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A. ELECTROCHEMICAL METHODS IN HYDRODYNAMIC EXPERIMENTS

In the course of trying to develop an electrochemical model for olfactory stimulation, we have happened upon a number of electrochemical effects associated with fluid motion past electrodes. Certain of these were remarkable enough that, instead of taking measures to eliminate them, as is usually done in electrochemistry, we thought it worth while to try using these effects to make the motion visible or to measure fields of flow in experimental hydrodynamics. We have a number of such methods, of which the following seem to hold promise.

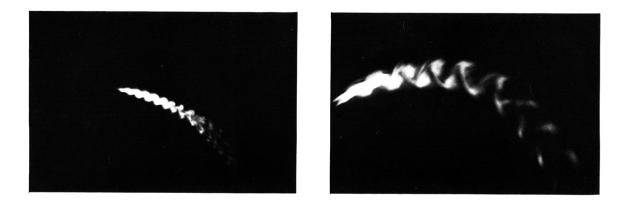
1. Electrochemiluminescence

In Quarterly Progress Reports Nos. 57 and 58 we showed that the bright glow produced at a platinum anode hydrofoil immersed in an aqueous alkaline solution of luminol (5-amino-2, 3-dihydro-1, 4-phthalazinedione, which is more likely to exist as 5-amino-3, 10-dihydroxy phthalazine) and H_2O_2 afforded a clear, detailed picture of the turbulent boundary layer about the hydrofoil. During the past few months we extended the scope and usefulness of the methods considerably.

In the first place, the phenomenon occurs in methanol as well as in water, provided that tetramethylammonium hydroxide in appropriate amounts is used as a base in place of NaOH, that $N(CH_3)_4Cl$ instead of KCl is used to provide conductivity, and that a solution of H_2O_2 in methanol is used (made from 30 per cent H_2O_2 in H_2O , thus introducing a small amount of water). This solution exhibits approximately the same pH as the purely aqueous one, that is, approximately 11.5. This measurement is made by diluting the methanolic solution with water.

However, if the methanolic solution is now gradually acidified with glacial acetic acid, novel effects appear. The glow from the turbulent boundary layer immediately next to the electrode dims rapidly and disappears, but the fluid wake behind the electrode begins to glow brightly. As more acid is added, an increasingly long section of the trailing fluid becomes luminous. When approximately 20-40 cm of it are visible, it has the maximum brightness per unit area; when the solution is made still more acid, the visible part of the trail continues to lengthen, up to a total distance of 3-4 meters

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(b)

Fig. XVI-1. Von Kármán vortex street resulting from circular flow past a cylindrical platinum anode. (a) Reynolds number, 90; (b) Reynolds number, 100.

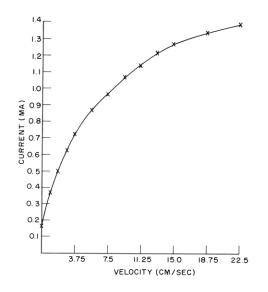


Fig. XVI-2. Values of electrode current at a constant potential for several rates of steady flow. The electrode is a platinum wire, 1-cm long and 0.5 mm in diameter, which is immersed in 1 N aqueous solution of KCl with 0.015 per cent H_2O_2 added.

at least, but the brightness decreases. Even at the acid extreme, the trail is still visible, and the solution (which is of course well buffered) shows, after dilution with water, a pH of approximately 5.3. This luminous wake can be seen to consist of fluid that has passed through the boundary layer immediately next to the anodal electrode itself. Figures XVI-1a and XVI-1b, which were obtained with a moderately acid solution, show a classical von Kármán vortex street, with the lines of flow from the boundary layer of a smooth platinum cylinder passing between the alternate members of the double row of vortices. The separation of the two rows is probably caused by the centrifugal currents set up in our system. The hydrofoil is placed in a rotating bath in order to avoid the complexities of constructing a linear flow chamber. This is arranged in such a way that both the hydrofoil and the bath can be rotated at different speeds around a common center; with this arrangement the trail can be held fixed with respect to the camera.

The series of changes that occur with increasing acidity is quite reversible. If $N(CH_3)_4OH$ is added gradually to the acidified solution, the luminous trails become shorter until, finally, the turbulent boundary layer at the electrode is again the only thing that glows. The shortening and lengthening process can be repeated any number of times.

