Dissolution Process of Copper Sulfate into Water in a Heterogeneous Vertical Magnetic Field

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The dissolution process of copper sulfate into water in a heterogeneous magnetic field vertical to the interface was examined from the viewpoint of the self-organization process of the nonequilibrium fluctuations. In a heterogeneous magnetic field, a material receives a magnetic force proportional to the product of the magnetic susceptibility, the magnetic flux density B and its gradient (dB/dz). As the reaction proceeds, a diffusion layer develops in the area where changing susceptibility is formed at the interface because of the dependence of the magnetic susceptibility on the solute concentration. In the case of a vertical magnetic field, a magnetic force is applied to the diffusion layer to induce magnetic force fluctuation and the resultant concentration fluctuation of the solute, which self-organize numerous convection cell. The cell pattern was then observed from the morphology of the dissolved copper sulfate pentahydrate crystal. As a result, it was concluded that the pattern changes from a roll-type to a rectangular-type as the magnetic flux density increases.

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1. Introduction

In line with the recent development of the superconducting magnet, much attention has been paid to various phenomena in high magnetic fields. In the field of materials processing, magnetic fields have been used as an effective tool to suppress the thermal convection of a liquid material with high electric conductivity (e.g., for fused silicon) by Lorentz force resulting from the evolution of the eddy current.¹⁻³⁾ Braithwaite *et al.*⁴⁾ have reported another type of phenomenon; namely, since the magnetic susceptibility of gadolinium nitrate largely changes with the temperature, the gadolinium nitrate solution with the temperature difference induces a specific thermal convection in the magnetic field, which is sometimes accelerated and sometimes depressed, depending on the direction of the magnetic field. In this case, the magnetic force acting on a nonmagnetic material with a difference in magnetic susceptibility plays an important role. For example, in protein crystallization,⁵⁾ this force is utilized to cancel the gravitational force for blocking the sedimentation of minute crystals.

In the case of physico-chemical processes at a liquid-liquid, liquid-gas or liquid-solid interface, the mass transfer of the reactant or solute is an important process. Some interesting phenomena concerning the mass transfer in high magnetic fields have been noted by researchers,⁶⁻¹⁰⁾ i.e., i) the increase of dissolution rate of oxygen into deaerated water, ii) the increase of the mass flux through a membrane of a Millipore filter, and iii) the increase of the evaporation rate of water. All these phenomena have been elucidated by the magneto-convection arising from the difference of the magnetic potentials of the materials.^{6-8,11)} However, it is impossible to obtain the quantitative relationship between the mass flux and the imposed magnetic field with the thermodynamic equilibrium because

such convection flows are thought to be formed through a self-organization process characteristic of the nonequilibrium process.

The aim of this paper is thus to examine the self-organization process from the nonequilibrium fluctuations¹²⁻¹⁷⁾ by the morphological change of copper sulfate crystals when the copper sulfate dissolves into water in the magnetic field vertical to the interface.

2. Theoretical

2.1 Magnetic force fluctuation

When the magnetic field having a distribution in the vertical z -direction is, as shown in Fig. 1, applied to the material, the following magnetic force is induced at the surface of the material ($z = 0$),¹⁸⁾

$$F_z = \frac{\chi}{\mu_0} B \left(\frac{dB}{dz} \right)_{z=0} \quad (1)$$

The magnetic susceptibility χ is given by a function of the concentration of the solute C , i.e., within the first approximation,

$$\chi = \chi_s [1 + \alpha_0 (C - C_s)], \quad (2)$$

where χ_s is the magnetic susceptibility at the surface concentration C_s which is assumed to be saturated, and α_0 is the concentration coefficient of the magnetic susceptibility, i.e.,

$$\alpha_0 \equiv \frac{1}{\chi_s} \left(\frac{\partial \chi}{\partial C} \right)_\mu, \quad (3)$$

where μ implies that the composition of the solution but not the solute is kept constant.

