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T. J. Hanratty

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INTRODUCTORY REMARKS TO SESSION ON ELECTROCHEMICAL METHODS OF TURBULENCE MEASUREMENT

Thomas J. Hanratty and Larry G. Chorn

This session is concerned with the use of the polarographic electrochemical technique to study flow fields and scalar transport. The principle involved in the technique is that an electrochemical reaction is carried out on a solid electrode surface held at a high enough voltage that the process is mass transfer controlled. The measurement of the electric current then is directly proportional to the rate of mass transfer to the test electrode.

If the test electrode is embedded in, but insulated from, a larger electrode it can be used to measure the local mass transfer rates in a mass transfer surface. If the test electrode is contained in an inert surface bounding a flowing fluid or is immersed in a flowing fluid it can be used to measure the velocity field. In these introductory remarks I would like to outline how this technique is used to measure a velocity field, to give a brief summary of some of the advances that have been made since the last turbulence conference at Rolla and to indicate some needs for future research.

DESCRIPTION OF THE TECHNIQUE

When used to measure velocities these electrochemical probes are the mass transfer analog of a constant temperature hot-wire or wall-film anemometer. The concentration of the reactant species at the electrode surface is controlled at a constant value of zero and temporal variations of the current flow then reflect temporal variations in mass transfer rate caused by the flow field.

The principal application of these techniques has been with small, platinum electrodes operated flush with a wall, for which they are analogous to the heated wall film used to measure the velocity gradient at a wall. The mass transfer probe has a number of advantages over the heat transfer probe. Auxiliary equipment is less costly. The reaction surface can be completely isolated from the substrate so that problems such as those associated with heat losses to the substrate do not exist. The calibration curve for the probe can be calculated since the boundary conditions

and differential equations describing the concentration field are well defined. Multiprobe configurations can be fabricated without concern about "cross talk".

A disadvantage of the mass transfer probe is in its frequency response, which is entirely associated with the scalar boundary layer. Since the Schmidt number characterizing the process is 700 or greater the concentration boundary layer is much thinner than what is encountered with a thermal probe operating in air or water. This gives rise to a much greater attenuation of the influence of the high frequency velocity components on the transfer process. Another disadvantage is that a certain amount of experimental art has to be developed in order to use the electrochemical technique. Contamination of the electrode surface and the occurrence of side reactions have to be avoided since these factors can lead to erroneous measurements.

Figure 1 shows how these techniques can be used to measure the x-component of the turbulent fluctuating velocity gradient, s_y , at the wall of a pipe (1). A platinum wire, 0.10 mm in diameter, or a platinum sheet, 0.076 mm x 0.076 mm, is glued into a hole in the wall of a pipe and sanded flush with the wall. If a rectangular electrode is used the long side is oriented perpendicular to the direction of mean flow. For the situation shown ferricyanide ion is converted to ferrocyanide ion on the test electrode which is the cathode of an electrolysis cell. It is converted back to ferricyanide ion at an anode, downstream from the cathode and of much larger area. A large excess of a neutral electrolyte such as sodium hydroxide is used to keep the solution electrically neutral. The applied voltage at the cathode is controlled at a large enough value that the reaction occurs rapidly enough to maintain the concentration of ferricyanide ions at a value close to zero but not large enough that side reactions are occurring. The current in the electrolysis cell is determined by measuring the voltage drop across a feedback resistor of an operational amplifier.

Figure 2 shows how these techniques could be used to measure the local velocity gradient on the wall of a

cylinder immersed in a flowing fluid. The position, relative to the flow direction, of a rectangular electrode embedded in the wall can be varied by rotating the cylinder. By using a sandwich of two rectangular electrodes insulated from one another, the direction of flow can be determined since the downstream electrode will give a lower signal due to decreased mass transfer.

