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Pressure Determinations along Die Axis in a Round Tube Extrusion*

(Received June 14, 1967)

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Abstract

Pressure distributions along the die axis in a round tube extrusion for a commercial grade polystyrene and a low pressure polyethylene were carried out by using a modified commercial extruder and electric membrane type pressure gauges. Measured pressure drops along the length of the tube containing a larger preceding channel in diameter were discussed numerically in terms of the tube length correction term coefficient and the viscosity resistance of the flowing liquid. The residual pressure at the tube exit, expected to show the pressure energy necessary to cause the Barus effect, was apparently observed with a tendency to increase with shear rate. Operations of the elastic shear stress in the steady laminar flow were persisted.

I. Introduction

In a round tube extrusion, it has been pointed out that the direct determination of the pressure distribution along die axis has a most significant meaning not only to clarify the physical meaning of the end correction¹⁾ or the tube length correction term coefficient²⁾, but also to reveal the elasticity effect in the steady state flow of viscoelastic liquid²⁾⁻⁷⁾. A few papers^{8)-10),6)} have been presented on the pressure determination, but unfortunately they are not in terms of the coefficient. As a matter of fact, entirely different interpretations^{2), 5), 11)-14)} of the cause of the Barus effect¹⁵⁾ have been presented due to the scarcity of the informations on the experimental results of the pressure distribution.

In the present research, pressures are measured for the viscoelastic flows of polymer melt in a straight cylindrical die tube of an extruder preceded by a larger

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approaching channel. The measured pressure distributions along the axis are directly compared with the predicted one by Arai and Aoyama²⁾ and discussed in terms of the tube length correction term coefficient and the Barus effect observed with the same substance.

II. Experimental procedure

Materials investigated in the present work were the commercial grade high density polyethylene Sholex 6009 of Nihon Olefin Co. (Melt Index : 0.79) and polystyrene Styron 683 of Asahi Dow Co., both being fabricated with the help of screw extruder.

Pressure determinations were carried out for the steady state flow in a modified commercial screw extruder at several points along the length of the flow direction at the adaptor, reservoir and die. The inner diameter of the extruder barrel is 40mm. By using Mikipulley device, the revolution speed of the screw can change continuously between 4 and 88 rpm. The variation of the extrusion rate was estimated to fall in the range of between 1.3 and 20 litre/hr. The flow rate and the temperature were selected within the region where the melt fractures¹⁶⁾ or pulsations²⁾ didn't occur but shear rate dependence of the Barus effect was obvious. Two dies of different dimensions of the tube radius and length were used. The included die entry and exist angles were equally 180° . The cross-sectional view of one of the dies attached to the extruder barrel is shown in Figure 1. As seen in the figure, electrical membrane type pressure gauges were mounted in the specially machined taps, to each of which respective wall surface pressure was propagated through a

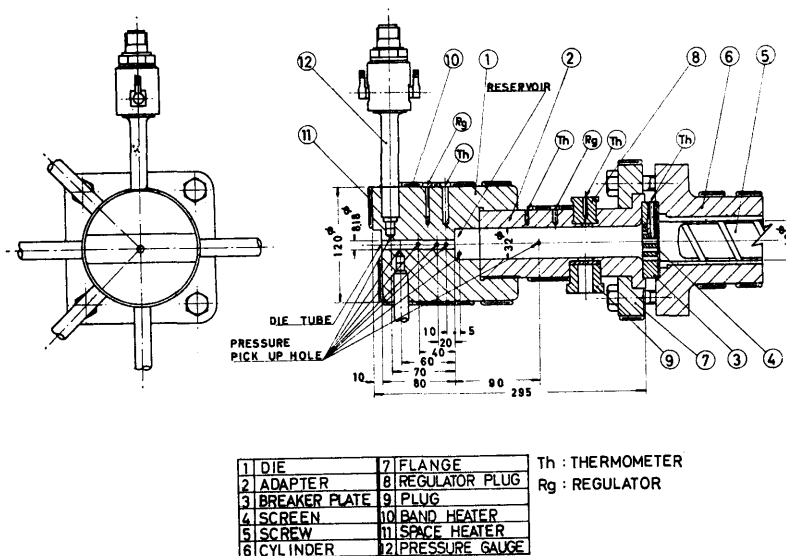


Fig. 1. Cross-sectional view of the die and the adaptor for the pressure distribution measurements

hole of 1, or 1.2 mm in diameter drilled vertically onto the wall surface, without disturbing the flow patterns of the flowing melt.

Figure 2 shows the overall view of the pressure gauges set up on the die and the reservoir. In order to screw the gauge into the tap under a constant torque, the use of a torque-wrench was recommended. Rubber tubes in the figure were for the circulation of cooling water for the pressure gauges. It is desirable to cover the gauge shaft with asbestos ribbon to minimize the effect of temperature fluctuations of the room. Each electrical current output of the gauge was directly fed into the static potentiometer, from the reading of which the operating pressure was calculated. The reliability of the pressure gauge readings were occasionally confirmed at the operating temperatures of the extruder. The maximum error of the pressure gauge readings was found to be less than 0.6 kg/cm^2 .