Along with the occurrence of the reaction processes such as dissolution and deposition at the solid surface, a diffusion

Fig. 1 Distribution of the magnetic field along the vertical bore axis. a, coil of the superconducting magnet; b, sample; c, maximum point of magnetic flux density (defined as $z = 0$). Magnetic flux density B is applied vertically to the interface of the sample. Magnetic force F operates in opposite directions on both sides of the maximum point c.

layer is formed. The concentration distribution of the solute in this layer is expressed as

$$C = \langle C \rangle + c, \quad (4)$$

where $\langle C \rangle$ is the concentration averaged over the solid surface, and c is the fluctuation. $\langle C \rangle$ is assumed to have a linear distribution in the diffusion layer. Therefore, the average concentration gradient is, as shown in Fig. 2, defined by

$$L \equiv \pm \frac{|\Delta C_s|}{\delta}, \quad (5)$$

where δ is the thickness of the diffusion layer, and “ $\pm$ ” correspond to positive and negative concentration gradients, respectively. In contrary to the usual convection problem such as the Bénard cell convection,¹⁹⁾ in this case, δ is not given a priori, but given as an arbitrary constant. The value is, as discussed later, automatically determined from a self-organization process of the convection cells. ΔC_s is the difference between the bulk concentration C_0 and the average surface concentration $\langle C_s \rangle$, which is, as mentioned above, equal to the saturated concentration C_{sat} ,

$$\Delta C_s \equiv C_0 - \langle C_s \rangle. \quad (6)$$

According to eqs. (1) and (2), the magnetic force due to the magnetic field gradient in the z -direction is vertically imposed on this diffusion layer and its average value is written as

$$\langle F_z \rangle = \frac{\chi_s}{\mu_0} [1 + \alpha_0 (\langle C \rangle - C_s)] B \left(\frac{dB}{dz} \right)_{z=0}. \quad (7)$$

This average magnetic force is balanced with the static pressure $\langle P \rangle$.

However, the concentration fluctuation accompanied by the dissolution or deposition induces the fluctuation of the motion in the diffusion layer. In the hydrodynamically stable case, the fluctuation is suppressed by the energy dissipation through the fluid viscosity. On the contrary, in the unstable case, it develops into obvious convective motion owing to a kind of positive feedback cycle between the fluid motion and the mass transfer.

The magnetic force fluctuation generating the convection is

Fig. 2 Two kinds of average concentration distributions in different convective-diffusion layers. (a) The concentration gradient L is positive. (b) The concentration gradient L is negative. $\langle C_s \rangle$ is the average surface concentration, C_0 is the bulk concentration, and δ is the thickness of the diffusion layer.

then expressed as

$$f_z = \frac{\alpha_0 \chi_s}{\mu_0} B \left(\frac{dB}{dz} \right)_{z=0} c, \quad (8)$$

where the subscript z indicates that the magnetic field is oriented in the z -direction.

2.2 Fluctuation equations

In the convective-diffusion layer, with the continuity equations for the velocity fluctuation,

$$\nabla \cdot \vec{u} = 0. \quad (9)$$

The Navier-Stokes equation ignoring the second order of smallness is given by

$$\frac{\partial \vec{u}}{\partial t} = -\frac{1}{\rho_0} \nabla \delta P + \nu \nabla^2 \vec{u} + \frac{1}{\rho_0} \vec{f}, \quad (10)$$

where $\vec{u}$ is the velocity fluctuation, ν the kinematic viscosity, δP the pressure fluctuation, and $\vec{f}$ the gravitational force fluctuation whose vertical component is defined by eq. (8).

After taking the rotation of eq. (10), we can derive the following equation for the z -component of the vorticity.

$$\frac{\partial \zeta}{\partial t} = \nu \nabla^2 \zeta, \quad (11)$$

where $\zeta \equiv \partial v / \partial x - \partial u / \partial y$, and u and v are the x - and y -components of the velocity fluctuation, respectively. Taking the rotation of eq. (11) again, we obtain the equation for the z -component of the velocity fluctuation w in the following

$$\frac{\partial}{\partial t} \nabla^2 w = \frac{1}{\rho_0} \nabla_{\perp}^2 f_z + \nu \nabla^4 w, \quad (12)$$

where $\nabla_{\perp} \equiv (\partial / \partial x, \partial / \partial y)$.

