Fluid Elasticity Measurements from Hole Pressure Error Data*

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Synopsis

A slit die of variable height H (0.135 in., 0.1975 in., or 0.2488 in.) width W 2.5 in., and variable length L_e (12.0 in., 12.5 in., or 13.5 in.) has been designed and constructed for measuring the wall shear stress σ_w and the hole pressure error P_H. Values of P_H and σ_w are obtained directly and rapidly on a chart recorder from three commercial pressure transducers by means of an electronic differencing technique. The reliability of P_H measurements is established by the excellent agreement between viscosity η values obtained from this apparatus and a rheogoniometer and by measuring negligible values of P_H for the Newtonian fluid, glycerine, for $\sigma_w < 800$ dyn/cm² and values of $P_H > 0$ for $\sigma_w \geq 800$ dyn/cm² and values of $P_H < 0$ for a low-elasticity solution of polystyrene in dioctyl phthalate (DOP). Values of P_H are found to be independent of the distance E of the transducers from the die exit and the die height which indicated that P_H is independent of the undisturbed wall pressure P_w for constant values of σ_w. Our values agree well with those measured by Higashitani for a similar Separan solution in a different slit die. Using this new slit die, it is shown the P_H is sensitive to changes in molecular weight (MW) for a series of solutions of polystyrene in DOP and that P_H could be used to monitor continuously the changes in molecular structure which occurred during the mechanical degradation of a 2.0% aqueous poly(ethylene oxide) solution. The simple relation between P_H and N_1 ($P_H = -cN_1$, where c is a constant) is verified for solutions of polystyrene in DOP and aqueous Separan solutions over a large range of flow conditions ($\dot{s} = 1000$ sec⁻¹). Because c is found to vary slightly with polymer type, concentration, and molecular weight, a method is presented for obtaining N_1 directly from P_H data.

INTRODUCTION

In 1968, Broadbent et al.[1] discovered that measuring pressures in viscoelastic fluids flowing along a rigid wall by means of a pressure transducer mounted at the base of a small fluid-filled hole introduces a large systematic error. The difference between the pressure P measured at the base of the hole and the pressure P_w which would be measured if no hole were present has been defined to be the hole pressure error P_H (see Fig. 1). For viscoelastic fluids, P_H has been found to be negative, while for Newtonian fluids, P_H was found to be negligible at low Reynold's numbers,[2-4] becoming positive for large Reynold's numbers.[6]

In 1969, Tanner and Pipkin[3] calculated the magnitude of P_H for a viscoelastic fluid flowing across a hole of narrow rectangular cross section placed perpen-
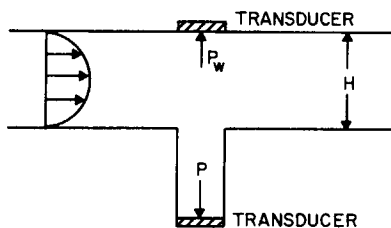
$$P_H = P - P_W$$

Fig. 1. Schematic drawing defining the hole pressure error. (In practice, the pressure difference due to the different vertical positions of the transducers is electronically offset.)

dicular to the flow direction using the second-order fluid theory. Their calculation, for very slow flows where the second-order fluid theory is valid, predicted that P_H should be simply related to the primary normal stress difference N_1 by the equation

$$P_H = -\frac{1}{4}N_1 \tag{1}$$

According to Tanner and Pipkin, measurements for a solution of polyisobutylene in cetane agreed well with this theoretical prediction. ($N_1 = P_{11} - P_{22}$, $N_2 = P_{22} - P_{33}$, where P_{ij} values denote physical components of stress for a unidirectional shear flow $v_1 = \dot{s}x_2$; $P_{11} > 0$ for a tensile normal component of traction.)

Other investigators have also found a simple linear relation between P_H and N_1.[4,5,7,11,12,13] The results of their experiments have shown that

$$0.15 \leq -P_H/N_1 \leq 0.25$$

It appears that the constant of proportionality in the relation $P_H = -cN_1$ depends on the polymer, concentration, and molecular weight, though not strongly.

More recently, Higashitani and Pritchard[8] estimated the magnitude of P_H for holes of various cross-sectional shapes. For viscoelastic fluids, they succeeded in obtaining relations between P_H and N_1, N_2 and the shear stress σ from a geometric analysis of the curvature of the shearing surfaces near the holes. Their result includes the calculations of Tanner and Pipkin (slot placed perpendicular to the flow) and Kearsley[4] (slot placed parallel to the flow) as special cases. Their theoretical values of P_H agree well with the experimental values of P_H for solutions of Separan AP 30 in water for the case of a circular hole.

Our interest in P_H stems primarily from this simple correlation between P_H and N_1. It is clear that an important new possibility arises: instead of regarding P_H as an error in the course of measuring pressures in viscoelastic fluids flowing along a wall, we may regard P_H as a measure of N_1 or at least a measure of some elastic property of the liquid if the constant c varies from fluid to fluid. We expect that P_H should show similar sensitivity to MW and molecular weight distribution (MWD) as N_1 does. In view of the fact that P_H can be generated in the flow of a viscoelastic fluid through a die, new possibilities arise for in-line quality control in polymer production and processing.
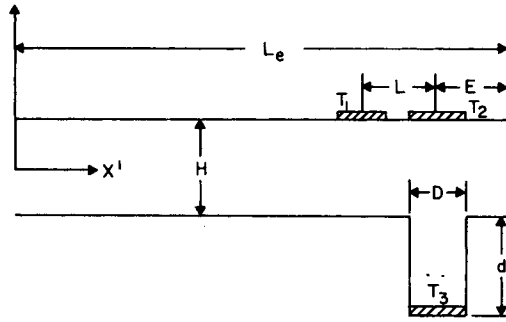
$$-(P_{22}-P_a) = \frac{\sigma_w}{H/2}(L_e - X')$$

$$P_H = P_3 - P_2 \qquad \sigma_w = \frac{P_1 - P_2}{L}\frac{H}{2} \qquad \dot{S} = \frac{Q_0}{3}\left(2 + \frac{d\ln Q_0}{d\ln \sigma_w}\right)$$

Fig. 2. Schematic drawing of the slit die showing the relative position of the pressure transducers in the die.

However, not everyone has found a simple relation between P_H and N_1. Han and Kim[10] found that P_H depends on the distance of the hole from the die exit and on the hole diameter, as well as on N_1, and that P_H was large and negative even for a Newtonian fluid. These results are in direct conflict with those of Higashitani.[2,5]

It should be pointed out that there is considerable difficulty in measuring P_H in flow through a die (this type of flow is termed a pressure-gradient flow) compared to measurements made in torque-generated flow or flow down an incline plane (nonpressure-gradient flow). The primary difficulty is that of obtaining a significant quantity from the small difference of two large pressures.

Thus far, only those who have recognized this problem have successfully measured P_H in a pressure-gradient flow. Novotny and Eckert[4] placed their pressure transducers 1 in. from the exit of their die. This permitted them to use fairly sensitive pressure transducers, since the local ambient pressures were small. However, they measured P_w and then recessed their pressure transducer and measured P. Furthermore, they checked neither the exit effects on their measurements nor the effect of using a shallow hole with a large diameter. On the other hand, Higashitani employed a suction/compression pump to reduce the pressure at the center of his die. He used his very sensitive transducers as null-point indicators and read the pressure differences from manometers.

