SEDIMENTATION IN TILTED VESSELS(1)

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Not a few papers have been published on *Boycott's effect*—apparent acceleration of sedimentation when the tube is tilted—among which we are to mention Nakamura and Kuroda's work as the standard one. Nakamura's theory, however, is a phenomenology, and he does not go into details of the mechanism of the "*levelling action*," which plays an important rôle in his theory. Recently, several workers have performed theoretical calculations on *Boycott's effect* assuming that the *levelling* is due to diffusion. But the author suspected the validity of this assumption, and carried out a series of experiments to prove that the *levelling* is not due to diffusion, but to a hydrodynamical current in the suspending medium (a kind of convection) caused by the uneven pressure distribution. The work was executed with the assistance of a number of students of the Faculty of Engineering.

§ 1. Introduction

Boycott(2) found in 1920 that the sedimentation of red blood corpuscles is apparently accelerated when the test-tube is tilted (Fig. 1). Many workers, mainly of the medical circle, have since engaged in the study of this curious phenomenon(3), among which we should quote Nakamura and Kuroda's work(4), (5) as the most complete.

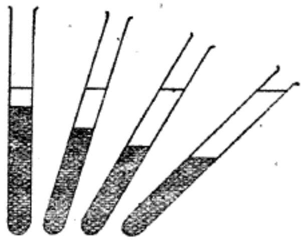

Fig. 1.

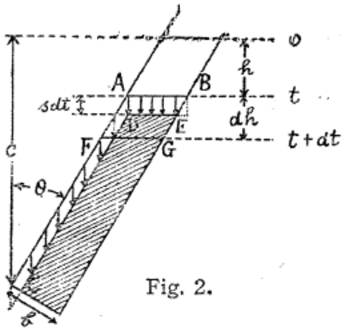

Fig. 2.

Nakamura's idea is as follows: Let the upper boundary of the suspended phase which at the beginning of time $t = 0$ coincided with the surface of the suspending medium be at AB at time t (Fig. 2). Now if the sedimentation velocity of the particles be s (which is considered to be identical with the velocity of descent of the boundary surface when the tube is held vertical), we should primarily expect

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(1) Short notes on this series of experiments have been published in the *Kagaku* (in Jap.). Kinosita-Hukaya-Taki-Isomura, Kagaku, **18** (1948), 125; Kinosita-Murase, *ibid.* **19** (1949), 230; Kinosita-Tsukada, *ibid.*, (in press).

(2) A. E. Boycott, Nature, **104** (1920), 532.

(3) Resumé of these investigations can be found in Ref. (4).

(4) H. Nakamura and K. Kuroda, Keijo J. Med., 8 (1937), 265 (in Fr.).

(5) H. Nakamura, J. Appl. Phys. Jap., **12** (1943), 410 (in Jap.).

that the space marked with arrows of length *sdt* would be clarified after the lapse of a short time interval *dt*. In reality, however, the boundary *CD* being unstable, the particles in the hatched volume drift to fill up the blank space above *CD* and form a horizontal boundary *FG*. The final result to be observed is thus nothing more than a descent by *dh* of the boundary from *AB* to *FG*. Nakamura assumes in the above process that the volume *ABGF* equals the volume of the arrowed space. In the case of a rectangular cylinder shown in Fig. 2, we can write*

$$\text{the arrowed volume} = \{b \sec\theta + (c - h)\tan\theta\} as\, dt$$

and

$$\text{the volume } ABGF = ab \sec\theta \cdot dh,$$

where a represents the depth of the vessel. Equating these and solving for h, we get

$$h = (c + b\sec\theta)\left(1 - e^{-\frac{st\sin\theta}{b}}\right)$$

which for the same value of the argument t is positively larger than

$$h = st,$$

the amount of sedimentation when $\theta = 0°$.

The relation described in the foregoing paragraph can also be stated in the form that the volume clarified in the lapse of time *dt* equals the horizontal projection of the upper boundaries of the suspended phase (including the free surface and the ones in contact with the upper walls) multiplied by *sdt*. The walls beneath the suspension have no essential influence**. Hence the relations illustrated in Fig. 3.

Fig. 3.

Sedimentation velocity, or, strictly speaking, the *descent velocity of the upper boundary of the suspended phase* in any vessel, can be calculated according to this simple principle, and the theory seems to explain the experimental data satisfactorily; at least in Nakamura and Kuroda's experiments on the sedimentation of cattle blood corpuscles the accordance of the theory and the observation is quite splendid.