On the other hand, substituting eq. (4) into the diffusion equation of the solute, and neglecting the second order of smallness, we can derive the equation of the concentration fluctuation,

$$\frac{\partial c}{\partial t} = -Lw + D_0 \nabla^2 c, \quad (13)$$

where D_0 is the diffusion coefficient of the solute.

2.3 Boundary conditions

It is assumed that two boundaries of the convective-diffusion layer remain flat, so that the z -component of the velocity becomes zero; therefore

$$w = 0 \quad \text{for } z = 0 \quad \text{and } z = \delta. \quad (14)$$

With this condition, the diffusion layer thickness δ is redefined as the thickness of the convective-diffusion layer. Because of the solid boundary of the solid surface, the x - and y -components must also be kept zero. With the continuity equation (eq. (9)), it follows that

$$\frac{\partial w}{\partial z} = 0 \quad \text{for the boundary at the solid surface.} \quad (15)$$

On the other hand, the boundary on the solution side is thought to be a free surface, so that there is no stress there. Since both the boundaries form flat and horizontal interfaces, the nonstress condition implies that the x - and y -components of the stress tensor are equal to zero, *i.e.*,

$$\frac{\partial u}{\partial z} + \frac{\partial w}{\partial x} = 0, \quad (16a)$$

$$\frac{\partial v}{\partial z} + \frac{\partial w}{\partial y} = 0. \quad (16b)$$

Considering eqs. (9), (14), (16a) and (16b), we can obtain the following boundary condition on the solution side,

$$\frac{\partial^2 w}{\partial z^2} = 0 \quad \text{for the boundary on the solution side.} \quad (17)$$

The concentration fluctuation is also assumed to vanish at both boundaries, *i.e.*,

$$c = 0 \quad \text{for } z = 0 \quad \text{and } z = \delta. \quad (18)$$

Then, from the definition of the z -component of the vorticity ($\zeta = \partial v / \partial x - \partial u / \partial y$) and the stationary conditions ($u = v = 0$) at the solid boundary, the solid surface gives the boundary condition of ζ as follows,

$$\zeta = 0 \quad \text{for the boundary at the solid surface.} \quad (19)$$

Since $\partial u / \partial z = \partial v / \partial z = 0$ holds at the boundary on the solution side,

$$\frac{\partial \zeta}{\partial z} = 0 \quad \text{for the boundary on the solution side,} \quad (20)$$

is derived. Equations (19) and (20) always lead to the static solution $\zeta = 0$ in eq. (11), so that it can be concluded that the flow mode of this convection is a nonvortex type.

2.4 Self-organization of convection cell pattern

The fluctuations of the velocity and the concentration have the following two-dimensional plane waves, *i.e.*,

$$w = w^\circ(z) \exp[i(k_x x + k_y y) + p t], \quad (21a)$$

$$c = c^\circ(z) \exp[i(k_x x + k_y y) + p t], \quad (21b)$$

where k_x and k_y are the x - and y -components of the wave number k ($\equiv (k_x^2 + k_y^2)^{1/2}$), p is the amplitude factor, and $w^\circ(z)$ and $c^\circ(z)$ are the amplitudes. For simplicity, we introduce the

nondimensional coefficients in the following; first, the thickness δ of the convective-diffusion layer is adopted as the unit length. Then, the nondimensional wave number $a = k\delta$ is introduced. At the same time, the coordinate z is replaced by $z'\delta$, changed to a nondimensional number z' . The thickness of the convective-diffusion layer is thus changed from δ to 1. The nondimensional symbol for the derivative $D \equiv d/dz'$ is also used, so that eqs. (12) and (13) are reduced to the following form especially in the steady state.