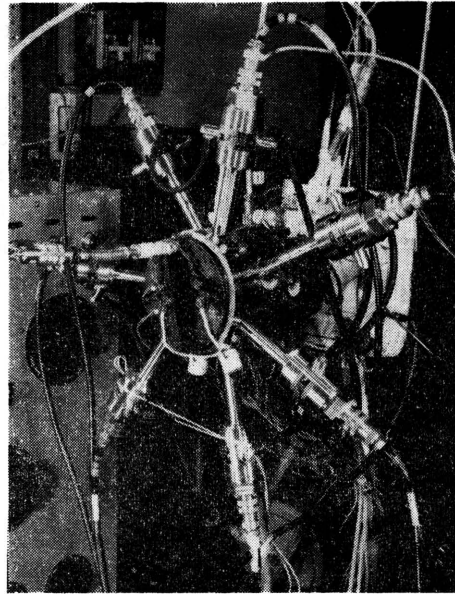


Fig. 2. Overall view of the pressure gauges attached to the extruder die

In carrying out the pressure determination, the temperatures of the breaker-plate, adaptor and die were self-controlled to maintain a prescribed temperature, so as to minimize the errors caused by the possible distributions and fluctuations of the flowing system there. It took about two hours to realize a steady condition. At higher revolution speed of the screw, it was found very difficult to realize the steady flow conditions. Under the conditions, the temperature readings at the die showed smaller variation than 0.5°C throughout the course of the measurements.

Determinations of the shear rate dependence of the tube length correction term coefficient were carried out by the separate experiments with a plunger extrusion type rheometer called Koka Flow Tester²⁾.

III. Experimental results and related theories

The terminology particularly used in this work is shown by the help of Figure 3, the explanatory diagram on the pressure distribution along the tube axis. There, P : total extrusion pressure, p : measured pressure at the wall, z : distance in flow direction, L : tube length, R : tube radius, n_c : Couette's correction term coefficient for the inlet portion²⁾, ν : tube length correction term coefficient, ξ : pressure

gradient supposed for the Newtonian flow, ξ_η : calculated pressure gradient due to the viscosity resistance alone, ξ_{obs} : real observed pressure gradient, ΔP_{Entr} : pressure depression observed at the entrance, ΔP_{Exit} : residual pressure at the tube exit, P_η and P_G : the pressures which are expected to contribute to the inelastic and elastic responses respectively as predicted in the previous paper²⁾. Experimentally, the pressure gradient in the reservoir was very small as compared with that in the tube where the pressure gradient was regarded to be uniform. Hence, as shown by the figure, ν_1 and ν_2 were defined by the intercepts of the total pressure P line and z axis by the pressure gradient line in the tube, respectively. To clarify the above definitions, ξ_N , ξ_η , ξ_{obs} , P_η and P_G are shown by the following formulas, respectively.

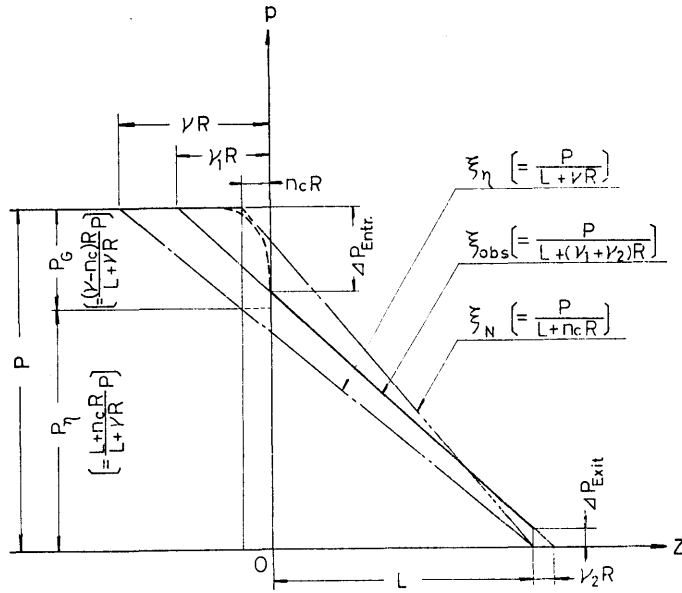


Fig. 3. Explanatory diagram of the pressure distribution along the die tube axis

$$\xi_N = \frac{P}{L + n_c R} \quad (1)$$

$$\xi_\eta = \frac{P}{L + \nu R} \quad (2)$$

$$\xi_{obs} = \frac{P}{L + (\nu_1 + \nu_2) R} \quad (3)$$

$$P_\eta = \frac{L + n_c R}{L + \nu R} P \quad (4)$$

$$P_G = \frac{(\nu - n_c) R}{L + \nu R} P \quad (5)$$

Here, it must be noticed that experimentally, ΔP_{Entr} showed appreciably a smaller value than the so-called entrance loss ΔP_E which was calculated directly from the Bagley plots or by the following formula :

$$\Delta P_E = \frac{\nu R}{L + \nu R} P \tag{6}$$

Experimental results are shown in the following figures.

