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Use of electrochemiluminescence in visualizing separated flows

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When a potential is applied between two electrodes immersed in a flowing chemiluminescent solution a blue glow appears at the surface of the anode. This glow may be used to observe the behaviour of liquid flows past solid surfaces. An electrochemical system suitable for generating the glow is described. Photographic studies of separated flows past cylinders, flat plates and cones are presented showing the application of the technique of electrochemiluminescence to flow visualization.

1. Introduction

The importance of flow-visualization techniques in studying the behaviour of complex flows has been long recognized. Consequently, through the years, many flow-visualization techniques have been developed including injection of marking fluids into the flow, neutral density beads, tufting, hydrogen bubbles, hot wires, and many others. Comprehensive summaries of these techniques may be found in the symposium volume entitled *Flow Visualization* (1960), and in the paper by Schraub *et al.* (1964), where it was pointed out that most of these methods require the introduction of foreign objects into the flow and are difficult to interpret in unsteady flow. In this paper a technique is presented which offers the possibility of observing the behaviour of liquid flows past solid surfaces continuously without disturbing the flow. This technique is based on the electrochemiluminescence (ECL) process.

When two electrodes are immersed in the flow field of a chemiluminescent solution and a voltage is applied between them, a blue glow appears at the surface of the anode. This phenomenon, while noted previously (Harvey 1940), was recognized only recently as a possible means for flow visualization (Howland, Pitts & Gesteland 1962). Springer (1964) demonstrated that in laminar flow, for a given solution and for a given applied potential, the intensity of the glow depends on the mass transfer of the active electrolyte to the surface of the anode. Howland *et al.* (1962) showed that the glow is generated within a few wavelengths of the anode, i.e. at a distance which is much smaller than the thickness of the boundary layer. Thus the technique of electrochemiluminescence is particularly useful for studying those phenomena that occur at the wall. Also, since the glow covers the entire surface, the technique may be used to study both local and gross effects. In the following an electrochemical system suitable for the experiments is described and photographic studies of separated flows are presented demonstrating the application of the ECL technique to flow visualization.

2. Electrochemical system

The two major variables that affect the glow are the composition of the chemiluminescent solution and the applied voltage. A solution suitable for the ECL experiments has been described by Howland *et al.* (1962) and is shown in table 1. This solution was found to give nearly the highest contrast and maximum brightness without significant accumulation of insoluble reaction products and without visible bubbling at the anode. A brief description of the purpose of each component is given in table 1.

Substance	Amount	Remarks
(1) H_2O	—	Solvent, should be distilled
(2) KCl	1 N	Supporting electrolyte
(3) KOH	0·01 N	Adjusts pH; Luminol exhibits chemiluminescence in alkaline solution only
(4) H_2O_2	$8{\cdot}5\times10^{-4}$ N	Active electrolyte; concentration strongly affects light intensity
(5) Luminol†	$4{\cdot}4\times10^{-3}$ N (150 mg/l.)	Chemiluminescent substance, soluble in alkaline solutions only
(6) EDTA‡	Very small	Ties up traces of iron and copper; only very small amount should be added

† Eastman Kodak Company Luminol = 5-amino-2,3-dihydrol-1,4-phthalazinedione.
‡ Eastman Kodak Company EDTA = (ethylenedinitrite)tetraacetic acid.

Table 1. Composition of chemiluminescent solution

It is important that all the chemicals, including the water, be free of impurities. Traces of iron or copper will catalyse the chemiluminescent reaction, resulting in an objectionable bulk glow of the solution. For this reason the water used in the experiments was either distilled or, when used in larger quantities, purified by ion exchangers. A slight bulk glow of the solution can be eliminated by the addition of a very small quantity of EDTA, a chelating agent that ties up small traces of copper and iron.

Satisfactory glow was also obtained using methanol, acetone and mixtures of water and glycerine as solvents. In the experiments that follow, the chemicals were proportioned exactly as given in table 1. It was found that small changes of the specified concentrations alter the intensity and contrast of the glow but have little effect on the appearance of the glow pattern. This solution was also found to be insensitive to simultaneous reduction of the concentration of all the reactants. The composition of the solution changes with time, evidenced by decreased light output and discoloration. Solutions were used successfully for periods of up to 7 days without anything being added. During this period sufficient light was developed to allow photographing of the glow pattern. The intensity of the glow could be increased temporarily by adding small amounts of hydrogen peroxide to the solution.