The purpose of the work presented here is to: (1) present a newly constructed slit die which employs commercial pressure transducers for measuring values of P_H more accurately and rapidly and over a larger range of values of σ_w than previously measured by Higashitani; (2) establish the reliability of measurements of P_H made in this new apparatus; (3) investigate the sensitivity of P_H to changes in the molecular structure of polymer fluids; (4) investigate the relation between P_H and N_1 over a large range of flow conditions and for different fluids then previously used. The first part of this paper will be concerned with describing the new apparatus and establishing the reliability of the measurements of P_H and σ_w. The second part will cover applications of this apparatus.
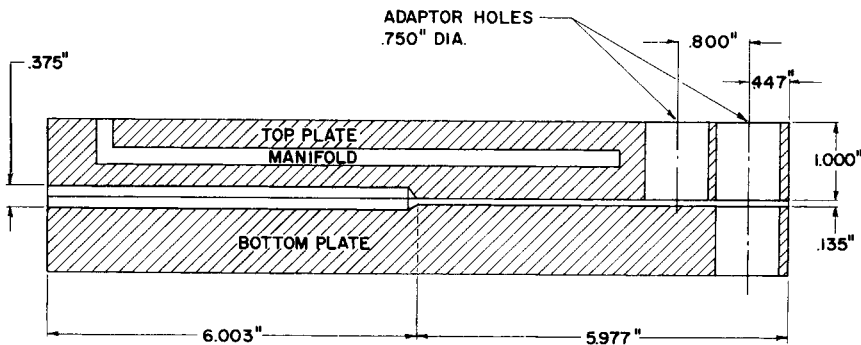
Fig. 3. Drawing of the slit die showing the basic dimensions.

EXPERIMENTAL

Apparatus

The basic principle of this apparatus was to reduce the ambient pressure for a given value of σ_w by placing the pressure transducers near the exit of the die and using a small aspect ratio (W/H). The arrangement of the three pressure transducers used to measure P_H and σ_w is shown in the schematic drawing in Figure 2. From transducers T_1 and T_2, σ_w is determined. P_H is obtained from the pressures measured by T_2 and T_3. The wall shear rate $\dot{s}_w$ is determined by means of the Weissenberg-Rabinowitch equation.[14]

The die consisted of two stainless steel plates in which were carefully milled the channel and a calming reservoir. The combined length of the channel and calming reservoir was 12 in., and the width was 2.5 in. The height of the channel was 0.135 in., and the reservoir height was 0.374 in. The height of the die could be increased to 0.1975 in. and 0.2588 in. by placing steel shims of thickness 0.0625 in. and 0.1238 in., respectively, between the top and bottom plates. A manifold was milled into the top plate through which water from a temperature control bath circulated. The position of the pressure transducers relative to the exit could be changed by the addition of extensions of length 0.5 in. and 1.0 in. which bolted to the end of the die. With no extensions in place, the centers of transducers T_2 and T_3 were 0.447 in. from the exit. The basic dimensions of the apparatus are shown in the drawing in Figure 3.

The pressure transducers were attached to the die through adaptors which pushed into holes drilled into the two plates. The adaptors which fitted into the top plate held the flush-mounted transducers T_1 and T_2. The adaptors which held transducers T_3 were designed with holes of various cross sections. All P_H measurements reported in this paper were made with a hole of circular cross section having a diameter of 0.125 in. and a depth of 0.375 in.

Considerable care was taken in the machining of this apparatus. All the critical dimensions such as the die height were machined to the nearest 0.0001 in. The worst recession of a flush-mounted transducer was 0.0005 and was due to the inaccurate machining of the transducers.

The crux of the apparatus was the pressure transducers. Diaphragm-capacitance transducers (Model 242 TC, Setra Co., Natick, Ma.) $\frac{1}{4}$ in. and $\frac{1}{2}$ in. in diameter were used in our measurements. The transducers were evaluated in
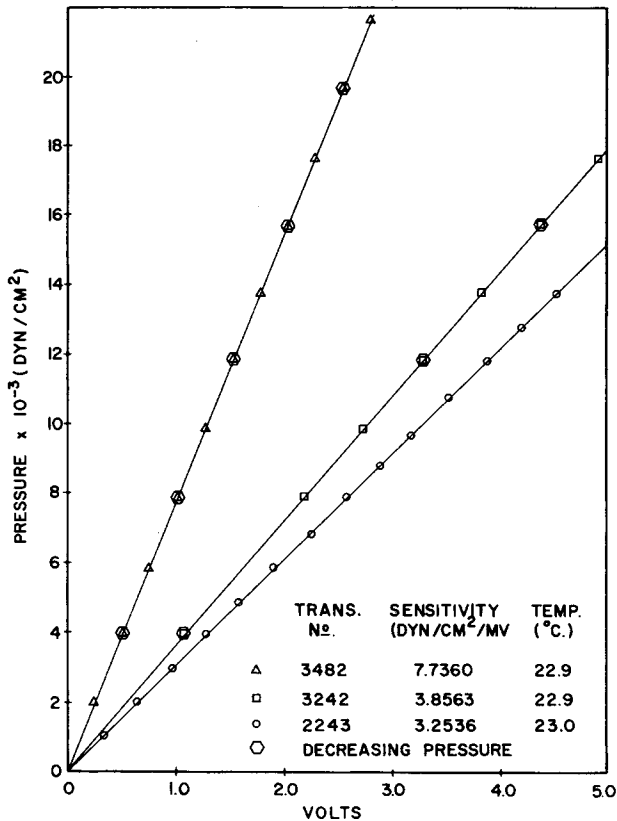
Fig. 4. Representative calibration data for three of the pressure transducers used in the slit die.

terms of their precision, temperature stability, and zero drift. A complete summary of the calibration data for these units is presented in Figure 4 for three of the pressure transducers. The worst deviation from the straight line through the data is ±31 dyn/cm^2. Although the reference pressure used in these experiments was atmospheric pressure, other reference pressures could be used.

An electronic differencing circuit was developed for evaluating P_H and σ_w from the outputs of transducers T_1, T_2, and T_3. The details of this circuit are given elsewhere.[15] The combined system of the pressure transducers and the electronic differencing circuit was calibrated against an inclined manometer using air pressure from a compressed air cylinder applied to both the manometer and the transducers. It was found that, at worst, P_H could be measured to the nearest ±17 dyn/cm^2 and that σ_w could be measured to the nearest ±3 dyn/cm^2.

It was discovered that the calibration factors of the transducers changed on mounting them in the die. For this reason, the end of the die was blocked off and air pressure was applied to the entrance of the die. The proper adjustments were made in the electronic circuit.

