

## Electrohydrodynamic Dissipative Structure in Unipolar and Bipolar Injection Cases

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(Received June 19, 1981)

An electrohydrodynamic dissipative structure appearing in a suspension system of  $\text{TiO}_2$  particles in organic solvents is analyzed from a viewpoint of thermodynamics. This analysis gives rather a general explanation for electrohydrodynamic instability caused by unipolar injection and for that caused by bipolar injection. Dimensionless parameters which are the criteria of the hydrodynamic instability are given as  $R = \varepsilon V / \eta \mu$  and  $R = \varepsilon^2 V^2 / n q \mu \eta c^2$  respectively for the former and latter cases. The lattice constant of the hexagonal dissipative structure is derived to be proportional to the thickness of the liquid layer.

It has been known that there exist regular structures which appear only by a constant supply of energy. The Bénard's thermal convection<sup>1–4)</sup> and a convection caused by the Félici instability<sup>5)</sup> are well known examples. In these cases, energy flux is flowing along a potential gradient across a liquid layer whose surfaces contact respectively with two different systems. The energy flux thus generated causes a periodic structure of convection which usually shows a two-dimensional hexagonal lattice and does not have the translational symmetry of the static liquid layer any more. Such lowering of symmetry means that only some components in the wavenumber spectrum of convective disturbance grows. Both the Bénard and Félici instabilities were considered from the above viewpoint by some authors.<sup>3,6,7)</sup> In their treatments the terms of higher order of the small disturbances were neglected and thus linearized equations were solved. In this sense it can be judged by their treatments whether a small fluctuation generated in a fluid system can begin to grow or not, but it is impossible to judge if the growing of the fluctuation can bring the system into a steady convective state.

In the previous paper,<sup>8)</sup> we reported a photoelectroexcited convective structure appearing in a suspension of  $\text{TiO}_2$  particles in an organic solvent. In the system,  $\text{TiO}_2$  particles (1  $\mu\text{m}$  in diameter) had a negative surface charge ( $q_a = -48e$ ,  $e$ ; unit charge) in the dark. When the system was subjected to a d.c. voltage between a pair of transparent electrodes and to light of the intrinsic absorption region of  $\text{TiO}_2$ , the sign of the electric charge of the particles is inverted on the anode surface and thus positively charged particles ( $q_+ = 3500e$ ) showed a photoelectromigration toward the cathode. On the cathode the positively charged particles again acquired a negative charge ( $q_- = -660e$ ) and migrated toward the anode.<sup>9)</sup> This process was repeated during illumination. Consequently,  $\text{TiO}_2$  particles showed an oscillatory motion between the two electrodes under a stationary illumination, and when the applied voltage exceeded a critical value, a convective flow of insulating fluid with a two-dimensional hexagonal symmetry appeared with a lattice constant proportional to the thickness of the suspension layer.<sup>10)</sup> The system looks similar to that of Félici in the sense that the convections in

the both cases were generated by a movement of charged particles through a liquid layer, though the charged particles in the Félici instability are ions or electrons and ours were photosensitive  $\text{TiO}_2$  particles dispersed in the suspension.

The Félici instability was treated by Schneider and Watson<sup>6)</sup> for the case of unipolar injection, while in our case<sup>8–10)</sup> there are positively charged  $\text{TiO}_2$  particles and negatively charged ones in a suspension system. In the case of Schneider and Watson, the problem was treated as a space charge limited conduction of unipolar carriers. In the present case, however, there is no space charge in the static state with no macroscopic convection. Thus the both cases are physically different from each other. In the present paper we consider which of the two stationary states, *i.e.* the convective state and the non-convective state of the present system, is more favorable and discuss the transition between the two states from the irreversible thermodynamic standpoint, instead of discussing the problem in terms of the growth of small convective fluctuations. The size of a convection cell in the stationary state is estimated with the condition for minimum energy dissipation.<sup>11)</sup>