Figure 4 shows the measured pressure plotted against the length from the tube inlet for Styron 683. In the figure, θ : temperature ($^{\circ}\text{C}$), D_{res} : inner diameter of the cylindrical reservoir, $\dot{\gamma}_R$: apparent shear rate at the tube wall given by the following equation :

$$\dot{\gamma}_R = \frac{4Q}{\pi R^3} \tag{7}$$

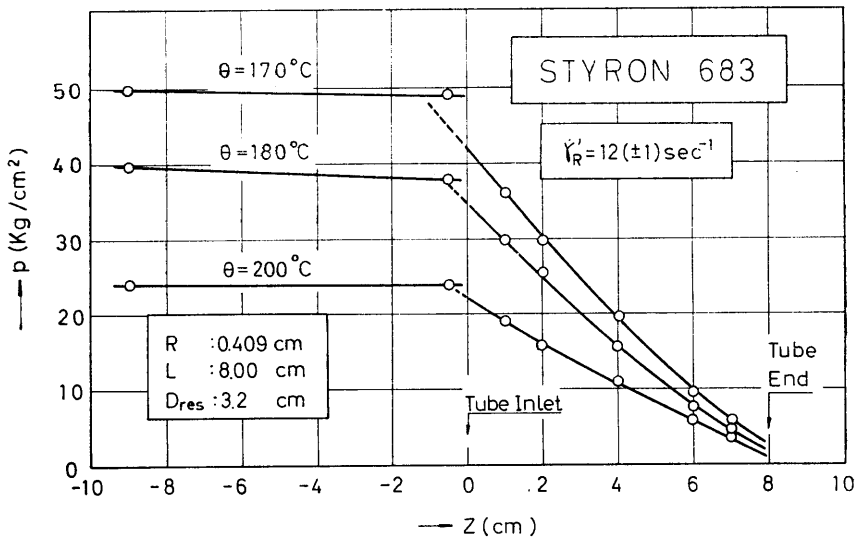


Fig. 4. Measured pressures for Styron 683 plotted against the distance in flow direction at the three temperatures as indicated

where Q is the volumetric flow rate. The gradually decreasing pressure gradient was attributed to the effect of large heat evolution¹⁷⁾ of the flowing melt caused by the high viscosity resistance and to the high temperature dependence of the viscoelasticity in the measured temperature region. For reference, the flow rate of Styron 683 at 180°C under the same testing conditions of nozzle dimensions and extrusion pressure as described in ASTM D 1238-52 T was $2.5 \times 10^{-4} \text{ cm}^3/\text{sec}$, which is compared with the corresponding value of $1.75 \times 10^{-3} \text{ cm}^3/\text{sec}$ for Sholex 6009 at 190°C. Gradually decreasing curves may be also conceivable for the cases where the effect of pressure on viscosity is remarkable. But this might not be the case with Figure 4 since former experiments with high viscosity Newtonian liquid of Novolak

resin¹⁸⁾ indicated that the change of viscosity by pressure fell in the range of experimental precision under the extrusion pressure lower than 80 kg/cm².

Figure 5 shows similar results for Sholex 6009. It was noteworthy that within the experimental precision the pressure gradient remained constant throughout the tube for every shear rate, and the entrance region within the tube where the gradient was expected to decrease was hardly observed. Residual pressures at the tube exit were apparent with a tendency to increase with shear rate. Further verification of these response was carried out with another die of different tube dimensions. The results were shown in Figure 6.

The values of the pressure gradient curves in Figures 5 and 6 were strictly checked with the experimental results obtained by Koka Flow Tester under the basic pre-supposition that regardless of the measuring instrument each viscoelastic liquid of uniform composition must show the same viscoelastic response of a material constant at any constant temperature and shear rate. With the rheometer, the flow rate of Sholex 6009 at 160°C didn't show any practical change by the repetition of test, whereas with the extruder it showed appreciable variation. But the dependence of the viscosity depression of the extrudate upon the screw revolution speed was hardly observed for the present study. Figure 7 shows the change of the tube length correction term coefficient through one course of extrusion at 160°C under the screw revolution speed of 35 rpm. The corresponding change of viscosity curve

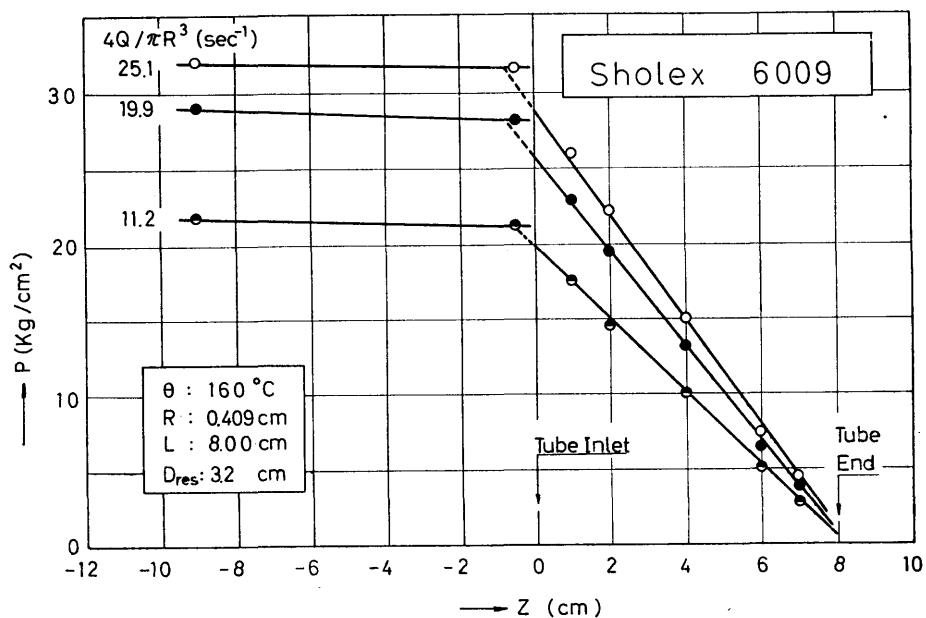


Fig. 5. Measured pressure for Sholex 6009 at 160°C plotted against the distance in flow direction at the three different shear rates, the value of L/R of the tube used being 19.6