Fluids

The fluids used in this work were glycerine, which is generally accepted to be a Newtonian fluid, 1.5% and 3.0% (3 g polymer in 100 g solution) aqueous solu-
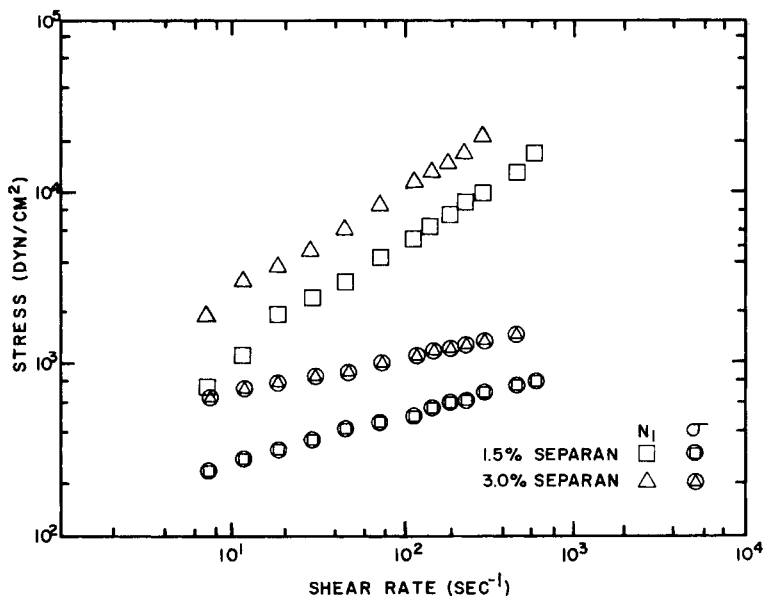
Fig. 5. Primary normal stress difference and shear stress of Separan AP 30 in water: (□) 1.5%; (△) 3.0%.

tions of Separan AP 30 (polyacrylamide) and 9.0% solutions of polystyrene in dioctyl phthalate. The polymers were supplied by Dow Chemical Co. and Pressure Chemical Co. The rheological properties of these fluids were measured by means of a Weissenberg Rheogoniometer (WRG) and are presented in Figures 5, 6, and 7. A list of all the fluids is presented in Table I.
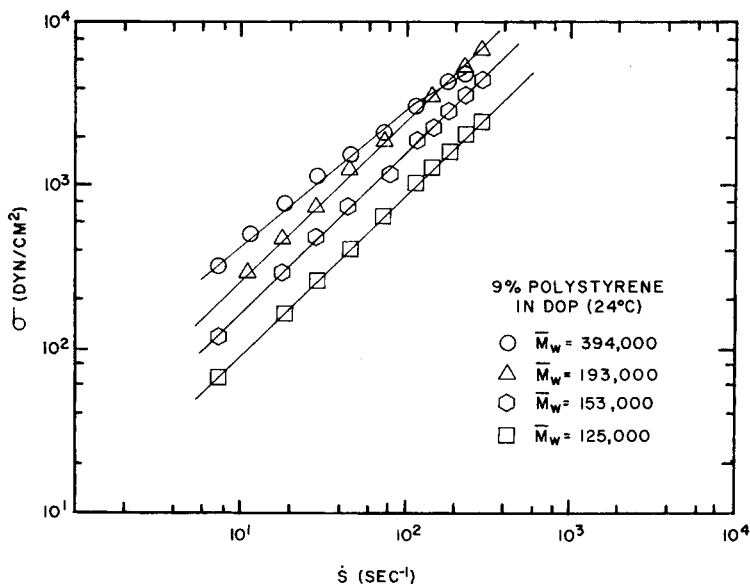

Fig. 6. Shear stress data for solutions of polystyrene in dioctyl phthalate: (○) $\bar{M}_w$ = 394,000; (△) $\bar{M}_w$ = 193,000; (○) $\bar{M}_w$ = 153,000; (□) $\bar{M}_w$ = 125,000.
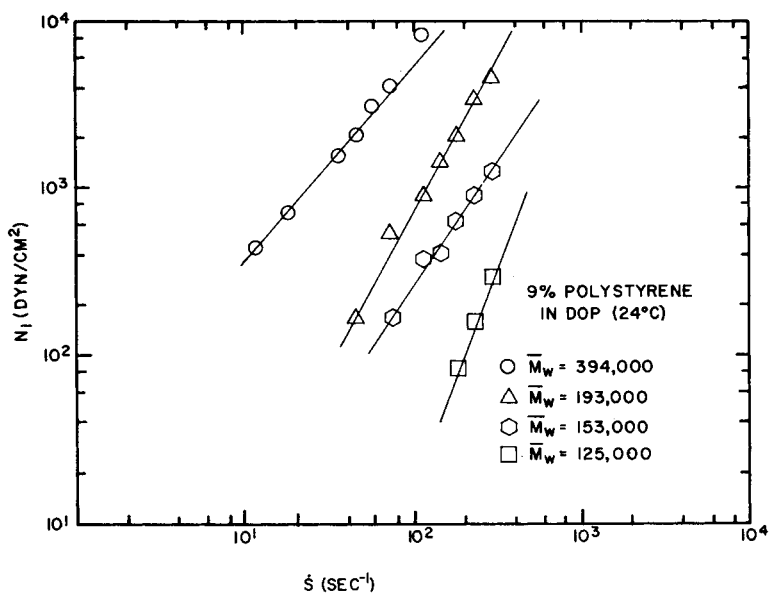
Fig. 7. Primary normal stress difference N_1 data for solutions of polystyrene in dioctyl phthalate: (O) $\bar{M}_w$ = 394,000; (△) $\bar{M}_w$ = 193,000; (O) $\bar{M}_w$ = 153,000; (□) $\bar{M}_w$ = 125,000.

These fluids were circulated continuously through the slit die by means of a gear pump. The flow rate was determined by measuring the motor rpm's.

RESULTS AND DISCUSSION

Reliability of P_H Measurements

As noted earlier, there is considerable difficulty in measuring significant values of P_H in the presence of large ambient pressures. It is possible for errors in individual pressure measurements to cover up values of P_H associated only with

TABLE I
Polymer Fluids Used In These Experiments

Fluid no.	Polymer	Solvent	Concentration, %	Molecular weight
1	polyacrylamide	water	1.5	
2	polyacrylamide	water	3.0	
3	polystyrene	dioctyl phthalate	9.0	125,000 $\bar{M}_w$
				119,000 $\bar{M}_n$
4	polystyrene	dioctyl phthalate	9.0	153,000 $\bar{M}_w$
				147,000 $\bar{M}_n$
5	polystyrene	dioctyl phthalate	9.0	193,000 $\bar{M}_w$
				182,000 $\bar{M}_n$
6	polystyrene	dioctyl phthalate	9.0	394,000 $\bar{M}_w$
				372,000 $\bar{M}_n$
7	polystyrene	dioctyl phthalate	3.0	862,000 $\bar{M}_w$
				750,000 $\bar{M}_n$
8	poly(ethylene oxide)	water	2.0	4,000,000 $\bar{M}_w$
9	glycerine			