**Photoelectrophoresis without Convection.**  $\text{TiO}_2$  particles may be in two charged states under the stationary illumination, *i.e.* the positively charged state ( $q_+ = 3500e$ ) and the negatively charged state ( $q_- = -660e$ ).<sup>9)</sup> Under an electric field,  $E$ , the velocity of  $\text{TiO}_2$  particles is given by Stokes' law,

$$v_{\pm} = \frac{q_{\pm} E}{6\pi\eta R}, \quad (1)$$

where  $R$  and  $\eta$  are the radius of a  $\text{TiO}_2$  particle and the viscosity of the surrounding liquid, respectively. Then, the mobility of the particles of each electric charge is given as

$$\mu_{\pm} = \frac{q_{\pm}}{6\pi\eta R}. \quad (2)$$

We take the Cartesian coordinates where the  $z$ -axis is in the direction vertical to the electrodes (see Fig. 1). We express the number density of total  $\text{TiO}_2$  particles, that of the particles carrying positive charge and that carrying negative one by  $n(z)$ ,  $n_+(z)$  and  $n_-(z)$ , respectively. Among the above three quan-

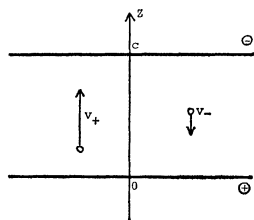


Fig. 1. Co-ordinates of the system for analysis, the anode being at  $z=0$  and the cathode at  $z=c$ .

ties the following relation should hold,

$$n(z) = n_+(z) + n_-(z).$$

By the law of conservation for the particles of each electrical sign under a stationary state, we have

$$\frac{\partial[n_+(z) \cdot v_+(z)]}{\partial z} = \frac{\partial[n_-(z) \cdot v_-(z)]}{\partial z} = 0. \quad (3)$$

The continuity condition at the electrode surfaces ( $z=0$  and  $z=c$ ) in a stationary state is given as

$$n_+(0) \cdot v_+(0) + n_-(0) \cdot v_-(0) = n_+(c) \cdot v_+(c) + n_-(c) \cdot v_-(c) = 0. \quad (4)$$

From Eqs. 3 and 4, we have a general condition holding also in the bulk of the liquid layer as

$$n_+(z) \cdot v_+(z) + n_-(z) \cdot v_-(z) = 0. \quad (5)$$

Using Eqs. 1 and 5, we can obtain

$$n_+(z) \cdot q_+(z) + n_-(z) \cdot q_-(z) = 0, \quad (6)$$

that is, there exists no space charge in the bulk of the liquid layer. This suggests that the electric field in the bulk is uniform and can be expressed as  $V/c$ , there  $V$  is the applied voltage. In consequence, the distribution function of particles,  $n_+(z)$  and  $n_-(z)$ , are also uniform from Eq. 3. Thus the current density in such a stationary state is given as

$$i = n_+ \cdot q_+ \cdot v_+ + n_- \cdot q_- \cdot v_- = nq\mu V/c, \quad (7)$$

where  $q \equiv \sqrt{|q_+ \cdot q_-|}$  and  $v_{\pm} = \mu_{\pm} E = \mu_{\pm} V/c$ .

**Photoelectrophoresis with Macroscopic Convection Stream Field:** As reported before,<sup>8,10</sup> the structure of convection showed a two-dimensional hexagonal lattice. In Fig. 2 intersections of solid lines are the lattice points with the lattice constant,  $a$ , and the hexagon given with broken lines is the Wigner-Seitz's unit cell. Vertices, A, B, C, D, E, F of the Wigner-Seitz's unit cell form a dual lattice. If the hexagonal lattice corresponds to the ascending flow, then its dual lattice corresponds to the descending flow of the fluid. The number of lattice points within the dual lattice is twice of that of original hexagonal lattice. Therefore, the ascending flow and the descending flow of the fluid are not symmetrical. This might be the result of the fact that the magnitude of charges carried by a positively charged particle and that by a negatively charged one are not equal. In a unit cell ABCDEF, the fluid flows up at the lattice point O, and spreading in the six directions, it falls downward at the boundary of this unit cell, that is, this flow is totally symmetric within this cell.