Nakamura's theory, however, is a phenomenology, and the mechanism of the drift of the particles to form a horizontal boundary—which we call in short the "*levelling action*" after Katsurai⁽⁶⁾—remains ambiguous. Nakamura employed the term "diffusion"; and some workers⁽⁷⁾⁽⁸⁾ are trying theoretical calculations on the basis of this assumption. But there is doubt if the *levelling* is actually due to diffusion, which means the drift of particles without any essential mass motion of the suspending medium.

The author understands the sedimentation with a well-marked boundary, which

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* Triangular volumes in the corners shown with dotted lines are neglected.

** On a horizontal bottom the particles are merely piled up; on an inclined wall some of them adhere and others slide down slowly. In any case, the motion of the suspended particles is not influenced essentially.

(6) T. Katsurai, *Theory of Colloid*, p. 62, 1947 (in Jap.).

(7) N. Saito and S. Oka, Kagaku, **18** (1948), 75 (in Jap.).

(8) O. Miyatake, An address at a Sectional Meeting of the Physical Society of Japan, Oct., 1948.

we treat here, to be characteristic of the case where diffusion is negligibly small†. Essential features of the sedimentation of this type might be pictured as follows: The particles descend with a uniform velocity so as to keep their initial relative positions; when they reach the bottom they settle and are piled up in order; the result is a maintenance of a nearly constant concentration in the suspended phase*. Careful examination will reveal that these ideas lie in Nakamura's theory itself.

Hence it seems unjustifiable to attribute the *levelling action* to diffusion. An alternative mechanism to be proposed here is a kind of convection, a hydrodynamic flow in the suspending medium which carries the particles with itself. Our principal interest in the following series of experiments lay in the verification of this working hypothesis.

§ 2. Sedimentation of Ammonium Chloride Smoke**,(9)

Ammonium chloride smoke was prepared by the usual process, poured gently into a vessel†† illustrated in Fig. 4, and its sedimentation was observed in detail.

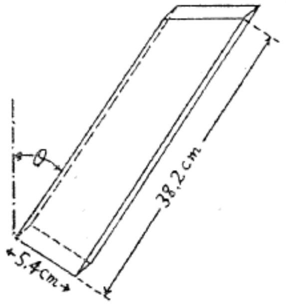

Fig. 4.

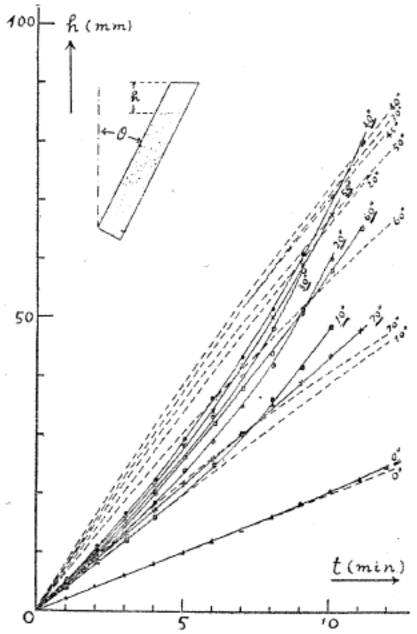

Fig. 5.

Fig. 5 shows the relation between the amount of descent of the boundary surface h and time t, observed for various inclination angles θ. It is clear at one glance that the descent velocity has a maximum near $\theta = 40°$. Broken lines were calculated after Eq. (1), in which s was taken to be equal to 2.0 mm/min., the observed value of the descent velo-

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† Well-kown exponential-tipe concentraton distribution is expected otherwise.

* The picture given here naturally concerns sedimentation in a vertical tube. Actual observations proved that this picture is substantially right, but details are more complicated. Cf. § 5.

** Experiments of this section were carried out by the assistance of Hukaya.

(9) Of the sedimentation of this aero-sol, T. Tachibana and H. Terada observed *Boycott's effect* recenty: J. Chem. Soc. Jap., **68** (1947), 2 (in Jap.).

†† We at first used a test-tube-type vessel of 3 cm dia. with ordinary cork, but found it inadequate because the inner side of the cork plays the rôle of a "tilted wall" and introduces unnecessary complications.

city of the boundary surface when the cylinder was held vertical. For a general idea of the accuracy of the observations refer to Fig. 6, where the original records of repeated measurements for the case $\theta = 40°$ are reproduced.