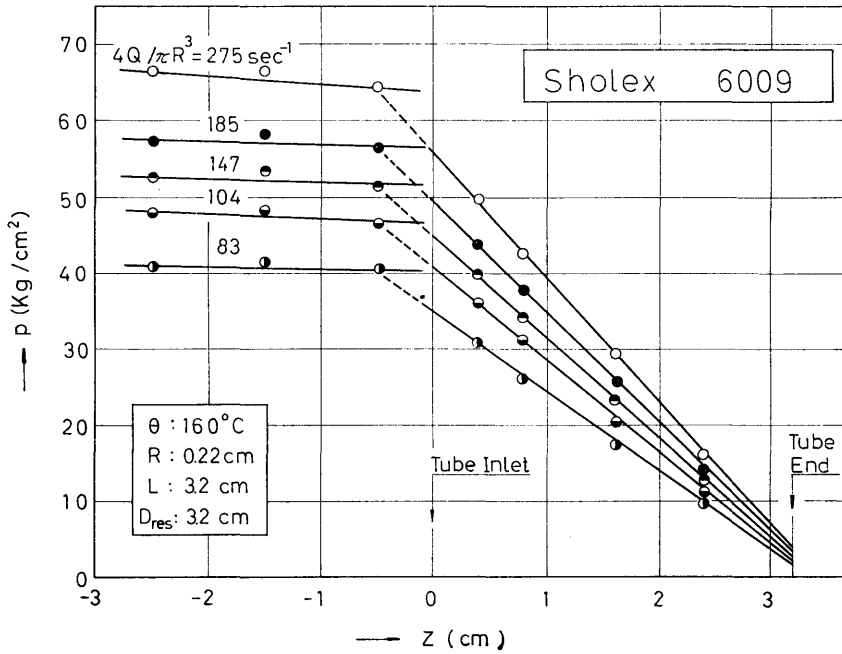


Fig. 6. Measured pressure for Sholex 6009 at 160°C plotted against the distance in flow direction at five different shear rates, the value of L/R of the tube used being 14.5

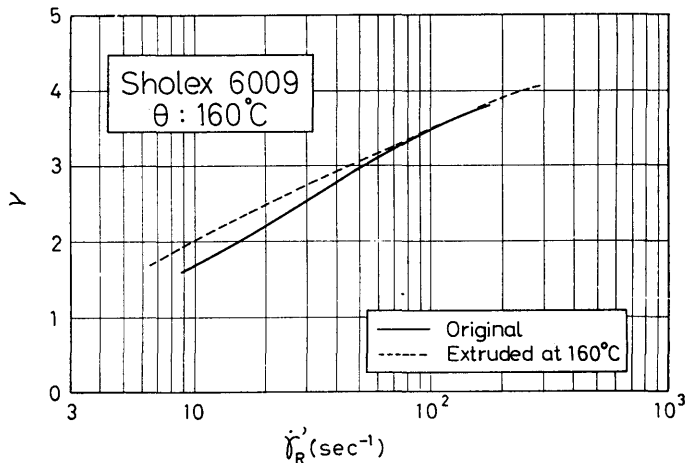


Fig. 7. Comparison of shear rate dependence of ν at 160°C for the original Sholex 6009 with that of the extrudate

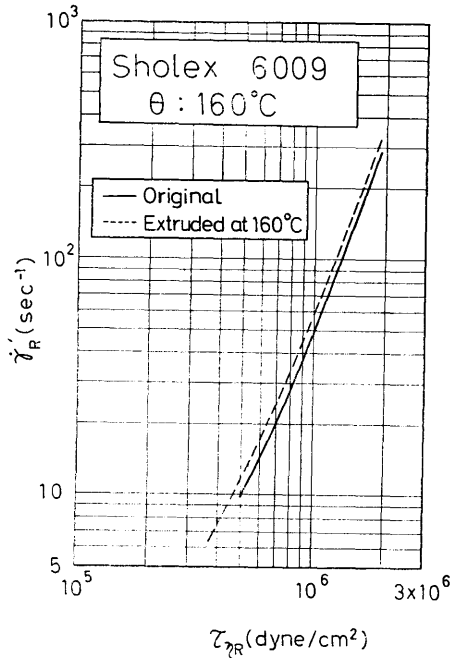


Fig. 8. Comparison of the viscosity curve of the original Sholex 6009 with that of the extrudate

is shown by Figure 8, where τ_{γ_R} is the inelastic shear stress at the wall defined by the following equation :

$$\tau_{\gamma_R} = \frac{PR}{2(L + \nu R)} \quad (8)$$

Comparing Figure 7 with Figure 8, we can notice that for the lower shear rate region the tube length correction term coefficient became larger in despite of the decrease of viscosity. The cause of this phenomenon has not been explained. The depression of viscosity, however, may be attributed to the polymer degradation within the barrel through undergoing high shear stress caused by the screw rotation. Taking these results into consideration, total extrusion pressure P versus $\dot{\gamma}_R$ relation of the extruder for the original test specimen was compared with that of the rehometer obtained with the corresponding extrudate. As a remarkable fact, it showed a perfect agreement as shown by Figure 9. Here, it must be notified that