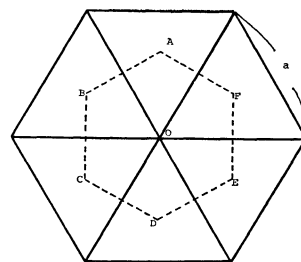


Fig. 2. Two-dimensional hexagonal lattice and the unit cell.

Intersections of real lines are the lattice points. The hexagon shown with dashed line is a Wigner-Seitz's unit cell.

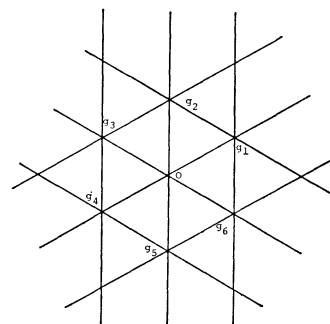


Fig. 3. Reciprocal lattice of convective structure.

We assume that the  $z$ -component,  $v_z(\mathbf{r})$ , of the hexagonal stream field,  $\mathbf{v}(\mathbf{r})$ , can be expressed as the following form,

$$v_z(x, y, z) = v \cdot f(x, y) \cdot g(z),$$

where  $v$  is a constant with the dimension of velocity. Then,  $f(x, y)$  must satisfy a periodicity of the two-dimensional hexagonal lattice and  $g(z)$  must satisfy a boundary condition at the electrode surfaces. The components of larger wavenumber in  $f(x, y)$  will be unfavorable for a stationary state because they will make the larger velocity gradient in the bulk of liquid layer and increase the energy dissipation rate. Thus, taking account of the total symmetry of the flow, we assume that  $f(x, y)$  is expressed by Eq. 8, which is constructed with six plane-wave whose wavenumbers are equal to the reciprocal lattice in Fig. 3,  $\mathbf{g}_i$  ( $i=1, 2, 3, 4, 5, 6$ ).

$$f(\mathbf{r}_1) = \frac{1}{6} \sum_{i=1}^6 \exp(i\mathbf{g}_i \cdot \mathbf{r}_1), \quad (8)$$

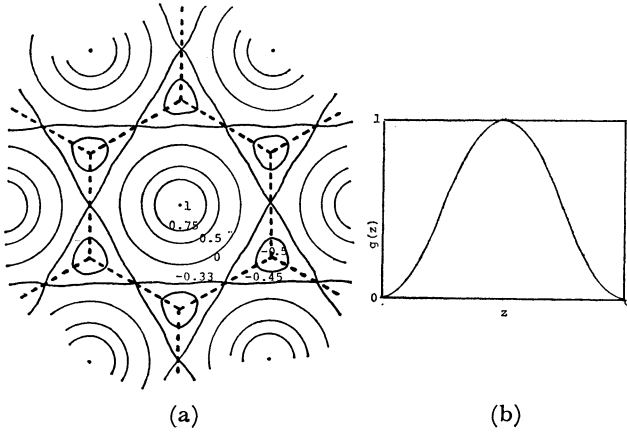
where  $\mathbf{r}_1 \equiv (x, y)$  and the factor,  $1/6$ , is a normalization constant making  $f(x, y)$  unity at the lattice points. The length of the shortest reciprocal lattice vectors are  $g=4\pi/\sqrt{3}a$ . Figure 4a illustrates the profile of the function,  $f(x, y)$ .

With the continuity condition of incompressible fluid flow,

$$\text{div } \mathbf{v} = 0, \quad (9)$$

and the relation

$$\left( \nabla_1^2 + \frac{4}{3}a^{*2} \right) f = 0, \quad (10)$$

Fig. 4. Profiles of functions,  $f(x, y)$ : (a) and  $g(z)$ : (b).

where  $\nabla_1 \equiv \left( \frac{\partial}{\partial x}, \frac{\partial}{\partial y} \right)$  and  $a^* \equiv 2\pi/a$ , one can obtain the stream field as

$$\begin{aligned} v_x(x, y, z) &= \frac{3}{4} \frac{1}{a^{*2}} v \frac{\partial f(x, y)}{\partial x} \frac{dg(z)}{dz}, \\ v_y(x, y, z) &= \frac{3}{4} \frac{1}{a^{*2}} v \frac{\partial f(x, y)}{\partial y} \frac{dg(z)}{dz}, \end{aligned} \quad (11)$$

and

$$v_z(x, y, z) = v \cdot f(x, y) \cdot g(z).$$

The function,  $g(z)$ , must be chosen to satisfy the boundary condition,  $v_x = v_y = v_z = 0$ , at the electrode surfaces ( $z=0, c$ ), so we assume that  $g(z)$  has the form as Eq. 12, which satisfies the above conditions with the smallest wave number.