The relation of the mass transfer rate to the flow field for the electrode configurations shown in Figures 1 and 2 is defined by the equations

$$\frac{\partial C}{\partial t} + S_{x}y \frac{\partial C}{\partial y} = D \frac{\partial^{2}C}{\partial y^{2}}$$
(1)

$$C = 0 \qquad y = 0$$

$$C = C_{\rm B} \qquad y = \infty$$
(2)

If a pseudo steady state assumption is made in that the $\frac{\partial C}{\partial t}$ term in (1) is ignored, the following relation between S_x and the electric current is obtained:

$$S_{x} = \frac{2}{3} \frac{\Gamma}{C_{B}} \frac{(4/3)}{F} \frac{I}{A_{e}} \frac{I}{n_{e}}^{3} \frac{9Le}{D^{2}}$$
 (3)

Sirkar (2) has shown that by measuring the sum and difference of the electric signals to two electrodes in a chevron arrangement both components of the velocity gradient at the wall S_x and S_z , can be measured simultaneously. The electrodes are 0.075 mm x 1 mm and are at an angle of 15° to the mean flow direction. The construction of these chevron electrode pairs, as indicated in Figure 3, is considerably more difficult than are single electrodes. It is of considerable interest in that the probe is quite sensitive at such low velocities. These results have been quite encouraging. We now have work in progress to see if we can use these probes in conjunction with wall probes to study turbulence structure. At the 1971 Rolla Conference we presented results obtained with the configuration of electrodes shown in Figure 7. These enabled us to measure simultaneously both components of fluctuating velocity gradient at a

RECENT ADVANCES

Advances have been made since the last turbulence conference at Rolla in the analysis of the frequency response, the choice of an electrochemical system, the design of electrodes, the use of electrodes as probes in the fluid and the use of multiple electrodes.

Fortuna (3) has examined the frequency response of wall electrodes by solving (1) and (2). The chief limitation in his analysis is that he considered a form of (1) which is linear in the fluctuating quantities. McMichael (4) has replotted Fortuna's calculations in the form shown in Figure 4. It is noted that the mass transfer boundary layer causes significant attenuation of the signal at $\omega^{+3/2}$ Sc^{1/2} L⁺ \cong 1. This clearly shows that the dimensionless frequency ω^+ , above which measurements need to be corrected for frequency response, decreases with increasing Schmidt number.

In more recent experiments from our laboratory we have been working with the iodine reaction used by Py and his students at the University of Nancy.

cathode:
$$I_3^- + 2e^- + 3I^-$$

anode: $3I^- + I_3^- + 2e^-$

This holds considerable advantages over the ferri-ferrocyanide reaction in that it is less subject to solution contamination and deterioration and that it is safer to use. In addition it has enabled us to carry out experiments on drag-reducing polymer solutions without being overly concerned about polymer degradation.

Eckelman (5) carried out experiments to explore whether electrochemical techniques can be used to measure velocity fluctuations in the bulk flow. He was unsuccessful in using wire probes of the type shown in Figure 5 because contamination of the probe caused a drift in the signal. We have traced this difficulty to the formation of string-like contaminants as a result of the degradation of the ferro-ferricyanide solution. More recently Bieg (6) and Zilker (7) have been successful by using the iodine electrolyte and by taking great precautions in filtering the fluid. The calibration curve shown in Figure 6 was obtained by Zilker. It is of considerable interest in that the probe is have been quite encouraging. We now have work in progress to see if we can use these probes in conjunction with wall probes to study turbulence structure.

At the 1971 Rolla Conference we presented results obtained with the configuration of electrodes shown in Figure 7. These enabled us to measure simultaneously both components of fluctuating velocity gradient at a number of locations at the wall (8). From these measurements we found that quite often the spatial variation of s_x and s_y showed the phase indicated in Figure 8a might dominate the flow. More recently Lee (8) has completed experiments in which he measured with a thermal probe the x and y components of the fluctuating velocity at y^+ = 13 at the same time he was measuring the wall velocity gradients with the electrode configuration in Figure 7. These results were in contradiction with the eddy structure shown in Figure 8a in that they showed a phase reversal of the y-component of the fluctuating velocity at a short distance from the wall. These results have led Lee to speculate that, on an average, the eddy structure might look more like what is indicated in Figure 9. Of course it would be expected the size of the small roller eddies would be continually changing and would even disappear for periods of time. The theoretical implication of these results

is that a pseudo-steady state model of the eddy structure 245

close to a wall might overlook important aspects of the flow.