It may be useful to include a recipe for making a solution that shows all of these effects, although the quantities given do not necessarily produce maximum brightness or the fastest pulse response: To a methanolic solution of 1/2 N tetramethylammonium chloride, add 100 mg/liter of luminol and 10 ml/liter of 3 per cent hydrogen peroxide in methanol (the 3 per cent solution is prepared from 30 per cent H₂O₂ in H₂O, diluted with methanol).

If 250 ml of this solution are used and $N(CH_3)_4$ OH, which is 1 N in methanol, is added slowly, we find the effects shown in Table XVI-1. (The length of the visible trail is estimated crudely by eye, for it fades out gradually.)

The solution now has a pH of approximately 11.5. When we added 1 N acetic acid in methanol, the results were those shown in Table XVI-2.

It should be evident that this method of visualizing flows past solid objects has particular advantages. The subsequent history of the fluid originating in the whole boundary layer or any part of it can be traced as far as desired by painting part of the electrode without disturbing the flow in the boundary layer in any way. The electrode can be made in any shape and inclined at any angle to the stream. The current can be supplied to the electrode in pulses instead of continuously. The electrochemical reaction is sufficiently rapid that pulse rates as high as several hundred per second (or even several thousand per second under some conditions) will produce discrete bursts of luminous trail. In this way, the motion and the velocities in the wake can be measured for rapidly changing unsteady flow as easily as for steady flow.

Various changes in the technique are possible, and we are engaged in testing some

Table XVI-1.

Amount of base added (ml)	Length of trails (cm)
0.33	15
1	10
2	6
4	4 (with slight glow at surface)
5.25	No trail; bright turbulent boundary layer crawling over electrode.

Table XVI-2.

Amount of acid added (ml)	Length of trails (cm)
1.3	1.5 (surface glow gone)
4	3.5
6.6	10
8	15
10	21
12.5	28 \rangle trail very bright
14	30
15.5	56
17	91
18.5	140 \rangle intensity diminishing
22.4	245
26.3	300-400

of them. The system can be used with other organic solvents, particularly those that have kinematic viscosities widely different from water or methanol. Such solvents have Reynolds numbers (and therefore equivalent free-stream velocities) that are very different from that for water for the same actual system velocity. A decrease in the effective Reynolds number can be obtained easily. The luminol-in-water system described in Quarterly Progress Report No. 57 can be prepared by using almost any ratio of water to glycerol. But some solvents should cause a substantial change in the reverse direction. Thus, liquid sulfur dioxide would yield a Reynolds number 3.34 times larger than an aqueous solution moving at the same speed, and liquid ammonia would multiply the Reynolds number by 3.03. It seems likely that the reaction should function in such solvents (possibly with minor chemical changes). The system operates quite well at -50°C in methanol. Sulfur dioxide and ammonia would not be difficult to handle in a linear flow system, since they are both standard refrigerants. Liquid hydrogen cyanide also has a most favorable viscosity factor. Of the ordinary solvents, the most promising seem to be diethyl ether, which increases the Reynolds number by a factor of 2.7 over that for H₂O, and acetone, which increases the Reynolds number by a factor of 2.4. Others can be found, but the solvents mentioned also satisfy the indispensable condition of being fairly good ionizing solvents for some salts and therefore provide the necessary conductivity. For a given Reynolds number, these solvents allow a longer photographic exposure because of lower hydrofoil velocities.

We have not yet tested the usefulness of the second famous chemiluminescent substance, lucigenin (10, 10'-dimethyl-5, 5'-bisacridinium dinitrate). When its luminescence is produced in the bulk of a solution by a chemical catalyst, the light intensity is said to be 100 times that of luminol, or more. In the literature on this subject, it has been stated (1) that the luminescence of lucigenin also can be produced electrically, but at the cathode. It is conceivable that lucigenin may provide enough additional light to reduce the sensitivity required by our photographic apparatus. This apparatus has been improved since the photographs of Fig. XVI-1 were taken; we can now photograph the structure of the flow in much greater detail. We shall try lucigenin as soon as we synthesize a reasonable supply of it. (Luminol is available commercially, but lucigenin is not; directions for making it can be found in Albert's treatise on the acridines (2).)