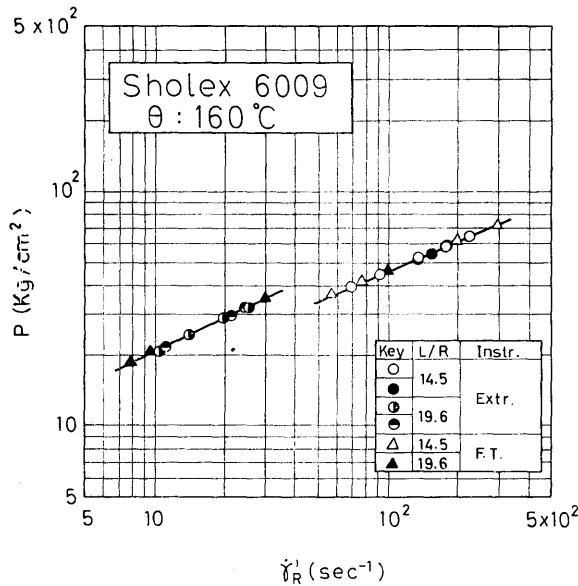


Fig. 9. Comparison of P versus $\dot{\gamma}_R$ relation of the extruder with that of the rheometer

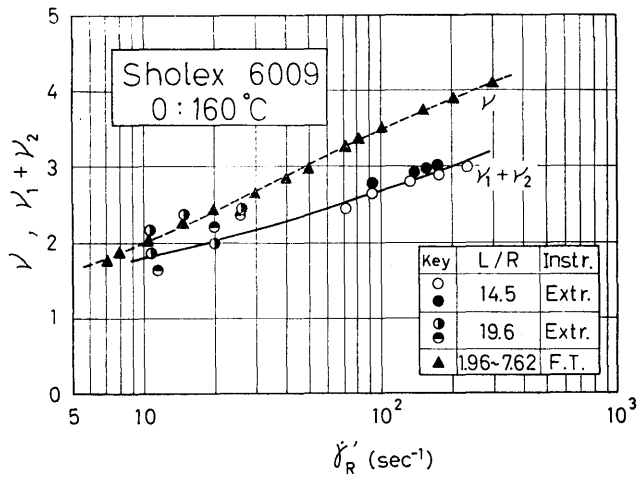


Fig. 10. Comparison of ν with $\nu_1 + \nu_2$ as a function of shear rate

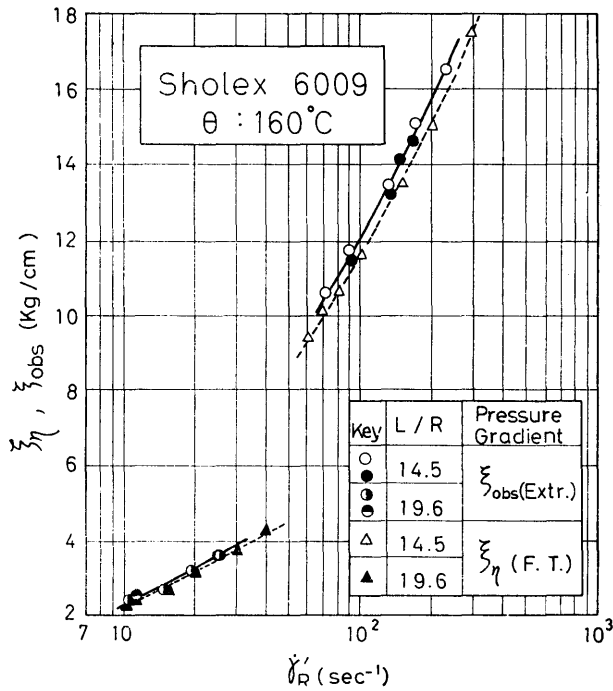


Fig. 11. Comparison of ξ_{η} with ξ_{obs} as a function of shear rate

for each shear rate the measured pressure gradient ξ_{obs} in the tube showed an appreciably larger value than ξ_{η} calculated from the rheometer data by Equation 2. This tendency may be apparent by Figure 10, where the value of $(\nu_1 + \nu_2)$ is compared with that of ν . Figure. 11 shows the direct comparison between ξ_{obs} and ξ_{η} as a function of shear rate. These results may be regarded as an evidence of the really operating shear stress in the laminar steady flow in the tube, because the inelastic shear stress alone can contribute only to ξ_{η} which is found smaller than the observed pressure gradient ξ_{obs} .