$$g(z) = \frac{1}{2}(1 - \cos(c^*z)) \quad (c^* \equiv 2\pi/c), \quad (12)$$

where the factor,  $1/2$ , is a normalization constant making  $g(z)$  unity at  $z=c/2$ . The function  $g(z)$  is illustrated in Fig. 4b.

Thus, the Eq. 11, together with Eqs. 8 and 12, give the stream field of two-dimensional hexagonal convective domain with the thickness  $c$  and the lattice constant  $a$ .

**Dissipation Function:** We assume that  $\text{TiO}_2$  particles move on the stream of fluid, then an energy dissipation by the friction between the particles and the fluid will be neglected compared with that within the fluids. The amount of heat generated in a unit volume and a unit time at any point in the fluid is given by the viscous dissipation function,

$$\Phi(x, y, z) = \frac{1}{2} \eta \sum_{i,j} \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right)^2. \quad (13)$$

Substituting Eq. 11 into  $v_i$  in Eq. 13, one obtains

$$\begin{aligned} \Phi(x, y, z) &= \eta v^2 \left[ \left\{ 4f^2 + \frac{9}{4} \frac{1}{a^{*2}} \left( \left( \frac{\partial^2 f}{\partial x \partial y} \right)^2 - \frac{\partial^2 f}{\partial x^2} \frac{\partial^2 f}{\partial y^2} \right) \right\} \left( \frac{dg}{dz} \right)^2 \right. \\ &\quad \left. + \left\{ \left( \frac{\partial f}{\partial x} \right)^2 + \left( \frac{\partial f}{\partial y} \right)^2 \right\} \left( g + \frac{3}{4} \frac{1}{a^{*2}} \frac{d^2 g}{dz^2} \right)^2 \right]. \end{aligned} \quad (14)$$

Substituting Eqs. 8 and 12 into Eq. 14 and integrating it over a unit cell, then dividing the result by the unit cell volume,  $(\sqrt{3}/2)a^2c$ , we obtain the mean viscous dissipation function,  $\langle \Phi \rangle$ ,

$$\begin{aligned} \langle \Phi \rangle &= \int_{\text{unit cell}} \Phi(x, y, z) v / \frac{\sqrt{3}}{2} a^2 c \\ &= \eta \left( \frac{v^2}{c^2} \right) \frac{\pi^2}{16} \left[ \frac{a^2}{c^2} + \frac{16}{3} \frac{c^2}{a^2} + 8 \right] \\ &= \eta \frac{v^2}{c^2} \pi^2 \left[ \frac{1}{3} \gamma^2 + \frac{1}{16} \gamma^{-2} + \frac{1}{2} \right], \end{aligned} \quad (15)$$

where  $\gamma = c/a$  is the axial ratio of this stream.

**Lattice Constant:** The stream field,  $v_x, v_y, v_z$ , contains the lattice constant,  $a$ , as a parameter, since  $f(x, y)$  must satisfy the hexagonal symmetry. Consequently, the parameter,  $a$ , also appears in the expression of the mean dissipation function,  $\langle \Phi \rangle$ . In Fig. 5, the variation of  $\langle \Phi \rangle$  corresponding to the virtual variation of the parameter is illustrated. Figure 5 will be interpreted as follows: Under a condition in which the lattice constant is smaller enough than the distance between the two electrodes, i.e. the thickness of the suspension layer,  $c$ , a large velocity gradient of  $v_z$  would occur in the  $xy$ -plane, making a large energy dissipation (Fig. 6a). On the other hand, larger enough value of the lattice constant compared with  $c$  also increases the dissipation, because the region of fluid transport along the direction perpendicular to the  $z$  axis increases and the directions of the flow in the upper and lower parts this region are opposite to each other, so the region where a gradient of  $v_x$  and that of  $v_y$  along  $z$  axis are large becomes wider (Fig. 6c). Consequently a lattice constant of the same order as the thickness of the liquid layer would be most favorable for the smallest dissipation rate of energy (Fig. 6b). We do not have any *a priori* principle to determine the stationary state under a given condition thermodynamically. However, if we assume that the principle of the minimum energy dissipation which