We have not yet made a systematic analysis of the mechanism underlying the chemiluminescence of luminol, but we have made a few incidental observations which do not seem to have been noted previously. In most of the studies of aqueous solutions containing luminol and H_2O_2 , the solutions were found to be strongly fluorescent below a certain critical pH (approximately 8 or 8.5). It was found impossible to evoke the luminescence by any catalyst whatever below this pH. Above this pH, the fluorescence vanishes, and a chemical catalyst (such as NaClO, $K_4Fe(CN)_6$, and iron and copper compounds) evokes luminescence which becomes gradually brighter and attains a

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maximum at pH 11.5. This fact has caused much difficulty in comparing the fluorescence and luminescence spectra – a comparison that is desirable for theoretical purposes. In our slightly alkaline and acid solutions, a chemical catalyst must evidently be generated at the electrode and pass into the luminous trail. This catalyst must differ from those used previously with luminol, since it is effective in neutral or even acid solutions of a pH as low as 5.3. This catalyst is not dissolved in Cl₂. It might be methyl hypochlorite, since hypochlorite esters are supposed to be the first product of the chlorination of alcohols. It seems volatile, because if the pH is adjusted to produce the brightest trail, we sometimes find on top of the solution a heavy vapor that causes a bright glow when it is blown back on the solution surface.

2. Measurement of the Velocity of Flow

The rate of flow of a fluid can be measured by determining the polarographic current resulting from the reduction of dissolved oxygen at a platinum cathode that is maintained at the correct potential; the faster the flow, the quicker the renewal of the depleted oxygen in the diffusion layer around the electrode, and the greater the current. The method has not been very popular, largely owing to its limited sensitivity.

It is well known that the presence of H_2O_2 produces a great increase in the rate of reduction of oxygen at such an electrode (the catalytic current). We hoped that the addition of H₂O₂ to a moving electrolyte containing dissolved O₂ would substantially augment the change in electrode current with velocity. S. J. Wiesner found at once that it did. Figure XVI-2 shows the current as a function of the velocity of the fluid. We believe that this increase in sensitivity is sufficient to render the method very practical for measuring speeds in fields of flow. We have also found that the electrode current will follow rapid periodic fluctuations in fluid velocity of 100 cps or more. This method, therefore, seems suitable for measuring large accelerations. The aqueous solution was 1 N in KCl and contained 0.015 per cent H_2O_2 . Wiesner also found that the less sensitive method mentioned above (that is, the solution without H_2O_2) could be modified somewhat to determine the direction of the streamlines as well as the speed at a single point along them. His preliminary trials were encouraging. He put one electrode upstream and forced enough current through it to deplete locally the surrounding electrolyte of dissolved oxygen and then used a second electrode downstream as a probe to measure the concentration of dissolved oxygen in the ordinary polarographic way. When this electrode was placed in the wake of the first there was a sharp drop in current; in this way the wake could be traced a considerable distance downstream. If the second electrode were placed immediately behind the first electrode, it would, of course, give the local direction of the streamline. The current through the first electrode would determine the speed. Presumably the sensitivity of this method would be much greater if H_2O_2 were added.

Because Wiesner's measurements were successful when the pH of the solution was approximately 8.3, it seems likely that this method could be adapted to the measurement of ocean currents as well as fluid flow in other natural circulatory systems. Although the experimental conditions will have to be changed, a useful method should not be difficult to realize, since the polarographic reduction of oxygen is a reaction that is known to be dependable in all kinds of fluids – even biological tissue.

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R. C. Gesteland, B. Howland, J. Y. Lettvin, W. H. Pitts, S. J. Wiesner

References

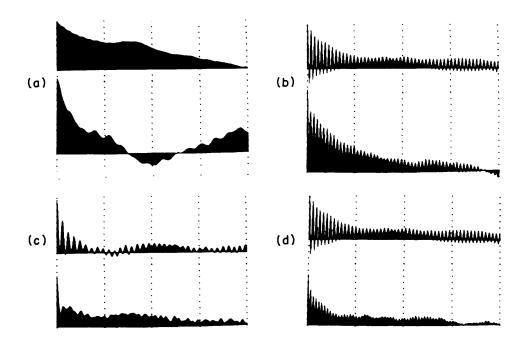
1. V. Vojiir, Chemiluminescence surrounding the dropping Hg electrode, Collection Czechoslov. Chem. Commun. 19, 862-872 (1954); in Russian.

2. A. Albert, The acridines; their preparation, physical, chemical, and biological properties and uses (E. Arnold and Co., London, 1951).

B. SERVOANALYSIS OF POSTURAL REFLEXES

This project was undertaken to determine the characteristics and approximate linear model of the peripheral self-regulating servomechanism through which voluntary movement and the maintenance of limb posture are achieved. Since the "input" to the system can be neither adequately localized nor driven, the only valid sets of measurements that can be made are the system output impedance when the internal sources are "fixed" and the noise output under the same conditions. Apparatus was devised to perform the measurements on the wrist joint in a state of flection. The subject was asked to keep his hand lined up with a pointer while his hand was being perturbed mechanically with filtered gaussian noise from a low-impedance (force source) driving mechanism. The torque and angle signals were recorded on two-track tape for subsequent data processing. In performing the required task the subject was exerting an "intention clamp" upon the environment, with the result that his internal energy sources could be assumed to be as fixed as possible. Output noise measurements were made under the same conditions, but with the hand splint swinging free; the small angular deflections were recorded.