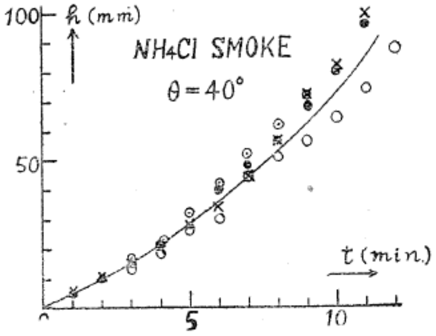

Fig. 6.

The smoke was washed by passing through water, dried through dessicative and directly introduced into the vessel; hence the size of the particles was not uniform*, and coagulation occurred soon after the commencement of the observation. The upward bend of the h-t curves at $t > 4 \sim 5$ min. may presumably be due to this latter effect. We dare not conclude this because the trend is not clear when $\theta = 0°$, but it would not be unreasonable to suppose that the coagulation is promoted when $\theta > 0°$ by the convection current which, as we shall show later, is the cause of the *levelling action* in this case.

In Fig. 7, the initial values of the descent velocity of the boundary suface estimated from Fig. 5 are compared with $(dh/dt)_{t=0}$ derived from Eq. (1). The value of θ which makes the latter maximum can be calculated analytically, and is found to be 41° in our case, which is in good agreement with the observational result. Thus Nakamura's theory explains the experimental results qualitatively very well, but the quantitative values of h or dh/dt calculated from the theory are not very satisfactory.

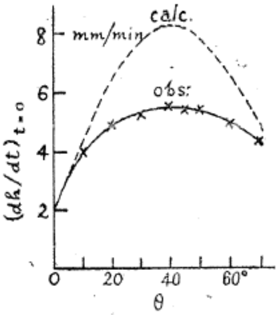

Fig. 7.

Fig. 8 illustrates the change in the appearance of the boundary accompanying the progress of sedimentation**. These sketches will suffice to show the fact that the *levelling* is mainly due to a current of air (a kind of convection) in the vessel. We shall have a small discussion on the mechanism of this convection later (§ 6), but the situation could be comprehended in the rough if one considers the distribution of the pressure exerted by the suspended system upon the medium (in this case, air): the diminution of the concentration of particles in the arrowed space in Fig. 2 will make the magnitude of the component of the gravitational pressure in the wall direction decrease near the upper tilted wall and increase near the lower one, the difference being decidedly

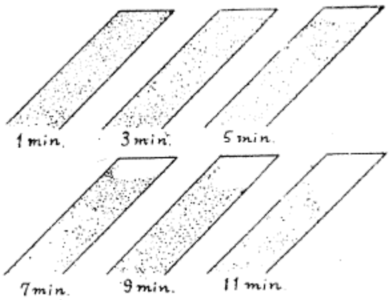

Fig. 8.

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* Mean diameter of the smoke particles was roughly estimated at 0.8 μ.

** The h values in Fig. 5 were read at the middle of the boundary surface.

larger at the bottom than at the upper part of the tube. In reality things would be naturally more complicated, especially in this case because coagulation takes place at the same time.

§ 3. Sedimentation of Fine Emery Powder† (10)

Fine emery powder for optical shops, of nominal size # 1200, after being made uniform by decantation, was suspended in water, poured gently into vessels like the one shown in Fig. 9, and its sedimentation was observed.

The relations of the amount of descent of the boundary surface h *versus* t for various inclination angles are plotted in Fig. 10. Broken curves in the figure correspond to the calculated values after Nakamura's formula, putting $s = 2.5$ mm/min. (the observed descent velocity of the boundary when $\theta = 0^\circ$). The dependence of h-t curves on θ is quite different from what was seen in § 2, according to the difference in the shape of the vessel. The general tendency of the curves can be explained by Nakamura's theory; especially, the accordance of the observed and calculated initial velocities $(dh/dt)_{t=0}$ is excellent (Figs. 10 and 11), but the observed h values lead the calculated ones more and more as time goes on.

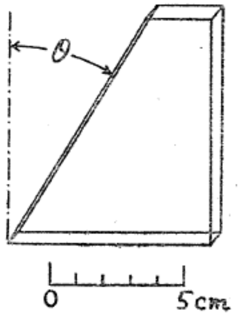

Fig. 9.