A schematic diagram of the circuit driving the electrochemical reaction is shown in figure 1. The power was supplied by a variable-voltage, low-resistance d.c. power supply. There were three electrodes placed in the solution: an anode, a cathode, and a saturated KCl–calomel electrode.

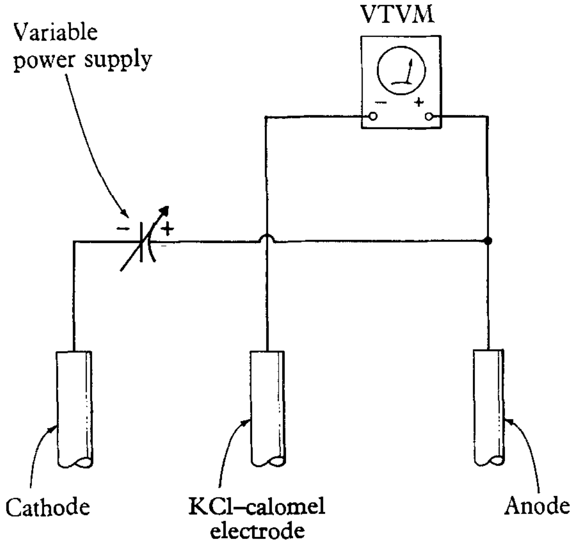

FIGURE 1. Circuit for driving electrochemical reaction and for measuring the anode-to-solution potential.

The glow is produced at the anode. Therefore, it is necessary to make the anode in the shape of the model about which the glow is to be studied. Good glow was obtained with platinum or platinum-plated anodes. None of the other materials that were tried as anodes (stainless steel, aluminium, copper, silver) gave satisfactory results. Before operation the anode was cleaned with a solution of KOH in methanol. If the pattern of glow at the anode still showed irregularities caused by surface effects, then the anode was further cleaned by raising the voltage for a few seconds with the fluid flowing past it. This process was repeated until the glow passed into the solution and the bubbles were eliminated. It is noted here that prolonged cleaning is harmful to both the electrodes and the solution and should be avoided.

The cathode can be of any shape, but it should have about the same surface area as the anode in order to prevent bubbling of the solution. The cathode may be located anywhere in the flow but, as expected, best results were obtained when it was close to the anode. The cathode may be made of a conducting material other than platinum. In the present experiments both platinum and aluminium cathodes were used.

A saturated KCl–calomel electrode was used to measure the potential between the anode and the solution. This electrode was placed near the anode at a position where it did not disturb the flow. It was found that at potentials less than about 0·2 V the glow disappeared. If the potential was raised above 1·2–1·5 V the glow left the surface of the anode and illuminated the flow in the wake of the anode. In each experiment, the potential was adjusted within the 0·2–1·5 V range to give the maximum contrast of the glow.

3. Experimental apparatus

Two types of experimental apparatus were used in the experiments: a small rotating flow chamber, and a large blowdown-type water tunnel.

A schematic diagram of the rotating-flow-chamber apparatus is shown in figure 2. The flow was provided by a circular flow chamber driven by a variable-speed d.c. motor. The flow chamber was constructed from a 10 in. diameter Plexiglas cylinder. Concentric with the wall of the chamber was a perforated 5 in. diameter Plexiglas cylinder. The purpose of this cylinder was to minimize the disturbances in the flow near the anode (model). As will be shown in the next section, this apparatus was useful for demonstrating a number of interesting ECL phenomena. It was limited to maximum flow velocities of about 2·0 ft./sec, because of the centrifugal effects on the fluid.