It is theoretically possible to derive a system function from input and output data by finding, for example, the autocorrelation function of the input and the crosscorrelation function of input and output and then finding the power density spectra by Fourier transformation. The ratio of the two power density spectra becomes the frequency-domain representation of the system function. The usual problems encountered are those of sampling errors and data truncation, but an even worse difficulty presented itself here. The autocorrelation of the applied torque and the crosscorrelation of that torque with the



- Fig. XVI-3. Autocorrelation functions of angular deflections of splint when driving torque is zero; total time shift = 8.5 sec.
- (a) Top: normal subject, right hand strapped; $\theta_{\rm rms} = 0.061$ cm.
 - Bottom: normal subject, left hand free; $\theta_{rms} = 0.05$ cm, $f_{av} = 1.7$ cps.
- (b) Top: advanced Parkinsonism, right hand strapped; $\theta_{rms} = 0.172 \text{ cm}$, $f_{av} = 6.2 \text{ cps}$. Bottom: advanced Parkinsonism, right hand free; $\theta_{rms} = 0.236 \text{ cm}$, $f_{av} = 6.6 \text{ cps}$.
- (c) Top: cerebellar tremor, right hand strapped; $\theta_{rms} = 0.172$ cm, $f_{av} = 3.9$ cps. Bottom: cerebellar tremor, right hand free; $\theta_{rms} = 0.12$ cm.
- (d) Top: advanced Parkinsonism, right hand strapped (same as top of (b)). Bottom: normal subject, voluntarily rigid, right hand strapped; $\theta_{rms} = 0.092$ cm, $f_{av} = 7.9$ cps.

resulting angular deflection were practically identical; hence the transforms would differ by little more than a constant of proportionality, with the result that the impedance function amplitude spectrum would be so flat that the origin of the variations would, at best, be obscure. Consequently, computations in the time domain were performed by solving the convolution equation directly:

$$\phi_{TT}(\tau) = \int_{-\infty}^{\infty} h(t) \phi_{T\theta}(\tau-t) dt$$

where $\phi_{TT}(t)$ and $\phi_{T\theta}(t)$ are known, and we are to solve for h(t). Programs for the TX-0 computer were written for this computation; a discussion of their properties can be found in the thesis resulting from this work (1).

The result for all subjects, normal and abnormal, was an output impedance that

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resembled in every respect the character of a simple spring constant. It is not intuitively obvious that under the conditions of lowpass, random, low-impedance perturbations the deflection at the joint should be proportional to the driving force for frequencies from 0.2 cps to 30 cps. Tracking studies have shown that a sinusoidal signal of sufficiently low frequency can be followed with a very high degree of accuracy. However, with the proper choice of conditions and constraints, it can be shown that the mean-square angular deflection of the limb is a minimum for the output admittance characteristics actually found; that is, sH(s) is a realizable admittance, and the integral squared impulse response is fixed and nonoscillatory (specifically, $|H(\omega)|$ decreases monotonically with ω or is, at most, a constant).

Figure XVI-3 shows autocorrelation functions of the output noise of various subjects and indicates markedly different qualities that result from different nervous disorders. The $\theta_{\rm rms}$ figure quoted is the root-mean-square deflection of the tip of the 9-inch splint; a hand "free" means that the subject was holding the splint approximately 6 inches from the pivot, between two finger tips, and with the entire arm supporting itself naturally from the shoulder. The $f_{\rm av}$ is an estimate of the most obvious frequency component present. The origin of time is at the left, and 8.5 seconds of autocorrelation function are shown. The lowest frequency component visible, which also appears to be the frequency of "spindling" of the higher frequencies, is the respiration artifact.

The major effect of the results upon the derivation of a linear model that describes the self-regulating postural mechanism is that the loop gains of the various feedbacks from muscle, joint, and tendon are so large that they mask entirely the mechanical effects of the masses and geometry of the system.

A. R. Johnson

References

1. A. R. Johnson, The Servo-Analysis of Postural Reflexes, Ph. D. Thesis, Department of Electrical Engineering, M. I. T., May 1960.