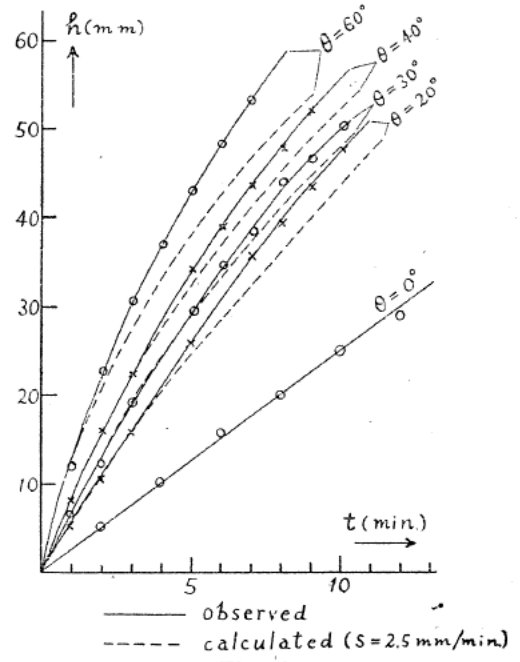

Fig. 10.

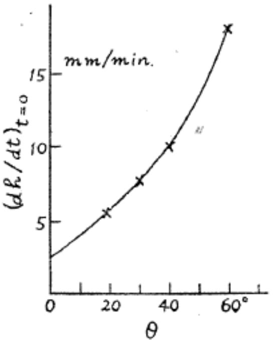

Fig. 11,

The change in the appearance of the boundary accompanying the advance of sedimentation is as follows: first, a thin wedge-type clarified layer appears just below the tilted wall, which proves the validity of Nakamu-

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† Taki and Isomura, and Tsukada assisted the author in these experiments.

(10) Similar odservations are reported in a recent issue of *Nature*, on the sedimentation of *potter's bone* in a conical flask: R. Johnson and E. Smith, Nature, **160** (1947), 27. However, it seems that they were ignorant of the relation with *Boycott's effect.*

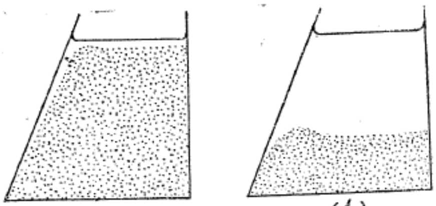
(a) (b)

Fig. 12.

ra's basic ideas (Fig. 12 (a)); convection takes place then to fill up this blank space, and the boundary descends gradually in a form with a protuberance near the tilted wall-side end as shown in Fig. 12 (b). Convection in this type of vessel is not very clear, but in a test-tube-type vessel the current is so violent that one can hardly execute any measurement.

The appearance of the wedge-type clarified layer, which shall be called the *V-layer* hereafter, signifies that the *levelling* occurs with a time-lag behind the sedimentation, which may be one of the causes of the discrepancy between the actual observation and Nakamura's theory. The *V-layer* was not distinctly recognized in the case of ammonium chloride smoke (presumably because the definition of the boundary was inferior), but we can point out several cases in which we could perceive it just under the upper tilted wall.

Most of the observations so far described were carried out in the summer vacation of 1947. The original purpose of our work, to establish the validity of the hypothesis that the *levelling action* in *Boycott's effect* is not due to diffusion but to a kind of convection current in the suspending medium, seems to have been accomplished by these experiments. But it is desirable to confirm this again in the case of erythrocyte sedimentation, because the fact that the sedimentation is much slower in the latter case may support the diffusion theory. Thus the following study was undertaken in the next summer.

§ 4. Sedimentation of Red Blood Corpuscles*.

0.25 to 30 per cent of rabbit's blood was dissolved either in the physiologic salt solution or in Hayem's solution (0.5 g of $HgCl_2$, 5.0 g of Na_2SO_4, and 1.0 g of NaCl dissolved in 210 cc of distilled water), and the sedimentation in small glass vessels like the ones used in the preceding section of this solution was observed. Approximate values of the descent velocity of the boundary surface for various inclination angles are given in the following table.

Table I. 2.5 per cent Blood Solution.