$$(D^2 - a^2)^3 w^\circ(z') = -S a^2 w^\circ(z'), \quad (22)$$

where S is a nondimensional number defined below, which must be positive for the onset of convection.¹⁸⁾

$$S \equiv \mp \frac{\alpha_0 \chi_s |\Delta C_s| \delta^3}{\mu_0 D_0 \eta} B \left(\frac{dB}{dz} \right)_{z=0}, \quad (23)$$

where “−” and “+” correspond to the positive and negative concentration gradients, respectively, and η ($=\rho\nu$) is the viscosity. The boundary conditions eqs. (14) to (18) to solve eq. (22) are rewritten in the following,

a) for the solid boundary on the solid surface

$$w^\circ(z') = D w^\circ(z') = (D^2 - a^2)^2 w^\circ(z') = 0, \quad (24a)$$

b) for the boundary on the solution side

$$w^\circ(z') = D^2 w^\circ(z') = D^4 w^\circ(z') = 0. \quad (24b)$$

The sign of the product $\alpha_0 \chi_s$ ($= (\partial \chi / \partial C)_\mu$) depends on the magnetism, *i.e.*, plus for paramagnetism and minus for diamagnetism. According to eq. (23), if the negative concentration gradient of the solute with paramagnetism just like the copper sulfate dissolution is chosen, we can expect the convection for $(dB/dz)_{z=0} > 0$. When the concentration gradient of the paramagnetism material is positive, $(dB/dz)_{z=0} < 0$ is required. On the contrary, if we choose the diamagnetism materials, the conditions for the magnetic field gradient are reversed. The necessary and sufficient conditions for the onset of the magneto-convection are summarized as $\mp |\Delta C_s| \chi_s (dB/dz)_{z=0} > 0$, where the signs “−” and “+” correspond to the positive and negative concentration gradients, respectively.

In Fig. 3, we plot the data of S against a .¹⁸⁾ From the minimum value of S corresponding to the most stable state, the critical numbers are obtained; as the critical wave number and the critical nondimensional number, $a_c = 2.682$ and $S_c = 1100.65$ are determined.

From the value of a_c , we can estimate the size of a single convection cell as follows. At first, the dimensional wave number k_c corresponding to a_c is given as $k_c = a_c / \delta$. The wavelength λ_c is thus derived, *i.e.*, $\lambda_c = 2\pi / k_c = 2\pi \delta / a_c$. Within one wavelength, two convection cells moving in opposite directions can be thought to appear, so that the size of a single cell d is estimated by $d = \lambda_c / 2 = \pi \delta / a_c = 1.17\delta$, *i.e.*, nearly equal to the thickness of the convective-diffusion layer. The corresponding amplitude $w^\circ(z')$ is derived as¹⁸⁾

$$w^0(z') = \sin q_0 z' - 0.01707 \sinh q_1 z' \cos q_2 z' + 0.003456 \cosh q_1 z' \sin q_2 z', \quad (25)$$

where $q_0 = 7.1378$, $q_1 = 9.1108$, and $q_2 = 3.7893$.

Fig. 3 Non-dimensional number S (defined by eq. (23)) plotted against non-dimensional wave number a . S_c is the minimum marginal value of S , and a_c is the corresponding non-dimensional wave number, i.e., the critical non-dimensional wave number.

As mentioned above, the convection cell is characterized by a particular wave number a_c . Nevertheless, the pattern which the convection cell will exhibit is completely unspecified. This arises from the fact that a given wave vector $\vec{a}$ can be resolved into two orthogonal components in an infinite number of ways; moreover, the waves corresponding to differing resolutions can be superposed with arbitrary amplitudes and phases. The present theory is unable to distinguish between these many possibilities. However, if we should specify from considerations of symmetry, or otherwise, the type of cell patterns that can prevail, then the theory can describe the details of the resulting cell structure.