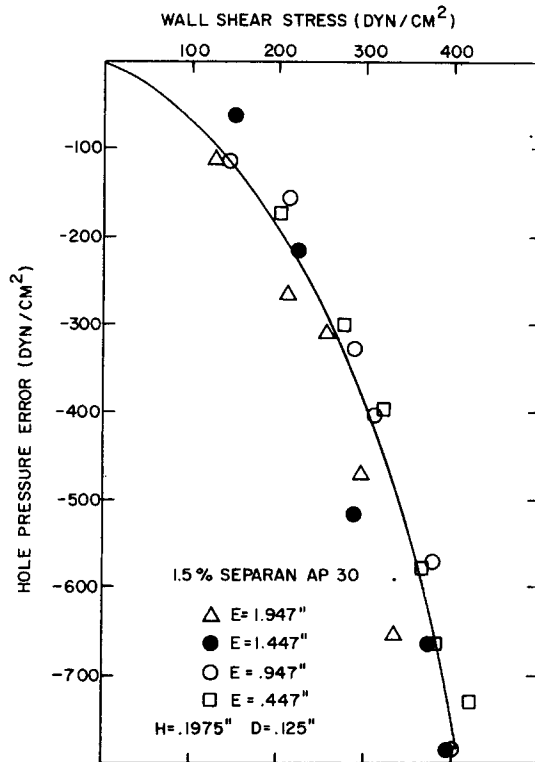WALL SHEAR STRESS (DYN/CM^2)

Fig. 8. Values of P_H recorded on a chart recorder for four positions of the transducers relative to the exit for a 1.5% Separan AP 30 solution in water.

fluid elasticity. For this reason, the first part of the paper will be concerned with establishing the reliability of P_H measurements using the new slit die.

Values of P_H were measured for four different distances E of the pressure transducers from the die exit and three die heights. The results are presented in Figures 8 and 9. The scatter in the data of Figure 8 appears to be random and indicates that there are no systematic errors in the measurement of P_H due to flow disturbances at the exit. Likewise, P_H is unaffected by changes of the die height even for the case of $W/H = 9.7$.

In addition to showing that P_H is independent of H and E, these data show that P_H is independent of P_w for a given value of σ_w. This is in disagreement with the results of Han and Kim[10] who found that P_H depends on the distance from die entrance and thus on P_w. This suggests that their results may have been subject to some systematic error.

These data are of further significance, since they give us confidence that the random errors are small. We obtain repeatable values of P_H for a given value of σ_w, even though P_w changes significantly.

Viscosity values for aqueous solutions of Separan AP 30 in water and glycerine obtained from a Weissenberg rheogoniometer are compared with values obtained from the slit die. As is observed from Figure 10, the values are in good agreement. This result gives us confidence that there are no large systematic errors in our pressure measurements. Furthermore, it shows that this apparatus yields viscosity values which are consistent with those obtained from the rheogoniometer.
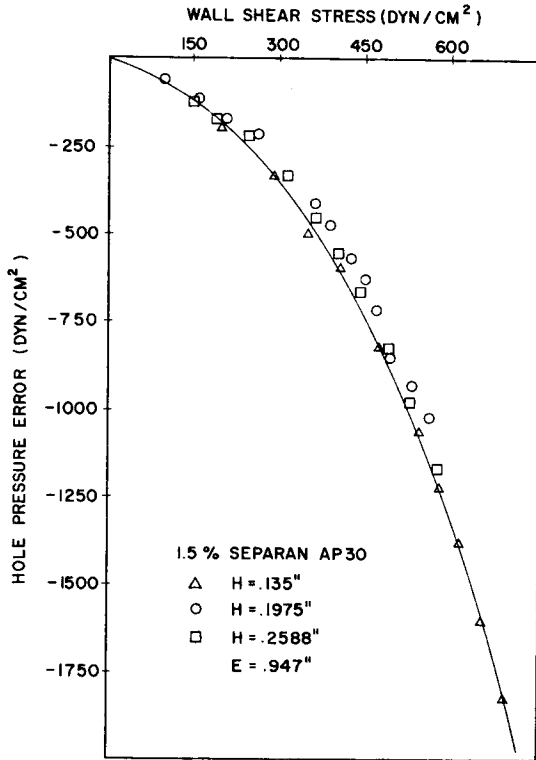
Fig. 9. Comparison of values of P_H for three different die heights.

We realize that on numerous occasions, other individuals have published viscosity data measured in a slit die,[4,16,17] and in some cases they compared their values with rheogoniometer data.[5,18] The purpose of our viscosity data is to give confidence in our P_H measurements taken with this apparatus and to show that
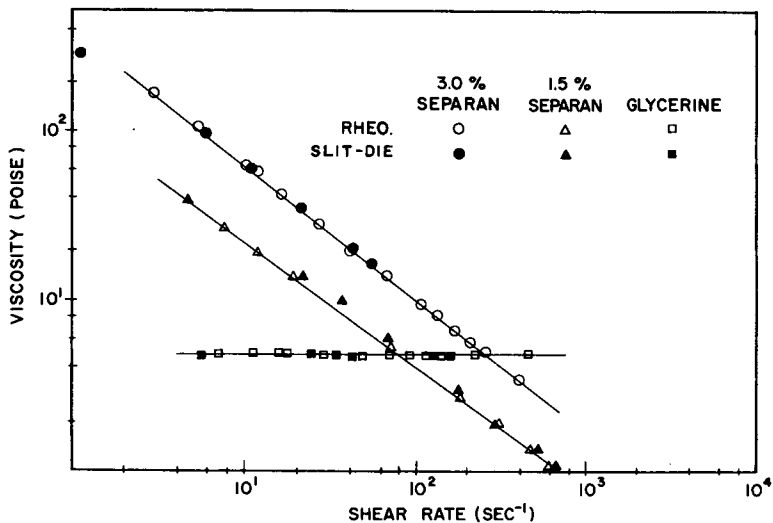

Fig. 10. Viscosity values for solutions of Separan AP 30 in water and for glycerine obtained from a rheogoniometer compared with values obtained from the stressmeter.
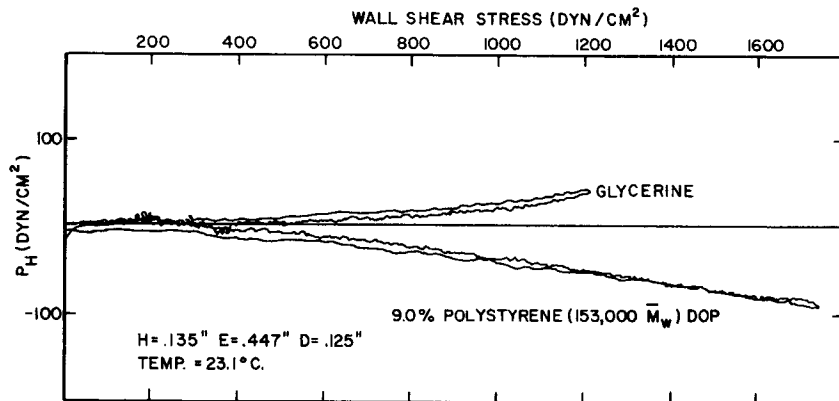
Fig. 11. Comparison of values of P_H for a Newtonian fluid and a non-Newtonian fluid of low elasticity (continuous traces taken from an X–Y recorder).

in the presence of a hole and with pressure transducers mounted near the exit of the die, one still obtains accurate viscosity data.