θ	0°	25°	40°	50°
dh/dt (mm/hr.)	4	6	7	8.5

As we expected considerable difficulty in detecting the convection current due to *levelling* in the case of blood, we tried a number of procedures including the *method of striae*; but what proved to be most satisfactory was tracing the movement of blood corpuscles through a microscope, by which means we succeeded in obtaining a rough estimate of the current velocity. An ordinary microscope (100 ×) was used. It was set horizontally, the vessel of blood solution being placed in the position of

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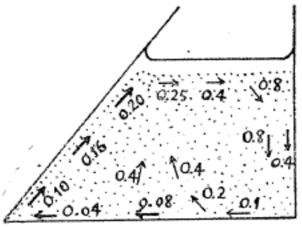
* Murase assisted me in these observations.

the object-glass. No special illumination was needed. The vessel was fixed on a carrier which could be displaced in both horizontal and vertical directions, so that we could perform an observation at any desired position.

A thin wedge-type clarified layer (*V-layer*) similar to the one mentioned in the preceding section appears again in this case. Microscopic observation reveals an upward movement of the blood corpuscles along the edge of this layer. The motion is quite orderly, the boundary with the *V-layer* never being invaded by the corpuscles.

A general idea of the movement of the corpuscles in the whole vessel may be obtained from Fig. 13, but care must be taken as to the interpretation of this illustration. The currents are unstable and are likely to be disturbed by illumination**, excepting the regular currents along the boundary of the *V-layer* and the bottom (esp. left half) of the vessel. The smaller the concentration is, the more unstable are the former currents, but we are obliged to use a dilute solution† for the purpose of observing the general situations as are illustrated in Fig. 13. Consequently, the directions and the velocities jotted in this figure are, except the ones represented by thick arrows, nothing more than an example to show the general trend. The currents represented by thick arrows are quite stable.



0.5 % of rabbit's. blood dissolved in Hayem's sol.

Fig. 13.

The numerals in Fig. 13 represent the current velocity (mm/sec.) read with ocular scale. They correspond to the values in a plane near the front wall. The velocities in the central plane are expected to be larger, as can be estimated from Fig. 14 which shows the influence of the viscous resistance near the wall, but actual measurement of them was not possible because of the limitations of the instrument.

The configuration of the *V-layer* and the velocity of the corpuscles along the lower edge of this layer vary with the inclination angle θ and the concentration of the solution c in the following way:

1°. When c is kept constant, both the thickness of the *V-layer* (or the opening of the wedge) and the current velocity increase with θ (Tab. II and Fig. 15).

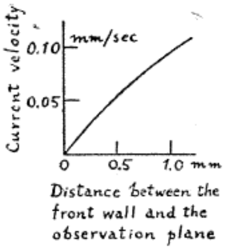


Fig. 14.

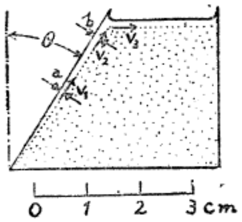


Fig. 15.

** Nakamura emphasizes the unreliability of microscopic observations in reference to these points; but our results are reproducible so far as what we positively maintain are concerned. They are also in accordance with the observations by the *methyl violet method*, which is to be described later.

† If the solution is concentrated, we are only able to observe the current along the *V-layer*.

Table II. 5 per cent Blood Solution.

θ	a (mm)	b (mm)	v_1 (mm/sec)	v_2 (mm/sec)	v_3 (mm/sec)
20°	0.13	0.20	0.08	0.08	0.07
40°	0.20	0.25	0.10	0.10	0.10
50°	0.30	0.40	0.17	0.17	0.11

2°. If θ is kept constant,

(a) the thickness of the *V-layer* decreases,

(b) the current velocity slightly increases, and

(c) the descent velocity of the boundary surface decreases

with increasing c; these changes, however, are confined to the range where c is not too large (Fig. 16).

An important remark should be added:

3°. Hardly any change can be recognized in the configuration of the *V-layer* and the movement of the corpuscles along the edge of the former during the period of the observation (1 to 1.5 hr.).

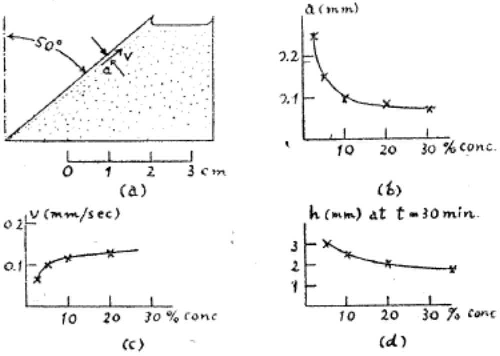


Fig. 16.