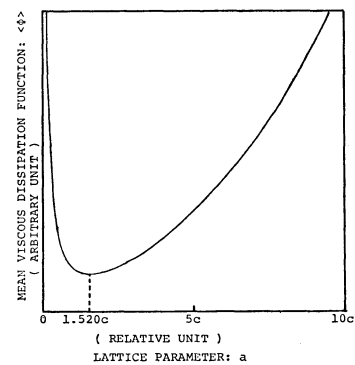
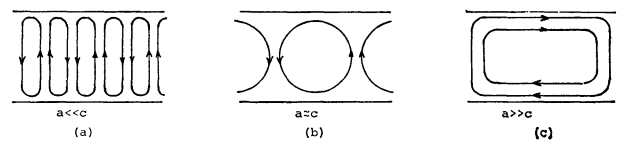
Fig. 5. Variation of  $\langle \Phi \rangle$  corresponding to the variation of lattice parameter,  $a$ .

Fig. 6. Illustration of the shapes of cellular convective flows.

(a): Lattice constant,  $a$ , is smaller than the electrode spacing,  $c$ , (b):  $a$  is nearly equal to  $c$ , (c):  $a$  is larger than  $c$ .

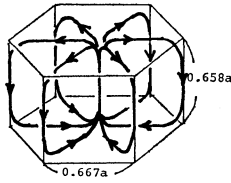


Fig. 7. Most favorable structure of convection.

has been known to be applied close to equilibrium also holds at the stationary state where a dissipative structure has been formed, the most favorable lattice constant can be estimated from Eq. 15 as

$$a = 2 \times 3^{-1/4} c \approx 1.520c. \quad (16)$$

Thus the lattice constant is proportional to the thickness of the liquid layer. The axial ratio of the stream,  $\gamma$ , is given as  $\gamma=0.658$  (Fig. 7). The experimental values of  $\gamma$  was 0.86 as reported in the previous paper, which was larger than the theoretical value to some extent. The mean viscous dissipation function at  $a=1.520c$  is given as

$$\langle \Phi \rangle = \eta \frac{v^2}{c^2} \frac{\pi^2}{2} \left( 1 + \frac{1}{\sqrt{3}} \right). \quad (17)$$

**Electric Current by Convection:** Assuming that  $\text{TiO}_2$  particles are moving on the convection stream of the surrounding fluid, their velocity coincides with that of convection of the fluid,  $\mathbf{v}(\mathbf{r})$ .

From the continuity equation of the flow in a stationary state,  $\text{div } \mathbf{v}=0$ , and that of the  $\text{TiO}_2$  particles flow,  $\text{div}(n\mathbf{v})=0$ , we obtain

$$\frac{dn}{dt} = \mathbf{v} \cdot \text{grad } n = \text{div}(n\mathbf{v}) - n \cdot \text{div } \mathbf{v} = 0. \quad (18)$$

Thus, in a stationary state, there is no change in the particle number density along a stream line. Therefore, there is also no change in the charge density along the stream line. More correctly, since the particles can discharge at the electrodes, there are two regions in a unit cell in each of which  $v_z$  is positive or negative, and the charge density along a stream line is kept constant within each region.