In Figure 11, values of P_H for glycerine are compared with values of P_H for a fluid of low elasticity (this fluid exhibits no values of N_1 for shear rates $\dot{s}$ up to 45.96 sec^{-1} and values of σ_w equal to 1200 dyn/cm^2. The negligible values of P_H for the Newtonian fluid are in agreement with those of other researchers[2-4] over the same shear stress range, and the positive values of P_H agree qualitatively with the theoretical predictions of Thom and Apelt[6] for a related problem. On the other hand, the values of P_H for the polymer solution were negative, reaching a value of -50 dyn/cm^2 at $\sigma_w = 1200$ dyn/cm^2. From these results we can conclude that (1) the negative values of P_H are due to elastic effects and not to systematic errors in the pressure measurements; (2) the slit die gives significant P_H data for fluids of relatively low elasticity ($N_1/\sigma^2 \sim 4.0 \times 10^{-5}$ cm^2/dyn).

It is important to compare our values of P_H with those measured by Higashi-tani using similar polymer solutions. In Figure 12, values of P_H from Higashi-tani's measurements are compared with values of P_H obtained from our apparatus for a 1.5% Separan solution. The agreement of the measurements is interesting for the following reasons. First, in view of the conflict between the conclusions of Han and Higashitani, these results give us confidence that Higashitani's findings are correct. Second, similar values of P_H can be obtained from two different instruments using different pressure measuring techniques. Third, there is further reason to believe that placing the transducers near the exit yields consistent values of P_H.

Applications of the Apparatus

The possibility of using this apparatus to detect changes in molecular structure of polymer solutions is explored in this section. In particular, we look at the sensitivity of P_H to changes in MW for a series of solutions of polystyrene in DOP and the capability of this apparatus to detect changes in MW on the continuous in-line basis.

The P_H was measured for solutions of polystyrene of different $\bar{M}_w$, but of narrow MWD in DOP. Traces of continuous curves of P_H versus σ_w obtained
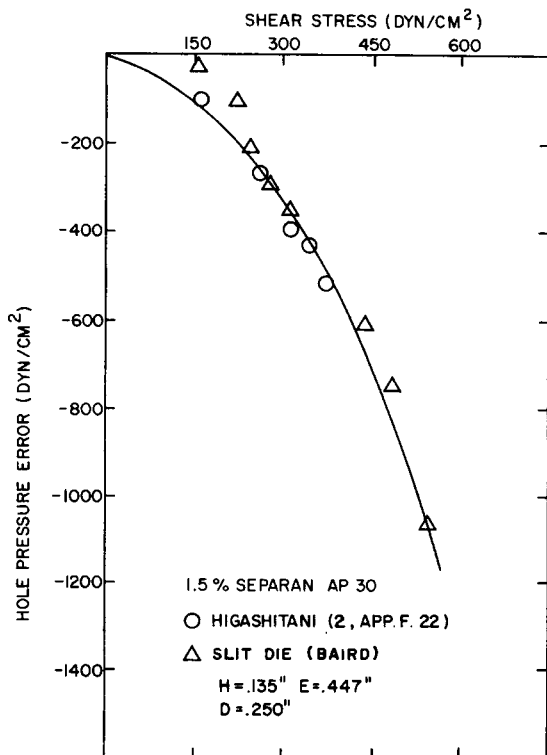
Fig. 12. Comparison of P_H values for a similar 1.5% Separan solution in water measured by Higashitani in a different slit die apparatus and this apparatus.
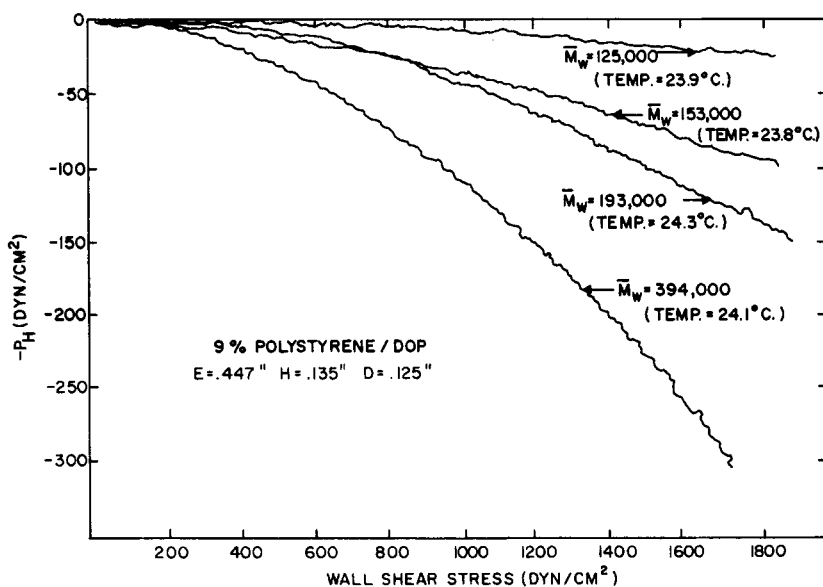

Fig. 13. Data showing the sensitivity of P_H to molecular weight for a series of 9.0% polystyrene/DOP solutions (traces taken from a X–Y recorder).
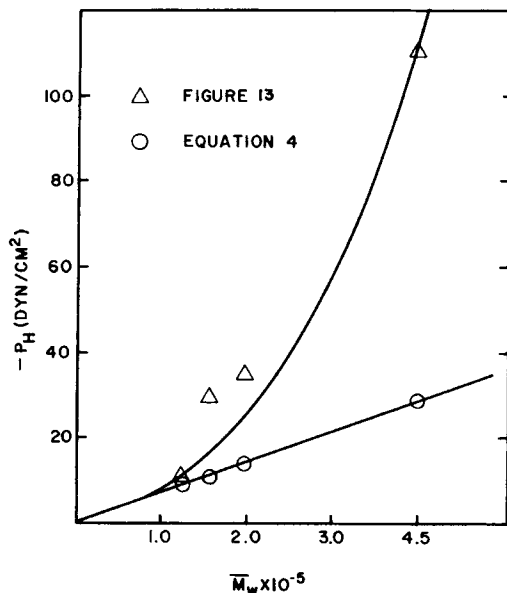
Fig. 14. Comparison of values of P_H calculated using eq. (4) with values from Fig. 13 taken at σ = 1000 dyn/cm^2.

from an X–Y recorder are presented in Figure 13. These results show that this slit die is capable of detecting MW changes.