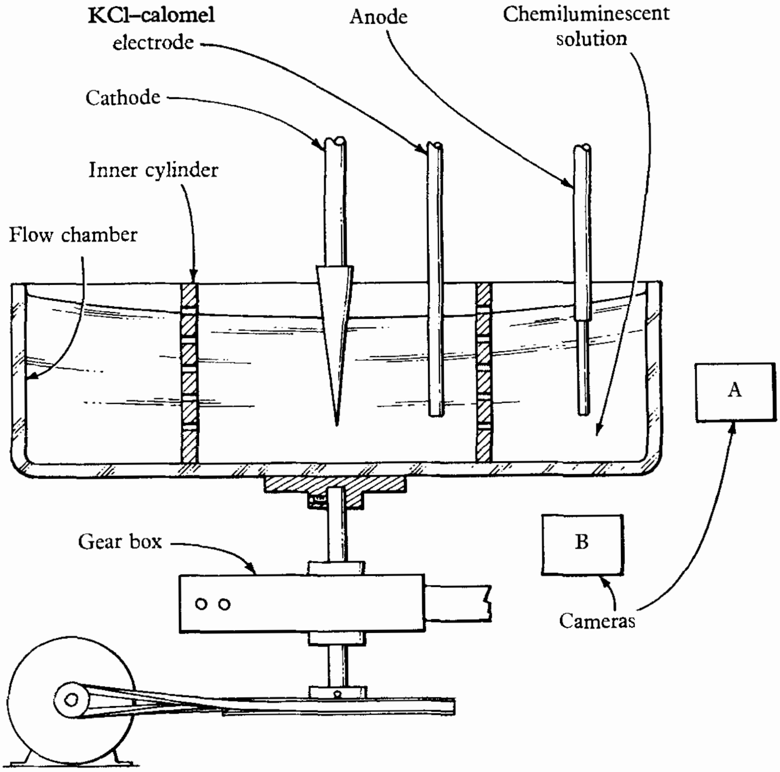

FIGURE 2. Schematic diagram of rotating-flow-chamber apparatus.

To attain higher fluid velocities, experiments were also performed in a blowdown-type water tunnel. A schematic diagram of the tunnel is shown in figure 3.

Details of the apparatus are given by Schiller (1964) and by Adams & Hill (1965). All those parts of the apparatus which were in contact with the fluid were made of materials resistant to the chemicals used. The model was mounted in the test section, which was a 36 in. long Plexiglas tube with an inside diameter of 4 in. (figure 4). In order to determine the fluid velocities and the velocity profiles in the test section, measurements were made by dynamic and static pressure taps located near the inlet and outlet planes of the test tube (figures 3, 4). Figure 5 shows typical velocity profiles obtained with the settling tank half full (approx. 750 litres) of solution at the start of the run. The change in velocity with time elapsed from the opening of the control valve is also shown in this figure. All actual test runs were made with the settling tank half full. In this case, the maximum attainable fluid velocity in the test section was approximately 10·5 ft./sec.

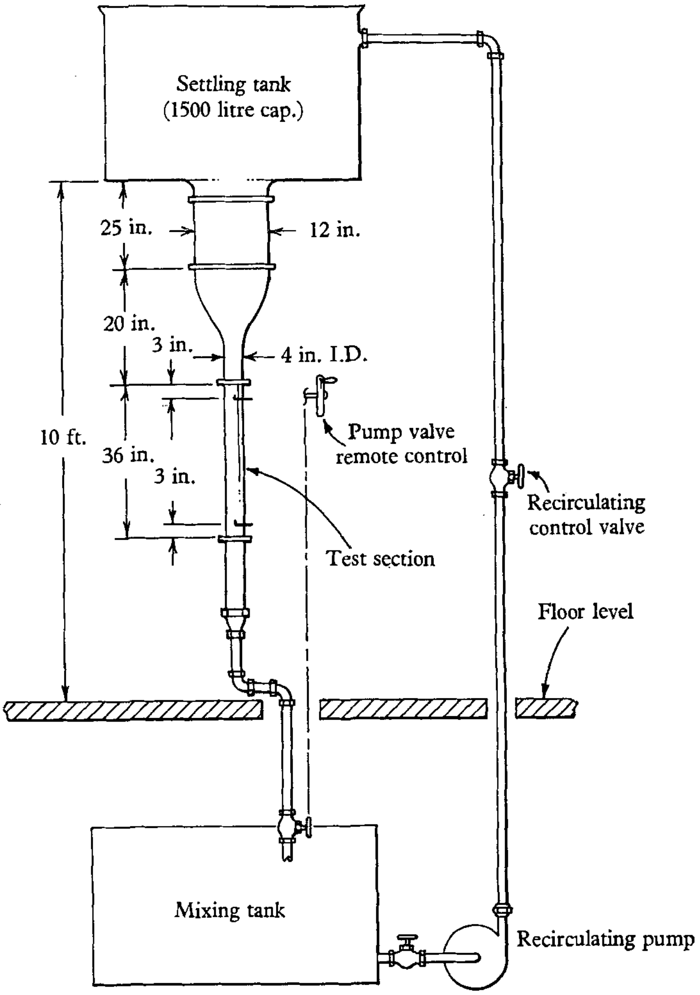

FIGURE 3. Schematic diagram of water tunnel.