The mean number density of  $\text{TiO}_2$  particles,  $n$ , in the present system was  $1.2 \times 10^{11} \text{ (cm}^{-3}\text{)}$ ,<sup>9)</sup> so if each of them have a positive charge ( $q_+=3500e$ ) and are distributed uniformly in the xy-plane, the charge density on a unit area of a electrode,  $\sigma_p$ , is  $3.15 \times 10^{12} e \text{ (cm}^{-2}\text{)}$ . The surface charge induced on an electrode by the application of external voltage,  $\sigma_e$ , is  $\epsilon V/c$ , where  $\epsilon$  is the dielectric constant of the fluid. In the present case we assume  $\epsilon=3\epsilon_0$ , where  $\epsilon_0$  is the dielectric constant of vacuum. Then, when an external voltage of 100 V is applied between the electrodes, the value of  $\sigma_e$  is  $2.25 \times 10^{10} e \text{ (cm}^{-2}\text{)}$ , thus  $\sigma_p$  is about 100 times as large as  $\sigma_e$ . When all the particles have a negative charge ( $q_-=-660e$ ), the absolute value of  $\sigma_p$  is about 20 times as large as  $\sigma_e$ . If the discharge of the  $\text{TiO}_2$  particles at the electrodes is a fast process, there exists positive charge in the region of positive  $v_z$ , while in the region of negative  $v_z$ , there exists negative charge. Thus, spatial separation of the positive charge and the negative charge

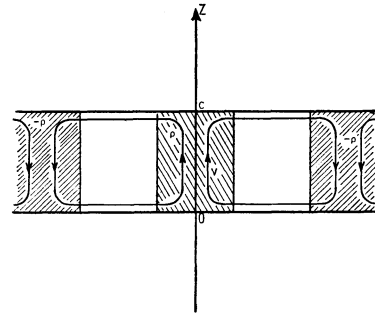


Fig. 8. Model of convective unit cell.

takes place in the periodical convective state, while there is no space charge in the static state without convection. However, the absolute value of the charge density per unit area of the electrode in each region does not exceed  $\sigma_e$ , since, for instance in the region of positive  $v_z$ , the increase of the positive charge density reduces the potential gradient near the anode and prevents the discharge of negatively charged particles at the anode, consequently too much increase of the positively charged particles in the region is suppressed. Electric current under this condition is just a space charge limited one. The situation is same in the region of negative  $v_z$ .

From the consideration mentioned above, together with Eq. 18, we assume a simple model for such a unit convective cell as shown in Fig. 8. It is assumed that the unit cell can be separated into three regions; the first one is the region in which the positively charged particles are distributed uniformly with the charge density,  $+\rho$ , and the fluid flows upward. The second region whose cross section area is equal to that of the first one, is that in which the negatively charged particles are distributed uniformly ( $-\rho$ ) and the fluid falls downward, and the third region between the above two regions is that in which no charge density and no fluid motion exist. The velocity of fluid is  $v$  in the first and  $-v$  in the second region. From Poisson's equation, we obtain

$$\rho \approx \frac{2\epsilon V}{c^2}, \quad (19)$$

where  $V$  is the applied voltage. The profile of the electric field in the suspension layer along  $z$  axis is given as follows:

$$E_1(z) \approx \frac{\rho}{\epsilon} z \quad (\text{in the region of positive charge}), \quad (20)$$

$$E_2(z) \approx \frac{\rho}{\epsilon} (c-z) \quad (\text{in the region of negative charge}). \quad (21)$$

Then we obtain the force by the external field acting on the fluid in the stream tube of a unit cell along a stream line as

$$\begin{aligned} F &\approx \frac{1}{2} S_e \int_0^c \rho E_1(z) dz - \frac{1}{2} S_e \int_0^c (-\rho) E_2(z) dz \\ &= 2S_e \epsilon V^2 / c^2, \end{aligned} \quad (22a)$$

where  $S_e$  is a sum of the cross section area of the first

and the second regions, *i.e.*  $S_e$  corresponds to the conducting area in a unit cell. If we assume that  $S_e$  is approximately equal to a half of the total cross section area of a unit cell,  $S_e = c^2$ , we obtain

$$F \approx 2\varepsilon V^2. \quad (22b)$$

Therefore, a work done by the external electric field on the fluid in a unit cell and a unit time is given as

$$W_e = v \cdot F \approx 2v\varepsilon V^2. \quad (23)$$

On the other hand, the energy dissipation within a unit cell and a unit time is written as

$$W_i = 2c^3 \langle \Phi \rangle = \pi^2 \left( 1 + \frac{1}{\sqrt{3}} \right) \eta v^2 c, \quad (24)$$

where  $2c^3$  is the volume of a unit cell. Since  $W_e$  and  $W_i$  are equal to each other in the steady state, we obtain the following expression for  $v$  by equating Eq. 23 to Eq. 24,

$$v \approx \frac{2}{\pi^2 \left( 1 + \frac{1}{\sqrt{3}} \right)} \frac{\varepsilon V^2}{\eta c} = 0.128 \frac{\varepsilon V^2}{\eta c}. \quad (25)$$

