; Katsuki, A.; Dunin-Barkovskiy, L.; Tanimoto, Y. *J. Phys. Chem. B* **2004**, *108*, 2527.

(11) Katsuki, A.; Uechi, I.; Tanimoto, Y. *Bull. Chem. Soc. Jpn*. **2004**, *77*, 275.

(12) Chakhashiri, B. Z. *Chemical Demonstrations*; University of Wisconsin Press: Madison, WI, 1983; Vol. 3.

(13) Iler, R. K. *The Chemistry of Silica*; John Wiley: New York, 1979.

(14) Jones, D. E. H.; Walter, U. *J. Colloid. Interface Sci.* **1998**, *203*, 286.

(15) Collins, C.; Zhou, W.; Mackay, A. L.; Klinowski, J. *Chem. Phys. Lett*. **1998**, *286*, 88.

(16) Gout, R.; Pokrovski, G. S.; Schott, J.; Zwick, A. *J. Solution Chem*. **2000**, *29*, 1173.

(17) Blkose, D.; Ozkan, F.; Kokturk, U.; Ulutan, S.; Ulku, S.; Nisli, G. *J. Sol-Gel Sci. Technol*. **2002**, *23*, 253.

(18) Cartwright, J. H. E.; Garcia-Ruiz, J. M.; Novella, M. L.; Otalora, F. *J. Colloid. Interface Sci*. **2002**, *256*, 351.

(19) Uechi, I.; Fujiwara, M.; Fujiwara, Y.; Yamamoto, Y.; Tanimoto, Y. *Bull. Chem. Soc. Jpn.* **2002**, *75*, 2379.

(20) Yokoi, H.; Usuba, S.; Kakudate, Y. Proceedings of the 7th Meeting, Symposium on New Magneto-Science 2003, Tsukuba, Nov. 2003, p 311.

(21) Katsuki, A.; Tanimoto, Y. *Chem. Lett*. **2005**, *34*, 726.

(22) Bielski, R.; Tencer, M. *Can. J. Chem*. **2003**, *81*, 1029.