By assuming the pressure in the tube to be equal in radial direction, the total shear stress at any radial position may be calculated from the mechanical equilibrium in the tube for the steady isothermal flow condition. At the wall, it is expressed by

$$\tau_{R-obs} = \frac{R}{2} \xi_{obs} \quad (9)$$

where τ'_{R-obs} is the total shear stress at the wall. Figure. 12 shows the comparison of the total shear stress with the inelastic one calculated from the rheometer data. From the standpoint of energy balance, the real elastic shear stress τ_{GR} at the wall must be given by

$$\tau_{GR} = \frac{R}{2} (\xi_{obs} - \xi_{\eta}). \quad (10)$$

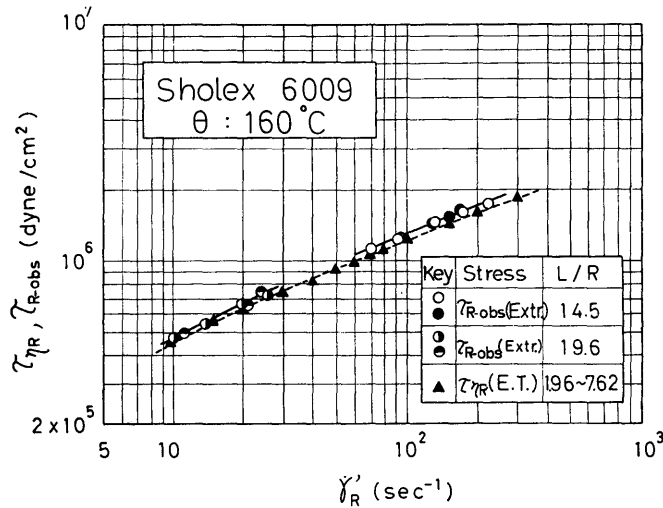


Fig. 12. Comparison of shear rate dependence of τ_{R-obs} with that of τ_{GR}

As pointed out in the previous paper²⁾, the Barus effect index $\bar{\alpha}$ which is the expansion ratio of the cross-sectional area of the extrudate to that of the tube is a simple function of the apparent elastic shear stress at the wall τ'_{GR} defined by¹³⁾

$$\tau'_{GR} = \frac{P_G R}{2(L + n \cdot R)} \quad (11)$$

Although the physical meaning of τ'_{GR} has not been explained, it can be expected that τ'_{GR} must be a unique function of ΔP_{Exit} since we could not find any other particular mechanical energy of the flowing liquid at the exit than ΔP_{Exit} which could cause the large elastic deformation of the Barus effect. Nevertheless, so long as the present study is concerned, the validity of this expectation is not so evident as shown in Figure 13. On the contrary, the sum of ΔP_{Entr} and ΔP_{Exit} showed a good agreement with the value of P_G calculated from the rheometer data.

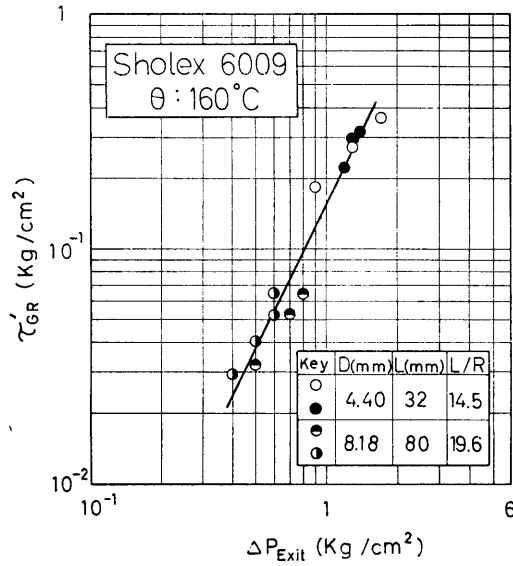


Fig. 13. Variation of τ'_{GR} with ΔP_{Exit}

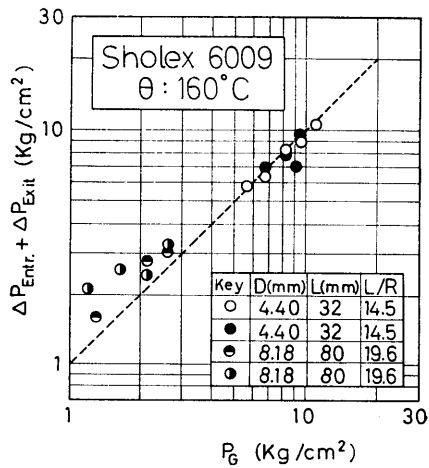


Fig. 14. Relationship between P_G and $\Delta P_{Exit} + \Delta P_{Entr}$

The results are shown by Figure 14. Furthermore, the values of $\bar{\alpha}$ plotted against τ'_{GR} fell fairly well on a single curve regardless of the measuring apparatus as shown in Figure 15.

The foregoing results for Sholex 6009 are summarized numerically by Table 1.

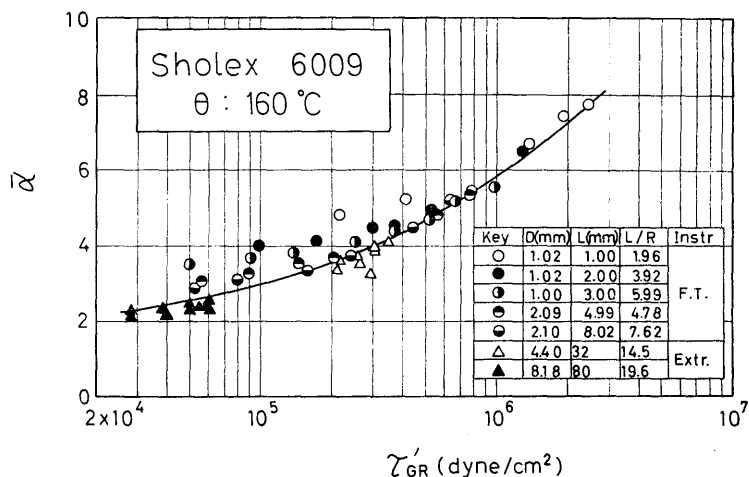


Fig. 15. Variation of $\bar{\alpha}$ for Sholex 6009 with logarithmic τ'_{GR} for the different values of L/R as shown