One can see in Eq. 25 that  $v$  is proportional to the square of the applied voltage. Consequently, the average current density,  $\langle i \rangle$ , is proportional to  $V^3$ :

$$\langle i \rangle \approx \frac{1}{2} \rho v = 0.128 \frac{\varepsilon^2 V^3}{\eta c^3}. \quad (26)$$

*Transition from the Static State of Fluid to Convective State.* When a d.c. voltage is supplied to the  $\text{TiO}_2$  particles dispersion system under illumination,

migration of the particles transports electric charge between the electrodes in the nonconvective state. The charge transport in this state occurs against the friction between  $\text{TiO}_2$  particles and their surrounding insulating fluid, and the current is proportional to the applied voltage as shown in Eq. 7. On the other hand,  $\text{TiO}_2$  particles are carried by a flow of fluid in the convective state. In other words, electric charge is transported by a flow of charged fluid between two electrodes. The charge transport in this state occurs against the friction within the fluid. The current in that state is proportional to the cube of the applied voltage as shown in Eq. 26. The two kinds of currents in the both states are illustrated in Fig. 9 as a function of the applied voltage. In the region

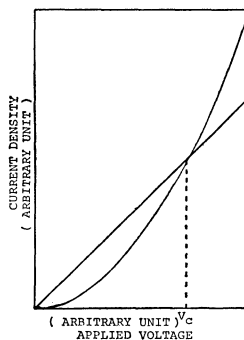


Fig. 9. Current-voltage curves by two state of conduction in bipolar injection.

A transition between two states occurs at the critical voltage,  $V_c$ .

of lower voltage than  $V_c$ , which is the voltage at which two  $i$ - $V$  curves in Fig. 9 cross each other, the static state is more favorable than the convective state for electric conduction. On the other hand, in the region of higher voltage than  $V_c$ , the state with the convection becomes more favorable. Consequently, a transition between the two states occurs at the critical voltage,  $V_c$ . From Eqs. 7 and 26, the critical voltage,  $V_c$  is given as

$$V_c \approx 2.8 \frac{\sqrt{nq\mu\eta}}{\varepsilon} c. \quad (27)$$

As mentioned in the experimental section of the previous paper,<sup>9,10)</sup>  $n = 1.2 \times 10^{11} \text{ cm}^{-3}$ ,  $q = 1520e$ ,  $\mu = 1.72 \times 10^{-5} \text{ cm}^2/\text{V s}$ ,  $\eta = 7.5 \text{ cP}$ ,  $c = 50\text{--}100 \text{ }\mu\text{m}$ ,  $\varepsilon = 3\varepsilon_0$ . When  $c = 75 \text{ }\mu\text{m}$ , we estimate  $V_c \approx 153 \text{ V}$ . The experimental value of the critical voltage<sup>10)</sup> was about 200 V which is in good agreement with the theoretical value. A dimensionless parameter indicating the transition between the states is given from Eq. 27 as

$$R_1 = \frac{\varepsilon^2 V^2}{nq\mu\eta c^2}. \quad (28)$$

From the above consideration we may expect that the transition occurs at  $R_1 \approx 8$ .

*Electrohydrodynamic Instability in the Case of Unipolar Injection.* The injection of carriers of one kind and one sign is called as "unipolar injection."

In such a case a space charge limited conduction may occur even in the static fluid state if the amount of the injected carriers is large enough (Fig. 10a). Such a case was analyzed by Schneider and Watson<sup>6)</sup> and Atten and Moreau,<sup>12)</sup> by solving a set of equations which express the turbulence in fluid in the system. However, our method mentioned above is also adequate for such a case and can give a reasonable explanation.