§ 5. Complemental Experiments and Observations*

This section will give an account of a number of complemental observations and experiments.

I. Currents in the blood solution caused by sedimentation described in the preceding section can be visualized by holding a small flake of methyl violet at an appropriate position in the vessel. A colored stream comes out slowly and shows an invisible current in the suspension. The general trend estimated by this very simple method agrees with what was illustrated in Fig. 13.

II. As a consequence of the convection in the suspension part, hydrodynamic flow takes place in the upper, already-clarified part of the liquid too. It is a very slow and not always stationary current, which can be detected by the *methyl violet method* or by means of the observation of red blood corpuscles remaining in the clarified region. (The concentration of the latter is of course very small.)

III. We supposed at the end of § 1, that in a vertical-wall vessel the particles should descend with a uniform velocity s retaining their relative positions. But the real state of things is much more complicated, and a weak convection is likely to occur even in this type of vessel. Unstable as this current is, one can detect it by the *methyl violet method.* The general tendency is sketched in Fig. 17.

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* Tsukada and Murase cooperated with the author in these experiments.

The current is assuredly caused by sedimentation, because if one fills the same vessel with water and tries the *methyl violet method*, nothing more than a straight downward motion of a colored stream can be seen.

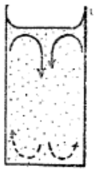

Fig. 17.

IV. If one places an obstacle on the surface of the tilted wall of the vessel used in the experiments of §§3 and 4, it may hinder the *levelling action* and lower the descent velocity of the boundary surface. This was suggested by Mr. Isao Osida of the Faculty of Engineering when our first report was published. The author adopted this idea as a possible way of detecting the convection due to sedimentation in the case of blood, before he started the experiments mentioned in the foregoing section.

We compared the sedimentation velocity in a vessel, on the tilted wall of which a numder of horizontal projections were attached (Fig. 18), with the one in the same vessel without projections.

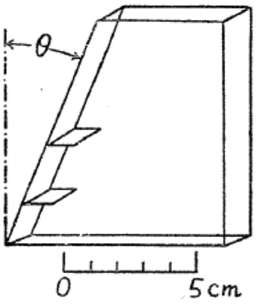

Fig. 18.

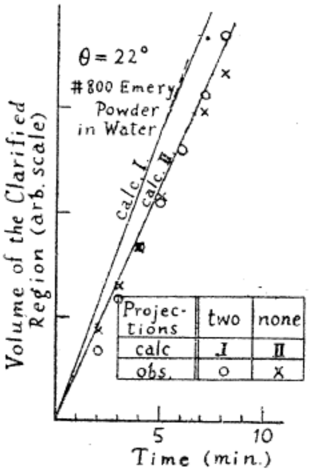

Fig. 19.

The emery powder suspension studied in §3 was observed at first. The results are illustrated in Fig. 19, in which the volume of the clarified region V is plotted *versus* t. Curve **I** in the figure was calculated according to Nakamura's theory, s being taken equal to 5.5 mm/min. (obs.), where the effect of the projections, *i.e.* that the lower surfaces of them should increase dV/dt as well as the increase in the area of the free surface, is taken into consideration. A knick in the upper part of the curve arises where the boundary surface passes the upper projection.

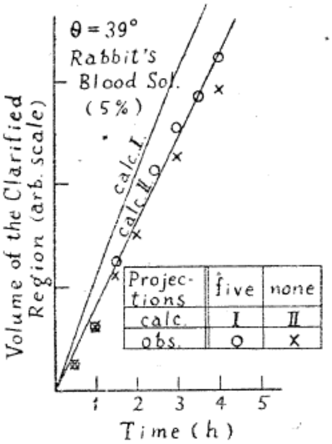

Fig. 20.

Fig. 19 shows that the lag of the observed value behind the calculated one is actually larger when the projections are attached. The lag, however, takes place mainly at the commencement of sedimentation, and the observed and calculated curves are almost parallel at $t \geq 2$ min.

The effect of projections on the tilted wall is much the same in the case of blood solution. No remarkable change can be seen if one increases the number of projections. Cf. Fig. 20 (rabbit's blood, five projections on the tilted wall).