Since there are no points or directions in the horizontal plane which are preferred, the convective-diffusion layer must be tessellated into regular polygons with the cell walls being surfaces of symmetry, i.e., a certain periodic cell pattern appears. Therefore, there is a unit cell which repeats itself regularly; on the cell walls the normal gradient of the vertical velocity vanishes. Such condition requires

$$(\vec{n} \cdot \nabla_{\perp})w = 0. \quad (26)$$

Using this condition, we can examine some special patterns, i.e., two specifically simple cases.¹⁹⁾ The simplest possible pattern is a roll-like one, which occurs when all the quantities depend on only one of the two horizontal coordinates, e.g., x' . In this case, the cells are infinitely elongated, which can be written as

$$w = w^{\circ}(z') \cos \frac{2\pi}{l} x', \quad (27)$$

where $l (= 2\pi/a)$ is a constant. Corresponding to this solutions for w , the x - and y -components are given in the following

$$u = -\frac{Dw^{\circ}(z')}{a^2} \frac{2\pi}{l} \sin \frac{2\pi}{l} x' \quad \text{and} \quad v = 0. \quad (28)$$

The width of the roll is l , which is the same as the wavelength

of the cell. A slightly more complex solution is obtained for a rectangular or square cell, i.e.,

$$w = w^{\circ}(z') \cos \frac{2\pi}{l_x} x' \cos \frac{2\pi}{l_y} y', \quad (29)$$

where l_x and l_y are the wavelengths of the cell in the x - and the y -directions; they are related to a by

$$a^2 = 4\pi^2 \left(\frac{1}{l_x^2} + \frac{1}{l_y^2} \right). \quad (30)$$

The horizontal components of the velocity are given by

$$u = -\frac{Dw^{\circ}(z')}{a^2} \frac{2\pi}{l_x} \sin \frac{2\pi}{l_x} x' \cos \frac{2\pi}{l_y} y', \quad (31)$$

$$v = -\frac{Dw^{\circ}(z')}{a^2} \frac{2\pi}{l_y} \cos \frac{2\pi}{l_x} x' \sin \frac{2\pi}{l_y} y'. \quad (32)$$

Finally, we seek the equation of the mass flux enhanced by the magneto-convection. In the present case, the convective-diffusion layer is self-organized on the solid surface, i.e., the most important parameter δ is automatically determined by the formation of the convection cells. This system by itself seeks the most stable nonequilibrium state, being settled there. Accordingly, S always tends to take the critical value S_c , i.e., δ instead of S functions as the adjustable parameter. Therefore, substituting $S = S_c$ into eq. (23), we can derive the equation of δ_c at the critical state¹⁸⁾ as

$$\delta_c = 10.32 \left\{ \frac{\mu_0 D_0 \eta}{|\alpha_0 \chi_s \Delta C_s|} \right\}^{1/3} \left| B \left(\frac{dB}{dz} \right)_{z=0} \right|^{-1/3}. \quad (33)$$

Using the data mentioned later, we can calculate the value of δ_c . Substituting δ_c for eq. (33) in the following equation of the average mass flux density,

$$\bar{j}_{\text{flux}} = D_0 \frac{|\Delta C_s|}{\delta_c}, \quad (34)$$

it follows that

$$\bar{j}_{\text{flux}} = 0.0969 D_0 \left(\frac{\alpha_0 |\chi_s|}{\mu_0 D_0 \eta} \right)^{1/3} |\Delta C_s|^{4/3} \left| B \left(\frac{dB}{dz} \right)_{z=0} \right|^{1/3}. \quad (35)$$

3. Experimental

The high magnetic field was generated by a superconducting magnet (Cryogen-Free 7 T High- T_c Magnet, Sumitomo Electric Industries, Ltd.), which has a vertical bore space at room temperature of 50 mm diameter and the maximum magnetic flux density of 7 T. Figure 4 shows the product of the magnetic flux density and its gradient, $B(dB/dz)$ at 7 T plotted against the position from the upper end of the bore.