In the space charge limited conductive state without convection, if the charge carriers are of one kind with mobility  $\mu$ , the current in such a state is given by the Langmuir-Child's law,<sup>13)</sup>

$$i = \frac{9}{8} \frac{\varepsilon \mu}{c^3} V^2. \quad (29)$$

Now we consider the current carried by a convective flow in the case of the unipolar injection. We assume tentatively that the carriers are cations. If all the cations which flow toward the cathode do not discharge as the cathode surface completely, a portion of the cations will remain in the flow toward the anode. When the probability of discharge of cations on the cathode is  $\kappa$ , the charge density in the downward flow is given as  $\rho' = (1 - \kappa)\rho$ , where  $\rho$  is the charge

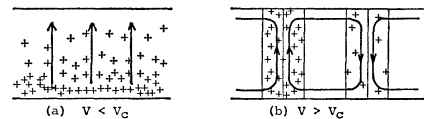


Fig. 10. Current-voltage curves by two states of conduction in unipolar injection.

A transition between two states occurs at the critical voltage,  $V_c$ .

density in the upward flow, *i.e.*  $\rho \approx 2\varepsilon V/c^2$  (see Fig. 10b). And the force acting on the region of upward flow along a stream line in a unit cell,  $F_1$ , is given in the same manner as Eq. 22,

$$F_1 \approx \varepsilon V^2.$$

Similarly, the force acting on the region of downward flow along a stream line,  $F_2$ , is given as

$$F_2 \approx -(1-\kappa)\varepsilon V^2.$$

Then, the work exerted on a unit cell within a unit time,  $W_e$ , is

$$W_e = v(F_1 + F_2) \approx \kappa v \varepsilon V^2. \quad (30)$$

Using Eqs. 24 and 30, we obtain the expression for the flow velocity,  $v$ , as

$$\begin{aligned} v &\approx \frac{\kappa}{\pi^2 \left(1 + \frac{1}{\sqrt{3}}\right)} \frac{\varepsilon V^2}{\eta c} \\ &= 0.064 \kappa \frac{\varepsilon V^2}{\eta c}. \end{aligned} \quad (31)$$

Therefore, the average current density,  $\langle i \rangle$ , is given as

$$\begin{aligned} \langle i \rangle &\approx \frac{1}{4} \kappa \rho v \\ &\approx 0.032 \kappa^2 \frac{\varepsilon^2 V^3}{\eta c^3}. \end{aligned} \quad (32)$$

One can see from Eqs. 29 and 32 that the space charge limited current without convection is proportional to the square of the applied voltage but, on the other hand, the current by the convection is proportional to the cube of the voltage. Current densities in the both conduction regimes are illustrated in Fig. 11 which shows that, in the region of the lower voltage, the static state without convection is more favorable for the electric conduction, and in the region of the higher voltage, the convective state is more favorable. The critical voltage,  $V_c$ , at which the transition between the two states occurs is given by comparing Eqs. 29 and 32,

$$V_c \approx \frac{27.8}{\kappa^2} \frac{\eta \mu}{\varepsilon}. \quad (33)$$

A dimensionless parameter indicating the transition is given by Eq. 33 as

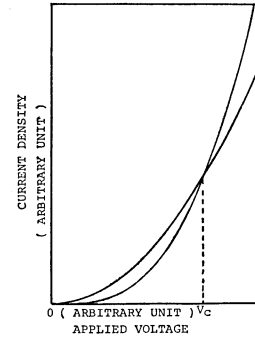


Fig.11. Current voltage curves in the two conduction regimes under unipolar injection.

A transition occurs at the critical voltage,  $V_c$ .

$$R_2 = \frac{\varepsilon V}{\eta \mu}. \quad (34)$$

The dimensionless parameter,  $R_2$ , coincides with one derived by Schneider and Watson<sup>6)</sup> as the criterion of the stability. The transition is expected to occur at  $R_2 \approx 27.8/\kappa^2$ , while Schneider and Watson calculated the value of  $R_2$  as 99<sup>6)</sup> although their boundary condition is somewhat different from the present case.

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