Table 1. Interrelations between the data on the Barus effect and that on the pressure distribution for Sholex 6009 at 160°C

L/R	$\dot{\gamma}'_R$ (sec ⁻¹)	JP_{Entr} (Kg/cm ²)	JP_{Exit} (Kg/cm ²)	ν_1	ν_2	$\nu_1 + \nu_2$	ν	$\frac{\Delta P_{Entr} + JP_{Exit}}{JP_{Exit}}$ (Kg/cm ²)	P_G (Kg/cm ²)	τ'_{GR} (dyne/cm ²)	$\bar{\alpha}$
14.5	70.8	4.7	0.9	1.99	0.45	2.44	3.27	5.6	5.6	—	—
	92.2	5.1	1.2	2.15	0.49	2.64	3.46	6.3	6.7	2.2	3.5
	133	6.9	1.3	2.31	0.45	2.76	3.70	8.2	8.4	2.7	3.8
	176	7.7	1.4	2.43	0.47	2.90	3.85	9.1	9.6	3.1	4.0
	229	8.8	1.7	2.52	0.45	2.97	3.96	10.5	11.0	3.5	4.1
1.45	93.1	5.6	1.2	2.31	0.47	2.78	3.47	6.8	6.8	2.2	3.4
	131	6.6	1.3	2.24	0.49	2.91	3.66	7.9	8.3	2.6	3.5
	155	7.5	1.3	2.42	0.55	2.97	3.76	8.8	9.1	2.9	3.3
	172	8.0	1.4	2.58	0.42	3.00	3.83	6.4	9.5	3.0	3.9
19.6	10.5	1.7	0.4	1.77	0.47	2.24	2.05	2.1	1.2	2.9	2.3
	14.4	2.0	0.5	1.91	0.49	2.40	2.26	2.5	1.7	4.0	2.4
	19.7	2.0	0.6	1.77	0.49	2.26	2.45	2.6	2.1	5.1	2.5
	25.3	2.7	0.6	1.96	0.49	2.45	2.63	3.3	2.6	6.4	2.6
19.6	11.2	1.1	0.5	1.30	0.37	1.67	2.10	1.6	1.3	3.1	2.2
	19.9	2.1	0.7	1.84	0.39	2.23	2.46	2.8	2.2	5.2	—
	25.1	2.3	0.8	1.96	0.49	2.45	2.62	3.1	2.6	6.3	2.5

IV. Discussion

The elastic shear strain of the flowing viscoelastic liquid so far appeared in literatures may be expressed by the following basic formula for the steady simple laminar flow^{3),5),7),19)-25)}:

$$s_{rec} = \frac{\tau_{\eta}}{G} \quad (12)$$

where s_{rec} is the recoverable shear strain, τ_{η} the inelastic shear stress and G the shear modulus. Including the case where both τ_{η} and G vary as a function of shear rate²⁵⁾, this equation means that s_{rec} is maintained or stored in the flowing liquid under the successive action of τ_{η} , which is regarded to contribute conclusively to the inelastic shear deformation in the process of its determination. Really, the apparent non-Newtonian viscosity is given in round tube rheometry by the quotient of $\tau_{\eta R}$ of Equation 8 divided by $\dot{\gamma}'_R$ of Equation 7 regardless of the magnitude of elastic strain energy of the flowing liquid. To describe these statements more briefly, Equation 12 presupposes that the same shear stress τ_{η} can play concurrently the two distinct roles of the inelastic and elastic stresses. This standpoint, however, might prove valid only under the contradictory assumption that the work done by τ_{η} does not dissipates as heat energy or s_{rec} does not play any role of mechanical energy but is treated as if it were an internal energy. Here, it must be emphasized that only the mechanical energy might be able to cause the Barus effect of large elastic deformation of the flowing melt, whereas the inelastic stress or the internal energy transformed from its work might not. The observed pressure gradient appreciably larger than the one calculated from the viscosity resistance alone, may be the experimental evidence of the really operating mechanical elastic shear stress which could not be defined by the inelastic shear stress alone but varies with the dimensions of the containers of the flowing system²⁾. The elastic strain energy of the flowing liquid could not be free from the wall restriction of the containers. In conclusion, Equation 12, which is incompatible with the authors' standpoint, may be basically difficult to accept from the view-point of energy balance.

As to the normal stresses, it may be reasonable to suppose that tensile stresses operate tangentially to the tube axis, since the elastic shear stresses alone as defined in the previous paper²⁾ could only pull back the extrudate at the centre and extend at the surface substantially without accompanying the post extrusion swelling, while the tensile stresses may be responsible for the swelling as suggested by Merrington²³⁾. Furthermore, when the tensile stresses are assumed to be equal along length of the tube, the above-mentioned supposition on the tensile stress holds well for the observed pressure gradient lines of the present investigations. Because the possible tensile stress distribution in radial direction might not give any effect on the pressure gradient line within the tube, although it may cause the operation of a resultant normal stress upon the tube wall to elevate the static pressure along the length of

the tube by a constant value of the observed residual pressure of ΔP_{Exit} at the tube end.