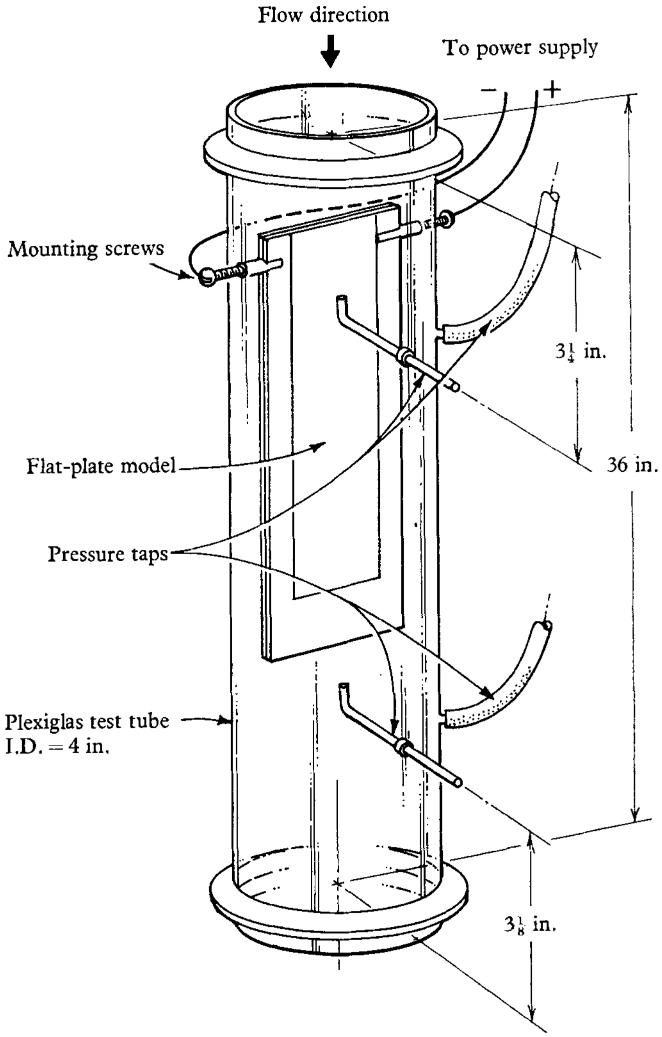

FIGURE 4. Test section used in the water tunnel.

4. Flow visualization

Experimental evidence was given by Howland *et al.* (1962) indicating that the ECL glow is generated within a few wavelengths of the surface; i.e. the distance from the surface at which the glow is generated is much smaller than the thickness of the boundary layer. For this reason, it was decided to perform preliminary studies on the use of the ECL technique of flow visualization in separated flows.

Figure 6 (plate 1) shows the flow past a cylinder immersed in the solution in the rotating flow chamber, at a Reynolds number of 6000† (based on the diameter of the cylinder, $d = 0{\cdot}315$ in.). The flow is from left to right and the view is normal to both the axis of the cylinder and the direction of the flow. Some ambient

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† In calculating Reynolds numbers the kinematic viscosity of water was used.

light was provided to show the surface of the fluid. This figure indicates the line of separation, which appears as a vertical dark line. Separation lines were also observed on cylinders at higher Reynolds numbers. Figure 7 (plate 2) shows the separation line on a cylinder at a Reynolds number of 130,000 (based on the cylinder diameter = 1·5 in.). The photograph in figure 7 (plate 2) was taken in the water tunnel. The line of separation at the end of the cylinder is affected by the wall of the test section. In this experiment approximately 750 litres of solution were used, illustrating that the ECL technique may be applied with large quantities of solution.