The steady-state compliance J_e^0 can be related to values of N_1 and σ^2 obtained from steady-shear experiments by the following relation:

$$J_e^0 = \lim_{\dot{s} \to 0} \frac{N_1}{\sigma^2} \tag{2}$$

According to the Rouse theory for concentrated polymer solutions,[19]

$$J_e^0 = 0.4 \, M/CRT \tag{3}$$

where M is the molecular weight and C is the concentration. Assuming that eq. (2) is applicable for finite values of $\dot{s}$ and using $P_H = -0.2N_1$ (see Fig. 18) for the polystyrene solutions, eqs. (2) and (3) yield the following relations between P_H and M:

$$-P_H = 0.16\sigma^2 M/CRT \tag{4}$$

In Figure 14, we compare values of $-P_H$ calculated from eq. (4) with values

TABLE II
Comparison of J_e^0 Values Calculated From Equation (3) and Using $N_1/2\sigma^2$ Values

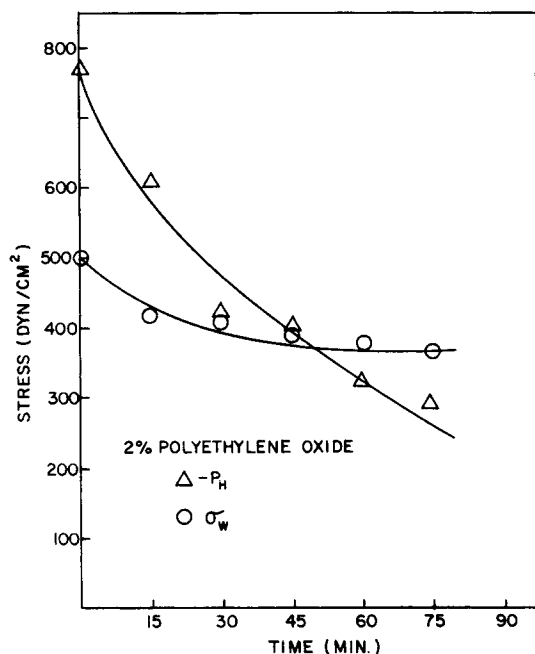| $\overline{M}_w$	$0.4 \dfrac{M}{CRT} \times 10^5$	$\dfrac{N_1}{2\sigma^2} \times 10^5$ cm^2/dyn
125,000	2.27	2.38
153,000	2.78	4.20
193,000	3.50	5.84
394,000	7.15	53.33

Fig. 15. Monitoring of the mechanical degradation by a gear pump of a 2.0% PEO/water solution using the stressmeter (flow rate 25 cc/min).

taken from the curves in Figure 13 at a value of $\sigma = 1000$ dyn/cm^2. We see that there is only qualitative agreement with the data.

There are several reasons why the agreement is only qualitative. First, the relation between J_e^0 and N_1 and σ^2 is not completely applicable at high shear rates. In our case, the values of $\sigma = 1000$ dyn/cm^2 corresponded to values of $\dot{s}$ from 25 to 114 sec^{-1}. Second, the difference between the values of P_H for the different MW polymers was of the order of the accuracy of the pressuring measuring system. Thus, the error in measuring P_H itself may be large compared to the difference of the P_H values. Third, it seems that the MWD of these polymers may have been broader than indicated. (This is possible, since the samples were stored for several years before being used.) It is known that J_e^0 is very sensitive to the high molecular weight tail of the MWD[20] and will be larger for polymers of broad MWD. We suspect this to be the case since values of J_e^0 calculated from eq. (2) using N_1 and σ values obtained from a WRG were larger than those values determined from eq. (3) (see Table II). In fact, we observed disagreements for the values of J_e^0 similar to those for the P_H values in Figure 14.

The apparatus was used to monitor continuously the mechanical degradation of a 2.0% aqueous solution of poly(ethylene oxide) (PEO) by a gear pump. Values of P_H and σ_w were recorded as a function of time during the degradation process and are present in Figure 15. (Note: Figure 15 presents individual data points obtained from continuous chart recordings, $(P_1(t), P_2(t)$, and was prepared before the electronic differencing circuit was constructed.) It is seen in Figure 15 that P_H changes from -771 dyn/cm^2 to -280 dyn/cm^2 after 75 min of degradation, whereas σ_w (which is related to the viscosity η) changed from 500 to 367 dyn/cm^2. This behavior is similar to that observed by Higashitani[2] who inves-
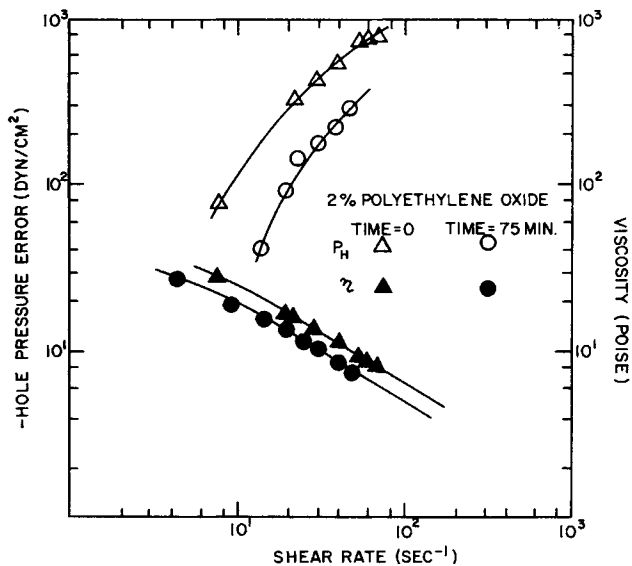
Fig. 16. Comparison of values of P_H and η before and after the degradation of a 2.0% PEO solution.

tigated the degradation of a similar PEO/water solution by means of a sampling procedure combined with rheogoniometer measurements.

In Figure 16, values of P_H and η versus the wall shear rate $\dot{s}_w$ measured at the beginning and the end of the degradation process are presented. It is seen that the change in the values of P_H was much larger than the change in the values of η.

The results are not only interesting, but important for the following reasons. First, this experiment demonstrates that P_H is more sensitive to changes in molecular structure (possibly molecular weight distribution in this case) than is viscosity. Second, the slit die is capable of monitoring continuously the properties of a flowing polymer solution whose molecular structure is changing with time. Third, as far as we are aware, this represents one of the first uses of a nonlinear elastic property to monitor continuously the behavior of a non-Newtonian liquid whose properties are changing with time.

Correlation Between P_H and N_1

For a hole of circular cross section, the theory of Higashitani and Pritchard predicts the following expression[8] for the magnitude of P_H:

$$P_H = - \int_0^{\sigma_w} \frac{N_1 - N_2}{3\sigma^2} \, d\sigma \tag{5}$$

where N_1 and N_2 are the primary and secondary normal stress differences, respectively, and σ_w is the wall shear stress. For fluids in which $N_1 \propto \sigma^n$ and N_2 is negligible, eq. (5) reduces to the following equation:

$$P_H = -N_1/3n \tag{6}$$

Because some agreement[5] with data has been found for eq. (6), it is possible that the theory of Higashitani and Pritchard may be more generally applicable than the theory of Tanner and Pipkin.
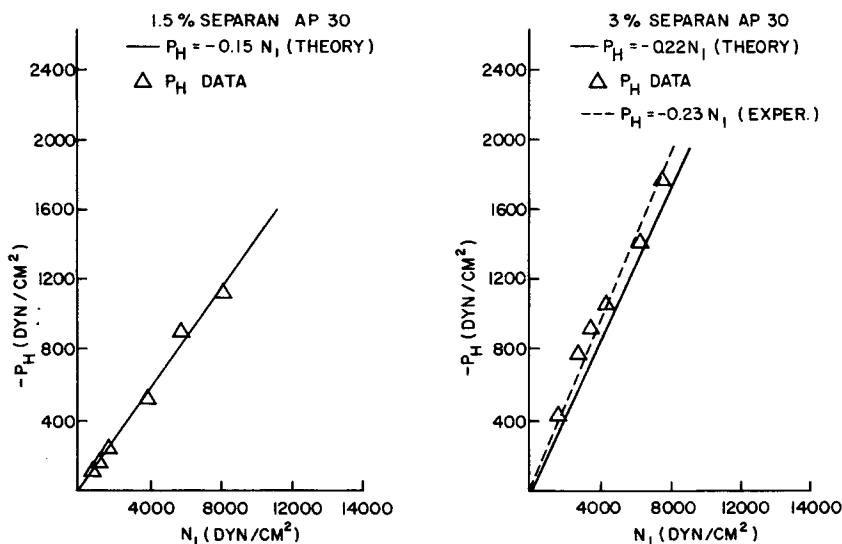
Fig. 17. Comparison of measured and calculated values of the hole pressure error P_H for solutions containing 1.5% and 3.0% Separan AP 30 in water. Lines represent the values predicted by the theory of Higashitani and Pritchard (from eq. (5) using $N_2 \approx 0$ and $N_1(\sigma)$ evaluated from rheogoniometer data).