The electrode configuration shown in Figure 7 was R used in a 20 cm diameter pipe. Its adaption to smaller S diameter pipes could be limited because of difficulties S with spatial resolution. For this reason the more compact electrode arrangements examined by Py (9) for measuring the two components of the wall velocity gradient t are of particular interest. We have been experimenting U with a twin electrode consisting of two semi-circles y used by Py. The problems involved in their fabrication y are not trivial. Figure 10 summarizes the method we have been using.

NEED FOR FUTURE WORK

We would like to close these remarks by pointing out some problems requiring the attention of future investigators.

Methods for dealing with the frequency response of these electrochemical probes are not completely satisfactory. The analysis carried out by Fortuna for wall probes should be extended so as to include cylindrical elements immersed in a fluid and the accuracy of the linear assumption made in the analysis should be determined.

The encouraging results recently obtained with cylindrical probes would suggest that more work be done to explore advantages that electrochemical probes might have over thermal probes in measuring flow fluctuations in the bulk fluid. Would it be possible to measure smaller velocities, to study the flow at smaller distances from the wall? Does the electrochemical method have advantages over the thermal method when using split-film probes since cross talk between the elements could be avoided?

Finally as can be seen from the construction techniques shown in Figures 3 and 10 there is a need to explore other methods to fabricate multi-electrode arrangements. For this purpose some of the techniques now being used to make printed circuits offer attractive possibilities.

SYMBOLS

A	Correction factor for frequency response
A _e	Area of electrode
C	Reactant species concentration
CB	Bulk concentration of reactant species
D	Molecular diffusivity of reactant species
F	Faraday's constant
I	Current
L ⁺	Dimensionless effective length of electrode

L _p	Effective length of electrode
n	Number of electrons transferred
Re	Feedback resistance
s	Schmidt number
s	Axial shear stress
s	Fluctuation component of axial shear stress
s,	Transverse shear stress component
t	Time
U	Fluid velocity
у	Distance normal to the pipe wall
y ⁺	Dimensionless distance
z	Circumferential distance
z+	Dimensionless distance

GREEK SYMBOLS

- $\Gamma(4/3)$ Gamma function of 4/3
- ω⁺ Dimensionless frequency
- ω^{\star} $\omega^{+}Sc^{1/3}$

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Figure 1. Configuration for Electrochemical Measurements in a Pipe



Figure 2. Electrode Configurations for Measuring Flow Around a Cylinder



(a) Preparation of Male and Female Plugs



(b)-(c) Mounting a Platinum Sheet Onto The Male Plug With Epoxy Glue



(d) Sand The Edge With Emery Paper To Separate Two Platinum Sheets



(e) Glue Two Plugs Together, Machine and Sand



(f) Cement, Machine and Polish

Figure 3. Fabrication of a Chevron Electrode



Figure 4. Frequency Correction for Wall Electrodes



Figure 5. Platinum Wire Mass Transfer Probe



Figure 6. Calibration Curve for Mass Transfer Probe of Figure 5



Figure 7. Multiple Chevron Electrode Configuration







Figure 8. Proposed Eddy Pattern



Figure 9. Lee's Proposed Eddy Pattern



(a) Prepare Two Resin Blocks With Groove and Glue Platinum Wires



(b) Milling and Sanding



(c) Glue Two Blocks Together Under Microscope



(d) Machine and Polish

Figure 10. Fabrication of Half-Circle Electrode