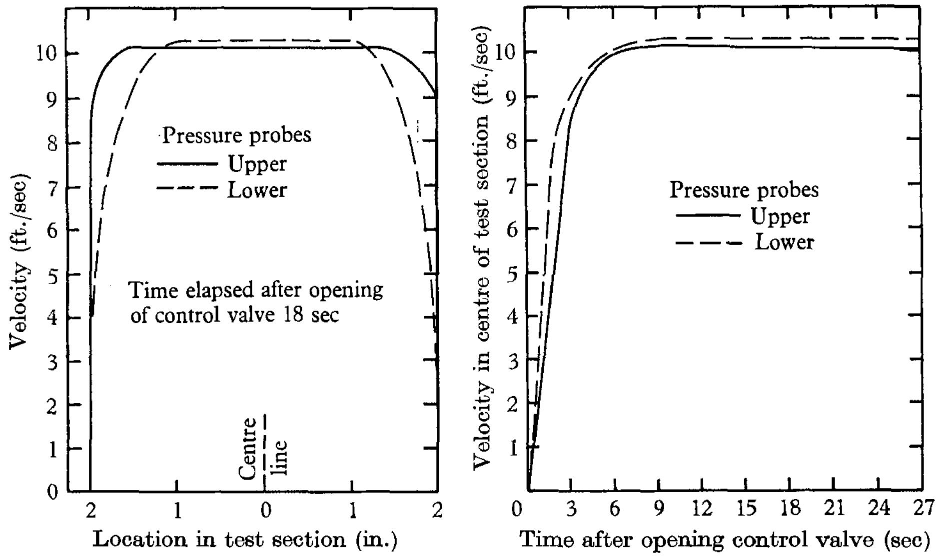

FIGURE 5. Velocity profiles and velocity–time characteristics in test section. Settling tank half full (~ 750 litres).

Using the rotating-flow-chamber apparatus, photographs were taken of transverse flows past cones at various Reynolds numbers (figure 8 (plate 2)). The flow is again from left to right and the pictures were taken from a direction approximately normal to the direction of the flow. The dark lines indicating separation are clearly defined. These pictures illustrate the changes in the positions of the separation with varying Reynolds numbers. The changes in the positions of the separation with increasing Reynolds numbers show trends similar to those quoted by Schlichting (1960) and by Grove, Shair, Petersen & Acrivos (1964) for circular cylinders.

In figure 9 (plate 3), flow past a flat plate is shown at two different angles of attack at a Reynolds number of 128,000 (based on the plate length of 5 in.). The separated regions behind the leading edge of the plate shown by these pictures are very similar, at least qualitatively, to the pictures presented by Eichelbrenner (1964). He photographed separated regions on annular wings using an ointment method (mixture of petrol and lampblack). The pictures in figure 9 were obtained in the water tunnel. Photographs of flows past a flat plate at lower Reynolds numbers (Re = 10,000, based on the plate length of 1 in.) show a very similar glow pattern (figure 10 (plate 3)). The pictures in figure 10 were taken in the rotating-flow-chamber apparatus.

In the foregoing pictures the voltage was adjusted so that the glow was generated near the surface of the anode. As was mentioned previously, when the applied voltage is increased the glowing substance leaves the surface and illuminates the fluid behind the body. This glow is quite bright and decays only gradually with time and distance. This effect is illustrated in figure 11 (plate 4), where the glow behind the cylinder is shown. Unfortunately, no practical application for this effect could be found, since these lines could not be identified to be either streamlines, streaklines, or pathlines.

The above examples were given to indicate the applicability of the ECL technique to flow visualization. It is expected that further investigation will reveal additional uses of this technique.

We wish to acknowledge the important contribution of W. H. Pitts, whose investigation of the ECL phenomenon led us directly to the experiments described here. We wish to thank F. G. Adams, R. C. Gesteland, M. Potash, and T. R. Schiller, who, in particular, helped in the experiments and the interpretation of the results. We are grateful to Prof. A. H. Shapiro for his suggestions and for his interest in this work. Some of the experiments described here were conducted at the Research Laboratory of Electronics, M.I.T., an inter-departmental laboratory supported by the U.S. Army, U.S. Navy and the U.S. Air Force. This project was also supported in part by grants to the Mechanical Engineering Department from the Shell Research Foundation and from the U.S. Navy and U.S. Coast Guard.

REFERENCES

ADAMS, F. G. & HILL, M. G. 1965 A study of the technique of electrochemiluminescence in determining points of separation and transition. S.M. Thesis, M.I.T.

EICHELBRENNER, E. A. 1964 Separation, reattachment and stall in three-dimensional laminar and turbulent boundary layers. *ASME Symp. on Fully Separated Flows* (ed. A. G. Hansen), pp. 1–9.