Copper sulfate pentahydrate crystals were made by a slow evaporation from a saturated copper sulfate solution at room temperature, where a seed crystal was suspended. Within a few months, it grew up to a triclinic crystal 3 to 4 cm in size. We cut off the (110) crystal face $8 \times 8 \text{ mm}^2$ in area after verifying it by the angle of two different faces of the crystal²⁰⁾ using a diamond cutter. Then, all the crystal faces except for the (110) face were imbedded into the visible light polymeric type of acrylic resin (Maruto Instrument Co., Ltd., Acryl-One

Fig. 4 Vertical distribution of magnetic flux density B and the product of the magnetic flux density and its gradient $B(dB/dz)$ at 7 T.

Fig. 5 Experimental cell for the dissolution of copper sulfate pentahydrate crystal. The crystal was cut to a 8 mm square, and was embedded in acrylic resin to expose the (110) plane into pure water.

#4111). After washing the resin surface with acetone, the crystal surface was polished with emery paper (8000 mesh). The crystal was, as shown in Fig. 5, placed upward on the bottom of a vessel to protect it from the gravitational convection. Then, pure water of 20 mL volume was poured into the vessel, and the vessel was set inside the bore space to fix the solid-liquid interface at the position under a given magnetic field gradient. The water was kept at $18 \pm 1^\circ\text{C}$.

After a certain time lapses, the morphology of the dissolved surface was optically observed by making a replicat of the surface with the SUMP method.^{21,22)} The amount of dissolved copper sulfate ions was electrochemically measured by the anodic stripping method.²³⁾

4. Results and Discussion

To examine the conditions under which the convection develops, we first measured the response of the dissolution rate of copper sulfate pentahydrate crystal at a constant magnetic flux density B by changing only the gradient (dB/dz) . Figure 6 represents the dissolved amount of copper sulfate plotted against the various gradients at a constant magnetic flux density of 4 T.

Since the concentration gradient was negative, according to the estimation based on eq. (23), the dissolved amount increases with the positive gradient, whereas for the negative area, it gave a constant value independent of the gradient. Therefore, the samples were placed at the bore space where the gradient took the positive maximum value. After a certain time lapses, the morphology of the dissolved surface was observed. Figure 7 shows the photos of the dissolved surfaces, which were taken by using the replicas made by the SUMP method.^{21,22)} In a zero magnetic field, *i.e.*, at the zero value of $B(dB/dz)$, the surface is flat without large irregularity (Fig. 7a). However, it can be seen that narrow ridges and valleys about at $500 \mu\text{m}$ intervals emerge at the middle value of $B(dB/dz)$ ($67 \text{ T}^2 \text{ m}^{-1}$, see Fig. 7b). We thus assure that at the middle $B(dB/dz)$, the convection cell is not the more symmetrical three-dimensional one such as a rectangular or hexagonal one, but the less symmetrical two-dimensional one, *i.e.*, the roll-type one (see eqs. (27) and (28)), which develops along a large axis adjoining both neighbor axes of other rolls rotating in opposite direction. This elucidation is also supported by the experimental result of the critical thickness of the convective-diffusion layer; in Fig. 8, the thickness are obtained from eq. (34) by using the mass flux data of copper sulfate. As discussed above, the size of convection cell d_c is given by $d_c = 1.178\delta_c$. Namely, the convection cell size is of the order of $100 \mu\text{m}$, which agrees with the order of the intervals of the ridges and valleys. At the higher value of $B(dB/dz)$, $212 \text{ T}^2 \text{ m}^{-1}$, however as shown in Fig. 7c,

Fig. 6 Average mass flux density against the gradient of the magnetic flux density. The temperature is 18°C , the volume of pure water is 20 mL, and the dipping time is 10 minutes.

Fig. 7 Photos of surface morphology after dissolution of the (110) face under various magnetic forces using the SUMP method (replica method). a, $B(dB/dz) = 0 \text{ T}^2 \text{ m}^{-1}$; b, $B(dB/dz) = 67 \text{ T}^2 \text{ m}^{-1}$; c, $B(dB/dz) = 212 \text{ T}^2 \text{ m}^{-1}$; the dipping time is 10 minutes, the solution temperature is 18°C , and the volume of pure water is 20 mL.

the morphology changes drastically, *i.e.*, the more symmetrical, rectangular-like pattern (see eqs. (29), (31) and (32)) appeared. This strongly suggests that the convection cell pattern changes from a roll-type one to a rectangular one as the vertical magnetic force increases.