Values of P_H at a given $\dot{s}_w$ are plotted versus values of N_1 obtained from a rheogoniometer at the same $\dot{s}$ in Figures 17 and 18 for aqueous solutions of Separan AP 30 and for solutions of polystyrene in DOP. The agreement between the experimental values and the theoretical values predicted by eq. (6) using $N_1(\sigma)$ values from the WRG is fairly good. (Note: For the 3.0% Separan solutions, only values of N_1 for $\dot{s}$ up to 100 sec^{-1} were considered accurate, since the normal forces generated by shearing this material exceeded the linear range of the spring in the normal force measuring system.)

In most cases, the experimental values of P_H were slightly larger in magnitude than those predicted by theory. Assuming that N_2 is negligible in eq. (5) may be partially responsible for the slight difference. However, before definite conclusions can be reached in regard to this slight discrepancy, accurate N_1 and N_2 values must be available.

The results of Figures 17 and 18 are important for the following reasons. First, the results of Figure 17 reconfirm the findings of Higashitani. Using Separan solutions similar to those of Higashitani, we measured values of P_H which agreed in magnitude with those of Higashitani. We also found that the theoretical and experimental values were in good agreement and that the simple relation between P_H and N_1 was similar to that found by Higashitani. Second, we have found that for solutions of polystyrene in DOP, P_H correlated well with N_1 alone by the simple relation $P_H = -cN_1$. Furthermore, the theoretical predictions of Higashitani and Pritchard agree fairly well with the experimental values of P_H. This result furnishes further evidence suggesting that the theory of Higashitani and Pritchard may be generally applicable at least for the case of a circular hole. Third, we see that even for fluids of relatively low elasticity ($N_1/\sigma^2 = 8.0 \times 10^{-5}$ cm^2/dyn) for the 9.0% polystyrene solution as compared to 260×10^{-4} cm^2/dyn for the 1.5% Separan solution) we can still correlate P_H with N_1.

The measurement of P_H and η for viscoelastic fluids in a die offers the advantage of obtaining rheological information at shear rates of practical impor-
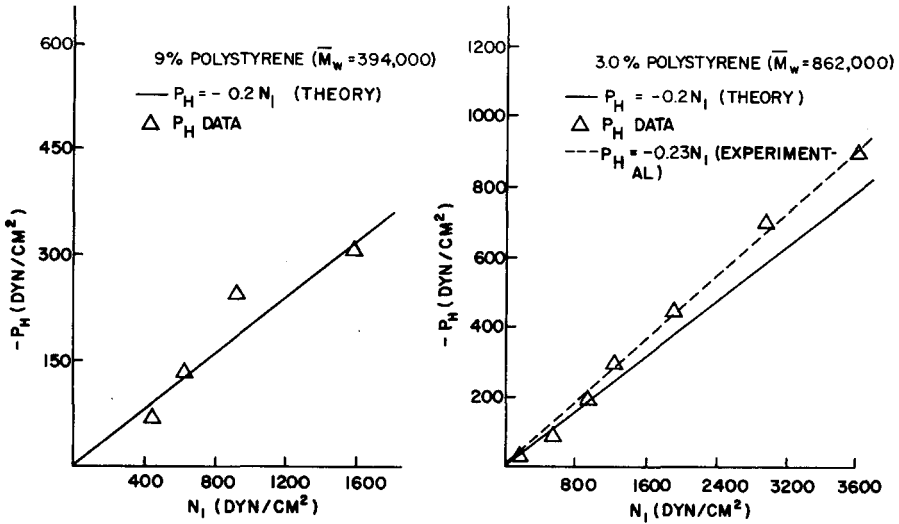
Fig. 18. Comparison of measured and calculated values of P_H for solutions of polystyrene in DOP. Lines represent the values predicted by the theory of Higashitani and Pritchard.

tance. In Figure 19, we have plotted P_H and N_1 versus $\dot{s}$ for a 1.5% aqueous Separan solution. It appears that P_H is still simply related to N_1 for values of $\dot{s}$ of up to nearly 1000 sec^{-1}. We note that the capacity of the gear pump employed here was the limiting factor which prevented us from obtaining higher $\dot{s}$. We note also that values of N_1 were measured on the rheogoniometer up to $\dot{s}$ = 578 sec^{-1}, and the line through N_1 was extrapolated to 1000 sec^{-1}.

This result disagrees with that of Novotny and Eckert[4] who found that at about $\dot{s}$ = 100, P_H/N_1 began to decrease and eventually went to zero for solutions of PEO in water. They attributed this to secondary flows in the hole which resulted in the streamlines diverging from the hole rather than dipping into the hole. On plotting their data versus the Reynold's number Re, where Re = $4\rho H \langle v \rangle /\eta$, where ρ = density, $\langle v \rangle$ = average velocity, and η = $\sigma_w/\dot{s}_w$, they found $P_H/N_1 = 0$ at Re
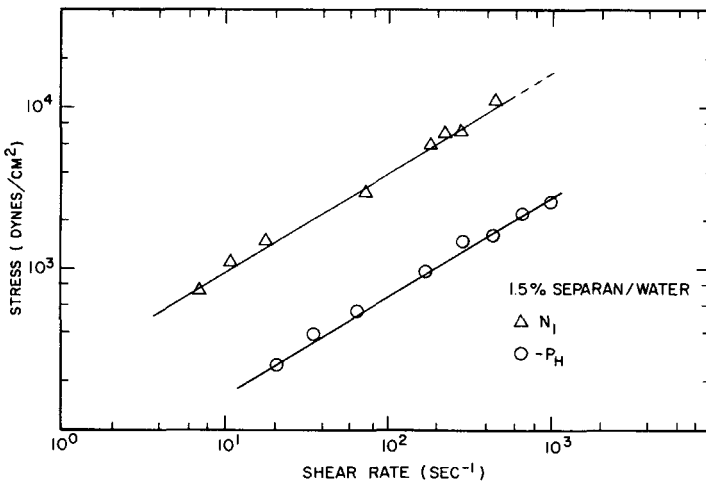