GROVE, A. S., SHAIR, F. H., PETERSEN, E. E. & ACRIVOS, A. 1964 An experimental investigation of the steady separated flow past a circular cylinder. *J. Fluid Mech.* **19**, 60–80.

Flow Visualization Symposium 1960 (Bound volumes available from *ASME*, 345 East 47th Street, New York, New York 10017.)

HARVEY, E. N. 1940 *Living Light*. Princeton, New Jersey: Princeton University Press.

HOWLAND, B., PITTS, W. H. & GESTELAND, R. C. 1962. Use of electrochemiluminescence for visualizing fields of flow. *Tech. Rep.* 404, Research Laboratory of Electronics, M.I.T.

SCHILLER, T. R. 1964 An apparatus for applying the technique of electrochemiluminescence to boundary layer studies. S.M. Thesis, M.I.T.

SCHLICHTING, H. 1960 *Boundary Layer Theory*, 4th edn., p. 155. New York: McGraw Hill.

SCHRAUB, F. A., KLINE, S. J., HENRY, J., RUNSTADLER, P. W. & LITTELL, A. 1964 Use of hydrogen bubbles for quantitative determination of time-dependent velocity fields in low-speed water flows. *ASME Paper* no. 64-WA/FE-20.

SPRINGER, G. S. 1964 Use of electrochemiluminescence in the measurement of mass transfer rates. *Rev. Scient. Instr.* **35**, 1277–80.