To validate the above explanation, we compared the experimental data of the mass flux density for the copper sulfate pentahydrate dissolution with the theoretical values calculated by eq. (35). Figure 9 exhibits the mass flux density plotted against the 1/3rd order of $B(dB/dz)$. A good linearity was obtained, and with the slope of this plot, we derived the concentration coefficient of the magnetic susceptibility of copper sulfate α_0 as follows; in the present case, the bulk solution is pure water, so that $|\Delta C_s|$ is assumed to be equal to the saturated concentration of copper sulfate at 18°C , *i.e.*, $C_{\text{sat}} = 1.256 \times 10^3 \text{ mol m}^{-3}$. Using the experimental data

Fig. 8 Plots of δ_c vs $\{B(dB/dz)\}^{-1/3}$. The solid circle plots calculated data from eq. (34).

Fig. 9 Plots of $\bar{j}_{\text{flux}}$ vs $\{B(dB/dz)\}^{1/3}$. Experimental conditions are the same as those in Figure 6. The solid line is the regression curve, and the broken line is the theoretical curve.

of the slope, $1.409 \times 10^{-3} \text{ mol m}^{-5/3} \text{ s}^{-1} \text{ T}^{-2/3}$, together with the data from other literature^{24,25)} $\mu_0 = 1.256 \times 10^{-6} \text{ H m}^{-1}$, $\chi_s = 1.479 \times 10^{-5}$, $D_0 = 3.79 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$, $\eta = 1.474 \times 10^{-3} \text{ Pa s}$, we can calculate $\alpha_0 = 1.10 \times 10^{-3} \text{ m}^3 \text{ mol}^{-1}$. On the other hand, we can also obtain the same value from the volume dependence of the magnetic susceptibility in the literature,^{24,25)} *i.e.*, $\alpha_0 = 1.29 \times 10^{-3} \text{ m}^3 \text{ mol}^{-1}$. Both data are in good agreement with each other.

Finally, the dependence of the dissolved amount on the concentration difference $|\Delta C_s|$ was examined. In the present case, as mentioned above, $|\Delta C_s| = C_{\text{sat}}$ holds. If we can control the saturated concentration C_{sat} by some means at a constant temperature, we can change the value of $|\Delta C_s|$. As exhibited in Fig. 10, the saturated concentration varies with the concentration of sulfuric acid. Therefore, we measured

Fig. 10 Relationship between the saturated copper sulfate concentration and the sulfuric acid concentration at 18°C.

Fig. 11 Plots of $\bar{j}_{flux}$ vs. $C_{sat}^{4/3}$. $(dB/dz) = 5.0 \times 10^2 \text{ T m}^{-1}$, and $B = 5.1 \text{ T}$. The temperature is 18°C, and the volume of pure water is 20 mL.

the dissolved amounts under various concentrations of sulfuric acid, which were then replaced by the saturated concentration of copper sulfate. As shown in Fig. 11, it was concluded that the dissolved amount is in proportion to the 4/3rd order of $|\Delta C_s|$.

5. Conclusions

From the change of the surface morphology of the dissolved copper sulfate crystal, it was concluded that the pattern

of the convection cell varies from a roll type to a rectangular one as the magnetic flux density increases. However, it could not be theoretically predicted what type of convection cell pattern occurs. This was because the energetics for the formation of such cell patterns can not be discriminated at least within the linear approximation. At the same time, the sizes of the cells were compared with the result of the theoretical calculation; it was decided that both data had the same order of values. Moreover, these results were supported by those of the measurement of mass flux density of dissolved copper sulfate. Namely, the experimental data were, as predicted theoretically, proportional to the 1/3rd order of the product of the magnetic flux density and its gradient and the 4/3rd order of the saturated concentration.

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