Recent experiments on the flow birefringence of polymer melt²⁷⁾ have already given the informations on the change of elastic strain with the axial length. But judging from the pressure gradient curves, the change may be said to be negligible for the present research conditions.

V. Conclusions

From the experimental results reported herein, the following conclusions may be derived.

(1) The viscoelastic response measured with the extruder showed a complete agreement with that observed with the rheometer.

(2) The pressure gradient in the tube remained constant throughout the tube for all the testing conditions where the shear rate dependence of the Barus effect was obvious. From this result, we can again conclude that even in these viscoelastic flow conditions the velocity profiles are fully developed on entrance to the tube and the time-dependent relaxation of the elastic strain in the flowing melt does not take place within the course of steady flow in the tube.

(3) The fact that the pressure gradient showed appreciably larger value than that calculated from the Bagley plots could be the evidence of the mechanically operating elastic shear stress in the steady laminar flow.

(4) The residual pressure at the tube exit was apparently observed with a tendency to become larger with shear rate. This pressure was expected to show the pressure energy to cause the Barus effect of the flowing melt when emerged from the restriction of the containers.

The third conclusion described here suggests the modification of the definition of non-Newtonian viscosity²⁸⁾ so far expressed by the total shear stress divided by the shear rate. To explain the interrelations between the residual pressure at the tube exit and the tube length to radius ratio in terms of the shear rate at the wall may be a most important but novel problem on mechanics, which is left undissolved in the present paper.

Acknowledgement

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Literatures

- 1) E. B. Bagley: J. Appl. Phys., 28, 624 (1957).
- 2) T. Arai, H. Aoyama; Trans. Soc. Rheology, 7, 333 (1963).
- 3) W. Philippoff, F. H. Gaskins: Trans. Soc. Rheology, 2, 263 (1958).

- 4) E. B. Bagley, A. B. Metzner: *Ind. Eng. Chem.*, **51**, 714 (1959).
- 5) A. B. Metzner, W. T. Houhton, R. A. Sailor, J. L. White: *Trans. Soc. Rheology*, **5**, 133 (1961).
- 6) T. Arai, I. Suzuki, M. Hatta, K. Miyaguchi: Paper presented at 17th Annual Meeting of the Chemical Soc. Japan, April, 1964.
- 7) A. G. Fredrickson: "Principles and Applications of Rheology", Chapters 7 and 9 (Prentice-Hall, Englewood Cliffs, N. J.) 1964.
- 8) B. C. Sakiadis: *A. I. Ch. E. Journal*, **8**, No. 3, 317 (1962).
- 9) R. Eswaran, H. Janeschitz-Kriegl, J. Schijf: *Rheologica Acta*, **Bd. 3**, Heft 2, 83 (1963).
- 10) H. Janeschitz-Kriegl: "Proceeding of the 4th International Congress on Rheology", Part 1, pp. 143—160 (Interscience, New York) 1965.
- 11) T. Arai, H. Aoyama: *Chem. High Polymers, Japan (Kobunshi Kagaku)* **18**, 53 (1961).
- 12) J. M. McKelvey: "Polymer Processing", pp. 85—97 (John Wiley and Sons, New York) 1962.
- 13) R. C. Kowalski: "Elastic Behavior in Molten Viscoelastic Polymeric Materials", doctoral thesis, Polytechnic Institute of Brooklyn, New York) 1963.
- 14) T. Arai: *High Polymers, Japan (Kobunshi)*, **12**, 352 (1963).
- 15) R. Houwink and W. G. Burgers: "Elasticity, plasticity and Structure of Matter", p. 203 (Cambridge Univ. Press, New York) 1937.
- 16) J. P. Tordella: *J. Appl. Phys.*, **27**, 454 (1956).
- 17) R. C. Gee, J. B. Lyons: *Ind. Eng. Chem.*, **49**, 956 (1957).
- 18) T. Arai, N. Suzuki: Paper presented at the 9th Annual Meeting of Chemical Soc., Japan, April, 1956.
- 19) K. Weissenberg: *Proc. 1st Interna Congr. Rheology*, pp. 1—29 (North Holland, Amsterdam) 1949.
- 20) M. Mooney: *J. Colloid Sci.*, **6**, 96 (1951).
- 21) M. Mooney: *J. Appl. Phys.*, **24**, 675 (1953).
- 22) W. Philippoff: *Trans. Soc. Rheology*, **1**, 95 (1957).
- 23) T. Kotaka, M. Kurata, M. Tamura: *J. Appl. Phys.*, **30**, 1705 (1959).
- 24) P. U. A. Grossman: *Koll. Zeit.*, **174**, 67 (1961).
- 25) H. Kato: *Bull. JSME*, **8**, 593 (1965).
- 26) A. C. Merrington: *Nature*, **152**, 663 (1943).
- 27) J. P. Tordella: "Unstable Flow of Molten Polymers at High Stress" (Contribution from the Research Division of E. I. du Pont de Nemours and Co., Wilmington, Delaware), private communication.
- 28) J. M. Burgers, G. W. Scott Blair: "Report on the Principles of Rheological Nomenclature", V—28 (North-Holland Pub. Co., Amsterdam) 1949.