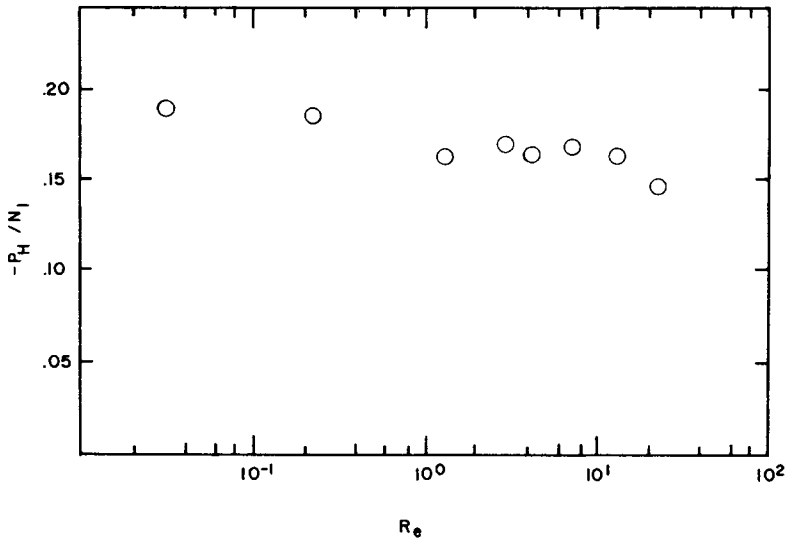Fig. 19. Comparison of P_H with N_1 at high shear rates for a 1.5% Separan solution in water.

Fig. 20. Values of $-P_H/N_1$ plotted against Re (Re $= 4\rho H \langle v \rangle /\eta$) for a 1.5% Separan AP 30 solution.

= 6 and became positive at Re = 10. In Figure 20, we have plotted the values from Figure 19 as $-P_H/N_1$ versus Re as defined above in order to compare our results with those of Novotny and Eckert. (We realize that there may be a more appropriately defined Re which should include the dimensions of the hole as is suggested by the recent calculations of O'Brien[21] for the Stoke's flow of a viscous Newtonian fluid over a cavity.) There appears to be only a small systematic decrease, but this is hardly significant in relation to the error in measuring P_H and N_1.

Novotny and Eckert speculated that the breakdown in the simple relation between P_H and N_1 would be characteristic of other viscoelastic fluids over
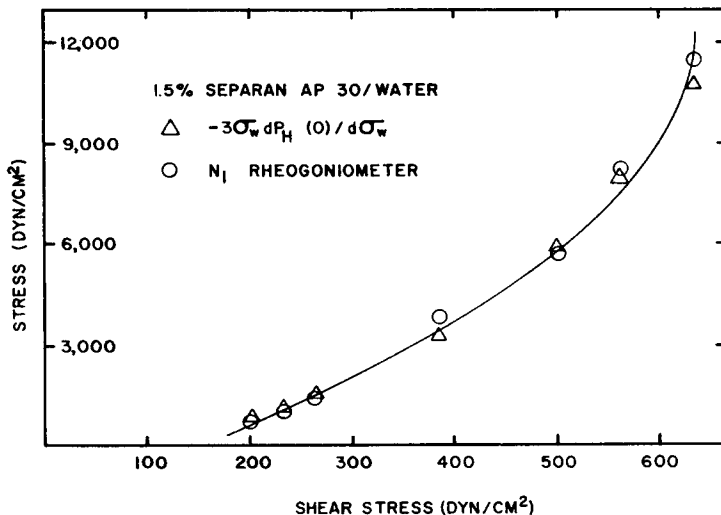

Fig. 21. Values of $-3\sigma_w dP_H/d\sigma_w$ obtained from a continuous $P_H(\sigma_w)$ curve compared with values of N_1 obtained from the rheogoniometer for a 1.5% Separan AP 30 solution in water.

similar flow conditions.   However, we feel that our results show that for a suitably designed apparatus the simple relation exists for a large range of flow conditions. Novotny and Eckert used a hole of diameter $D = 0.5$ in. and depth $d = 0.072$ in. (i.e., $D/d = 7$).   It is most likely that the influence of the flow at the bottom of the hole was not negligible and that the inertial effects at higher Re swamped the elastic effects.   In our apparatus, $D/d = 0.3$, and the influence of the fluid motion at the bottom of the hole was probably insignificant even at Re = 23.0.

It is fairly well established by now that for some polymer solutions a simple relation ($P_H = -cN_1$) between P_H and N_1 exists.   However, the value of c varies from fluid to fluid, although not greatly, and thus one cannot obtain N_1 from P_H data using this simple relation.   We have seen that eq. (5) gives a good estimate of P_H for holes of circular cross section for several different polymer fluids.   The question which arises is: can normal stress values be obtained from hole pressure error data directly?

Differentiation of eq. (5) yields the following expression:

$$-3\sigma_w dP_H/d\sigma_w = N_1 - N_2 \tag{7}$$

Provided that continuous curves of P_H versus σ_w are available, the theory of Higashitani and Pritchard is valid, and $N_2 = 0$; then in principle a reasonable estimate of N_1 can be obtained from P_H data.

In Figure 21, values of $-3\sigma_w dP_H/d\sigma_w$ are compared with values of N_1 obtained from a WRG (the values of $dP_H/d\sigma_w$ were evaluated by fitting a polynomial to the continuous curves of P_H versus σ_w obtained from the slit die).   The values of N_1 agree well with the values of $-3\sigma_w dP_H/d\sigma_w$.   This result also suggests that $N_2 \ll N_1$, which is in agreement with the results of Christiansen and Leppard,[19] who found that $0.03 \leq -N_1/N_2 \leq 0.2$ for solutions of polyacrylamide and poly-(ethylene oxide).

CONCLUSIONS

Although part of this work has been involved with establishing the reliability of P_H measurements made in this newly constructed apparatus and extending some of the ideas of others, we feel there are some important and new contributions presented here.   First, we have designed an apparatus which has the capability of measuring P_H rapidly and on a continuous in-line basis.   Second, we have shown that even for fluids of relatively low elasticity significant values of P_H can be measured.   Third, we have shown that P_H is sensitive to changes in the molecular structure of polymer solutions and that it is possible to continuously monitor these changes, at least for the case of polymer degradation, in this newly designed apparatus.   Finally, since continuous P_H-versus-σ_w curves can be obtained, the possibility of obtaining N_1 directly from P_H data is established.

Notation

c constant
C concentration, g/cc
d hole depth
D hole diameter

E distance from the die exit of the centers of transducers T_2 and T_3

H die height

J_e^0 steady-state compliance

L distance between the centers of transducers T_1 and T_2

L_e die length

P pressure measured at the base of a hole

P_w undisturbed pressure measured at the wall of a die

P_H hole pressure error measured using a circular hole

Re   Reynold's number

T_i transducer T_1, T_2, and T_3

W die width

N_1 primary normal stress difference ($P_{11} - P_{22}$, with $P_{11} > 0$)

N_2 secondary normal stress difference ($P_{22} - P_{33}$)

σ shear stress (P_{12} or P_{21})

σ_w shear stress at the wall

σ_{22} normal component of traction at the die wall

$\dot{s}$ shear rate

$\dot{s}_w$ wall shear rate

Q flow rate, cc/sec

Q_0 $6Q/WH^2$

η viscosity

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