Thus the hindering effect of projections was found to be small compared with what we had expected. The fact is that the current is too gentle for any considerable turbulence to be caused by passing the obstacles, as will be understood by Fig. 21 (microscopic observation). We do not mean, however, that there is no turbulence. A diffuse protuberance of the suspended phase into the clarified region in Fig. 22 must be attributed to the effect of turbulence.

Fig. 21.

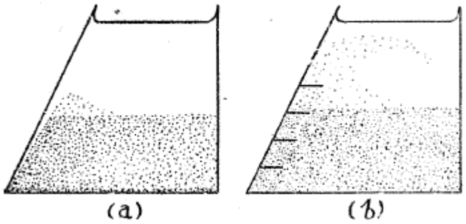

Fig. 22.

The above described slow and semi-stationary current may be of some interest for workers in the field of hydrodynamics.

V. Other than the diffuse protuberance of the suspended phase noticed in **IV**, detailed observation reveals that the blood corpuscles are scattered over the clarified region with but a small concentration (Cf. **III**). The apparently well-marked boundary surface of the suspended phase is, if one observes microscopically, of course of diffuse nature, and one can watch some blood corpuscles rising sporadically into the clarified region.

VI. Needless to say, particles are piled up on the bottom of the vessel in the form of a thin layer during the sedimentation, the layer thickening itself gradually.

Accounts in this section pertain mainly to the case of blood solution, but things are much the same in other suspensions.

§ 6. Conclusions and Discussions

A. *Boycott's effect* was observed on the sedimentation of ammonium chloride smoke (in air), fine emery powder (in water), and red blood corpuscles (in diluted blood solution); and it was established that the *levelling action* is not due to diffusion but to a mass motion (a kind of convection) of the suspending medium. We do not mean that diffusion has nothing to do with the sedimentation of these systems (Cf. §5, **V**), but its effect is very small, if any.

B. The velocity of this convection current is much greater than the descent velocity of the boundary surface of the suspended phase, which is usually regarded as the sedimentation velocity (Cf. Tab. I with Fig. 13 *et al.*). A general idea of the *levelling convection* could be obtained from Fig. 13.

C. Nakamura's theory for the velocity of descent of the boundary surface is qualitatively valid, but not quantitatively satisfactory in all cases.

According to the conclusion **A**, it is useless to discuss *Boycott's effect* on the basis of the equation of diffusion.

As was pointed out in **B**, the velocity of the *levelling convection* is some 10^2 times as large as the descent velocity of the boundary surface. Consequently, the author is inclined to picture the sedimentation in tilted vessels with a large horizontal vortex turned up in the suspended phase. The latter, whose upper boundary coincides with the boundary surface which we observe, flattens itself as time goes on, wherein the concentration of the particles in it is kept approximately constant by the excretion downwards of surplus particles (Fig. 23).

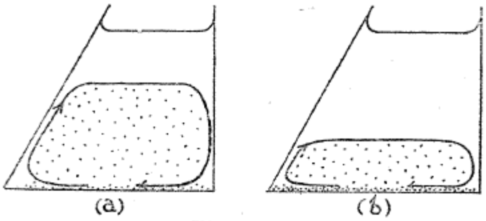

Fig. 23.

The crux is the mechanism of this vortex. It is not difficult to comprehend it qualitatively, as was mentioned before, by considering the uneven distribution caused by sedimentation of the pressure which the suspended system exerts on the medium, or, in other words, the non-uniformity of the density of the suspension (Cf. § 1). But if we go any farther to discuss the problem with hydrodynamical equations, we are beset with difficulties. If we wish an attack in the front, our discussions must ce based on Navier-Stokes' equations, as the viscosity of the medium plays an important rôle in this phenomenon. Writing down the fundamental equations is not essentially difficult —for instance, one can obtain the equations of motion and of continuity by regarding the forces acting on the particles, *i.e.* gravity, buoyancy, and Stokes' force due to the current in the medium, as body forces acting on the suspension**—, but solving them to arrive at a solution of practical value seems to be almost hopeless. The question remains in the future how to formulate and work out the problem.

The influence of diffusion was neglected in the above discussions. In reality, however, it does play a rôle, though of the second or third order. The author expects that a kind of convection in a vertical-wall vessel mentioned in §5, **III** could be reasoned if one could have carried out theoretical calculations on the above-stated principle with further corrections for this diffusion effect.

The phenomenon of sedimentation, which is ordinarily treated as a simple one-dimensional problem, reveals its rather complicated nature if one goes one step farther.

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