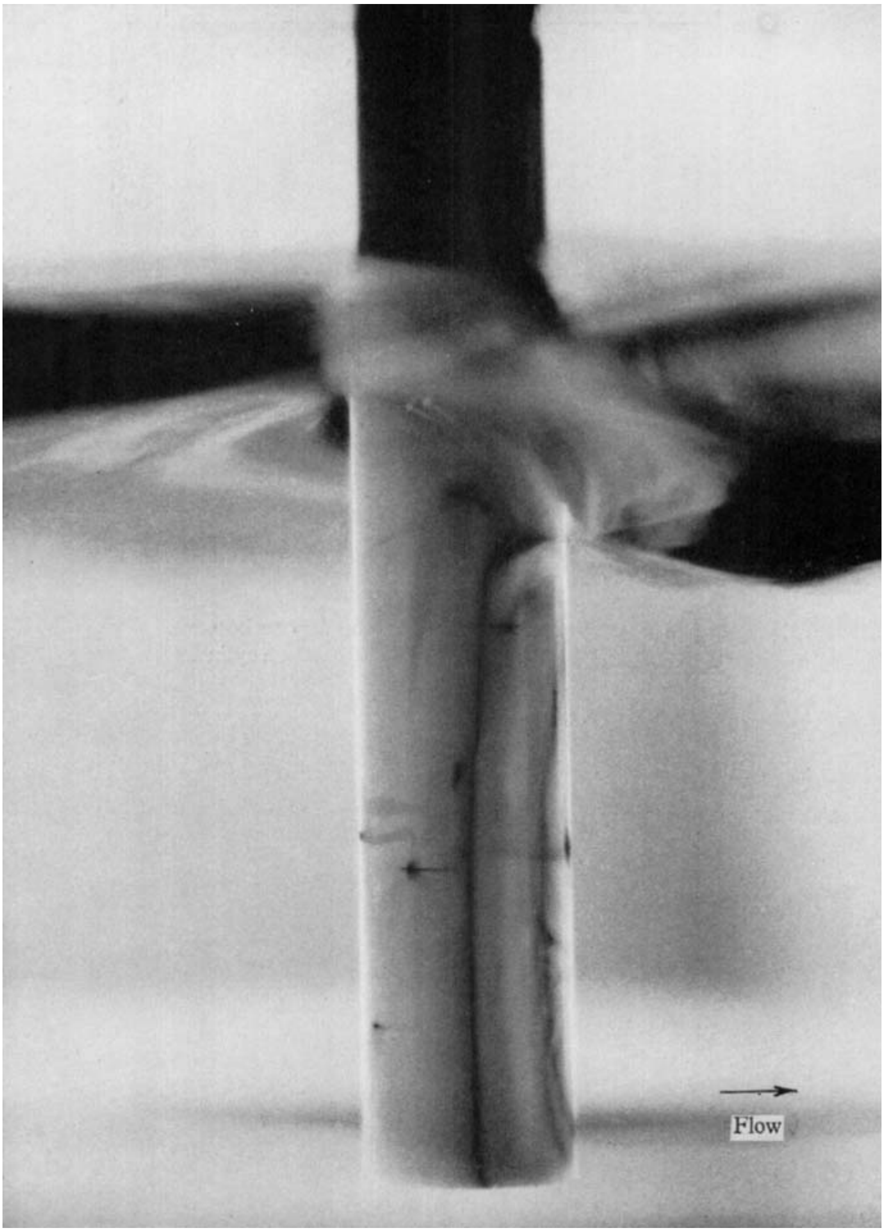

Figure 6. Flow past a circular cylinder. Flow from left to right. Dark line showing position of separation. Reynolds number = 6000 (based on cylinder diameter = 0·315 in.).

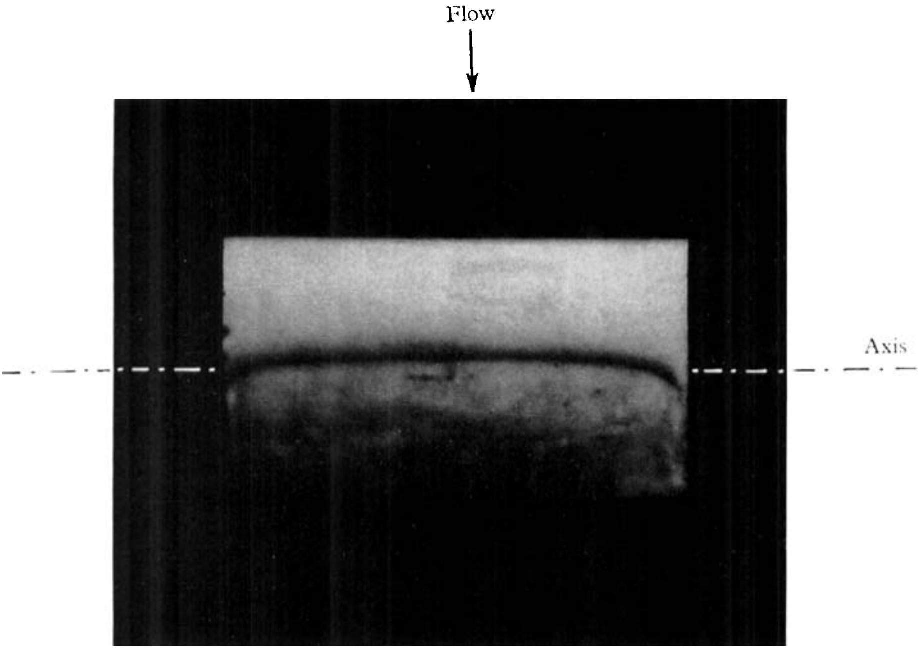

FIGURE 7. Flow past a circular cylinder. Flow from top to bottom. Dark line showing position of separation. Reynolds number = 130,000 (based on cylinder diameter = 1·5 in.).

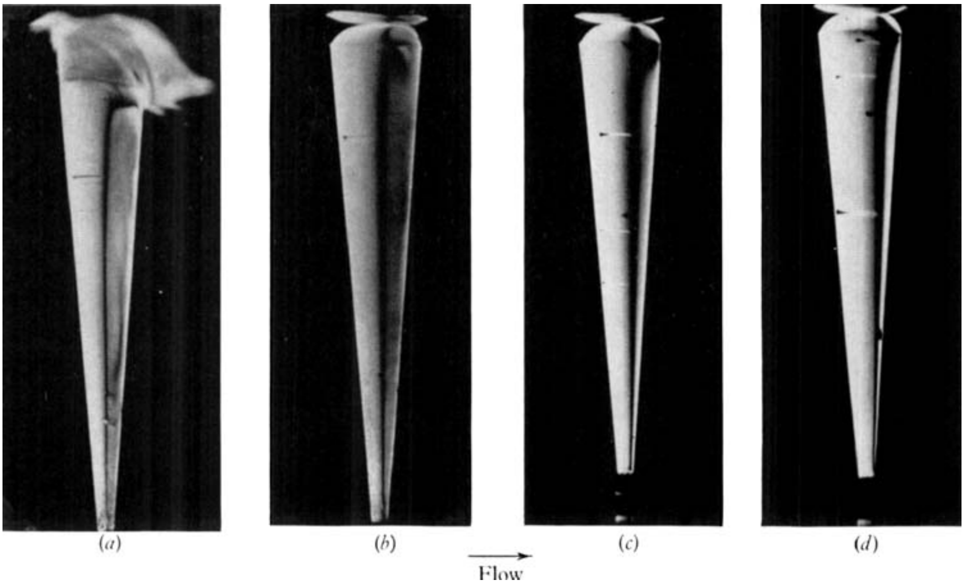
(*a*) (*b*) (*c*) (*d*)

FIGURE 8. Flow past a cone. Flow from left to right. Dark lines showing positions of separation at various Reynolds numbers. Reynolds numbers (based on maximum cone diameter = 0·5 in.): (*a*) Re = 4800; (*b*) Re = 1200; (*c*) Re = 200; (*d*) Re = 60.

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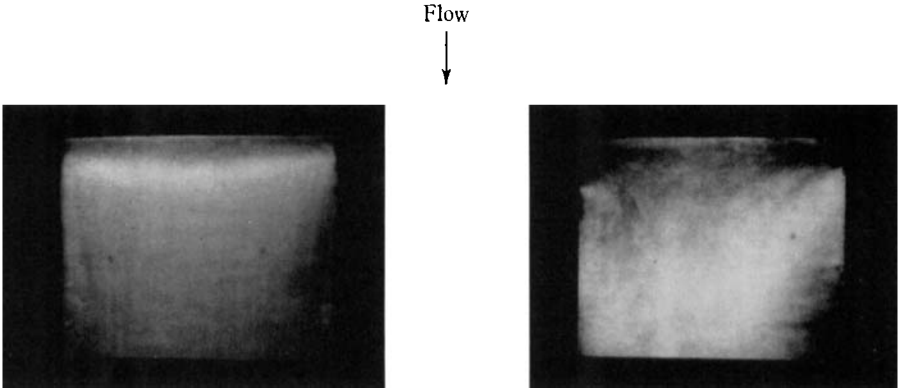

FIGURE 9. Flow past a flat plate. Incident flow in plane of paper from top to bottom. Glow patterns showing separated regions. Angles of attack are: (*a*) 3°; (*b*) 13°. Reynolds number = 128,000 (based on plate length = 5 in.).

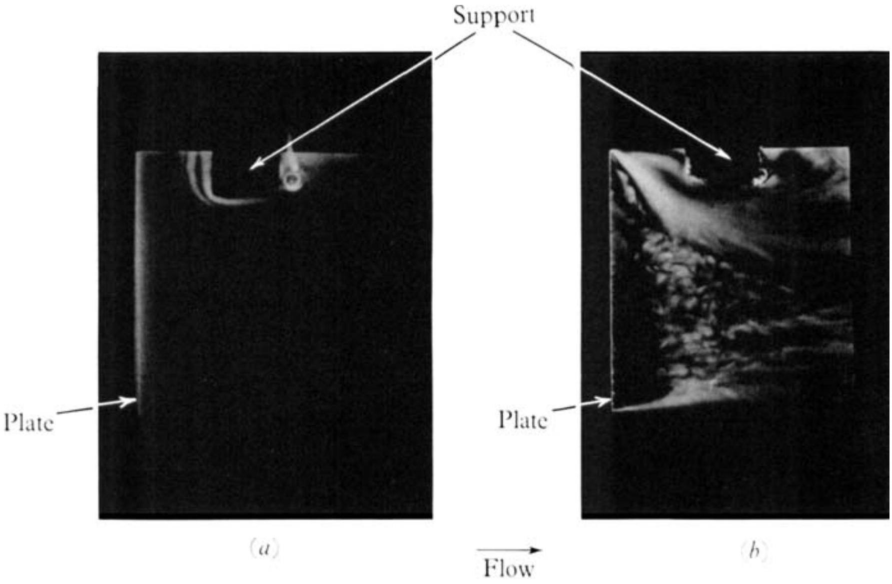

FIGURE 10. Flow past a flat plate. Incident flow in plane of paper from left to right. Angles of attack are: (*a*) 0°; (*b*) 10°. Reynolds number = 10,000 (based on plate length = 1 in.). Dark obstacle at top is plate support.

FIGURE 11. Illumination of the wake behind a circular cylinder. Flow is from left to right.