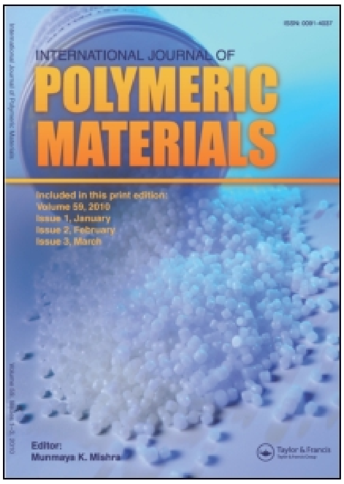

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Melt Viscosity and Elasticity of Low Density and Linear Low Density Polyethylene Blends

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Rheological properties of LDPE/LLDPE blends were determined using a capillary rheometer. The data obtained are presented to describe the effect of blend composition and shear rate on melt viscosity and melt elasticity. Melt viscosity of the blends varies smoothly and proportionately between those of the constituent polymers. Pseudoplasticity of the melt does not reveal large differences compared to those of the constituent polymers. Melt elasticity of the blends increases with LLDPE content, moderately, at low levels of shear and sharply at relatively high levels of shear.

KEY WORDS Melt viscosity, elasticity.

INTRODUCTION

Polyolefin blends have been studied extensively with a view to improving the properties and processability of the homopolymers involved.[1,2] The benefits claimed include, for example, improvements in impact strength,[3–5] environmental stress cracking,[6] optical properties,[7] crystallization rate,[8] low temperature impact strength,[9,10] rheological properties[11,12] and overall mechanical behavior.[1–5,13,14] A further reason for the study of such blends is that mixtures of such polymers often occur in plastics scrap and waste and effect the possible reuse of such low cost material.[15–17]

Linear low density polyethylene (LLDPE) has acquired greater commercial importance because of its superior mechanical behaviour compared to low density polyethylene (LDPE). Blends of LDPE and LLDPE are now regarded as excellent materials for film manufacture because they combine the processability of LDPE and the good mechanical properties and the environmental stress cracking resistance of LLDPE.[18–20] In this paper we report the rheological characteristics of blends of two selected grades of LDPE and LLDPE using a capillary rheometer.

EXPERIMENTAL

Materials

The materials used were commercially available thermoplastics with the following characteristics:

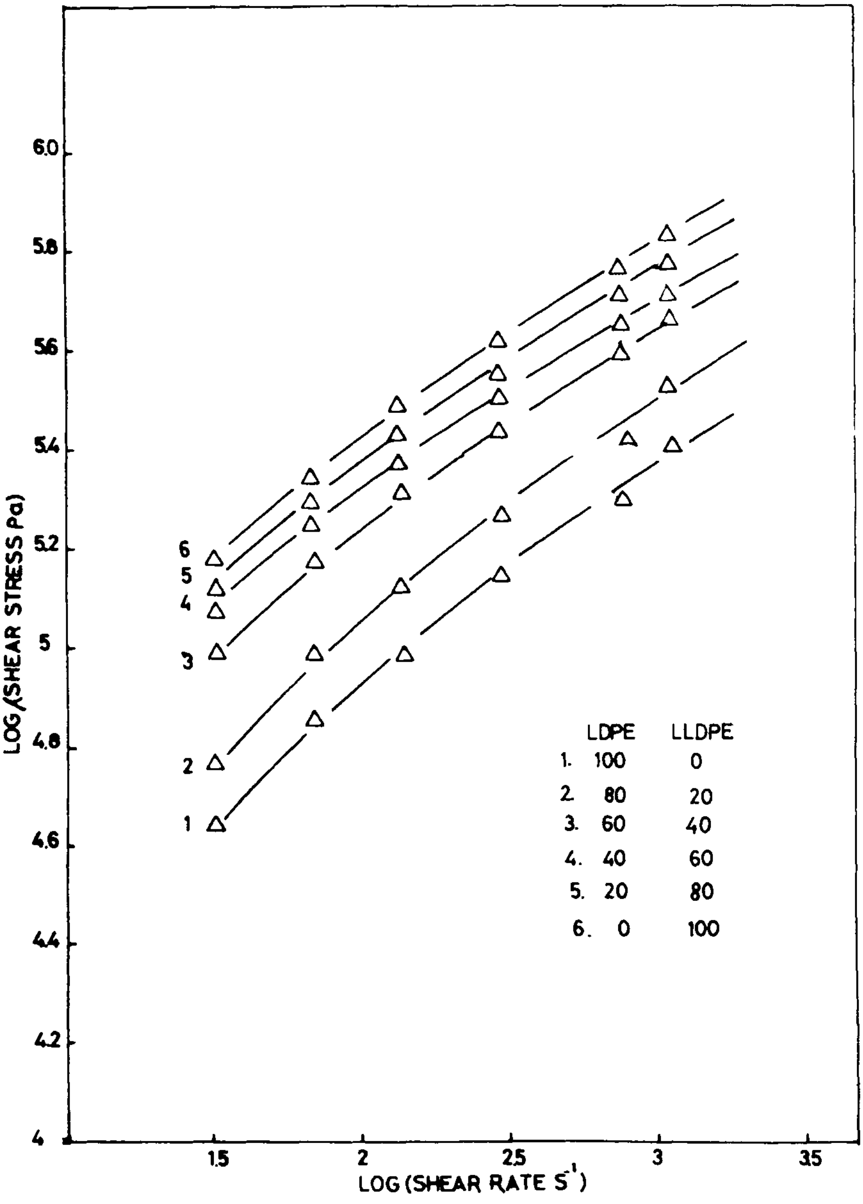

FIGURE 1 Apparent shear stress as a function of apparent shear rate for LDPE/LLDPE blends at 180°C.

TABLE I

Values of power law exponent for LDPE/LLDPE blends

Shear rate (S^{-1})	3×10^2					
LLDPE content (wt %)	0	20	40	60	80	100
n[eq.(3)]	0.44	0.47	0.49	0.42	0.42	0.42
n'[eq.(6)]	0.36	0.38	0.34	0.32	0.32	0.31

Low density polyethylene. Indothene FS 300 was supplied by IPCL, Baroda, density (g/cm^3) = 0.922, melt flow index (g/10 min) = 6.

Linear low density polyethylene. Ladene 218 W was supplied by IPCL, Baroda, density (g/cm^3) = 0.918, melt flow index (g/10 min) = 2.

Blend Preparation

The blends were prepared by melt mixing in a Brabender plasticorder model PL 3S at 160°C and 30 rpm for about 20 minutes. The pure polymers were also subjected to the same procedure in order to make the data comparable to those of the blends. The investigated compositions were 0, 20, 40, 60, 80 and 100 weight per cent respectively of LLDPE.

Rheological Investigation

A complete rheological investigation was performed on each blend with the aid of a capillary rheometer (Gottfert Viscotester model 1500) over a wide range of shear rates (25 to 1500 S^{-1}) at 180°C. Three different capillaries were used in order to evaluate the entrance and exit effects. The diameter was always 1 mm and the length to diameter ratios were 10, 20 and 30 mm.

Extrudate samples, cooled to room temperature, about 5 cm long were cut for diameter measurements which were made by means of a micrometer after solidification of the samples.

The swelling ratio, B, was in all cases evaluated as,[21]

$$B = \frac{D_e}{D} (\rho v_m)^{1/3}$$

where D_e is the diameter of the extruded samples, D the die diameter, ρ is the density of the polymer at room temperature, and v_m the specific volume at the extrusion temperature.

The surface of the collected samples was observed visually in order to evaluate the critical parameters, ie., the shear rate at which the extrudates appear irregular.

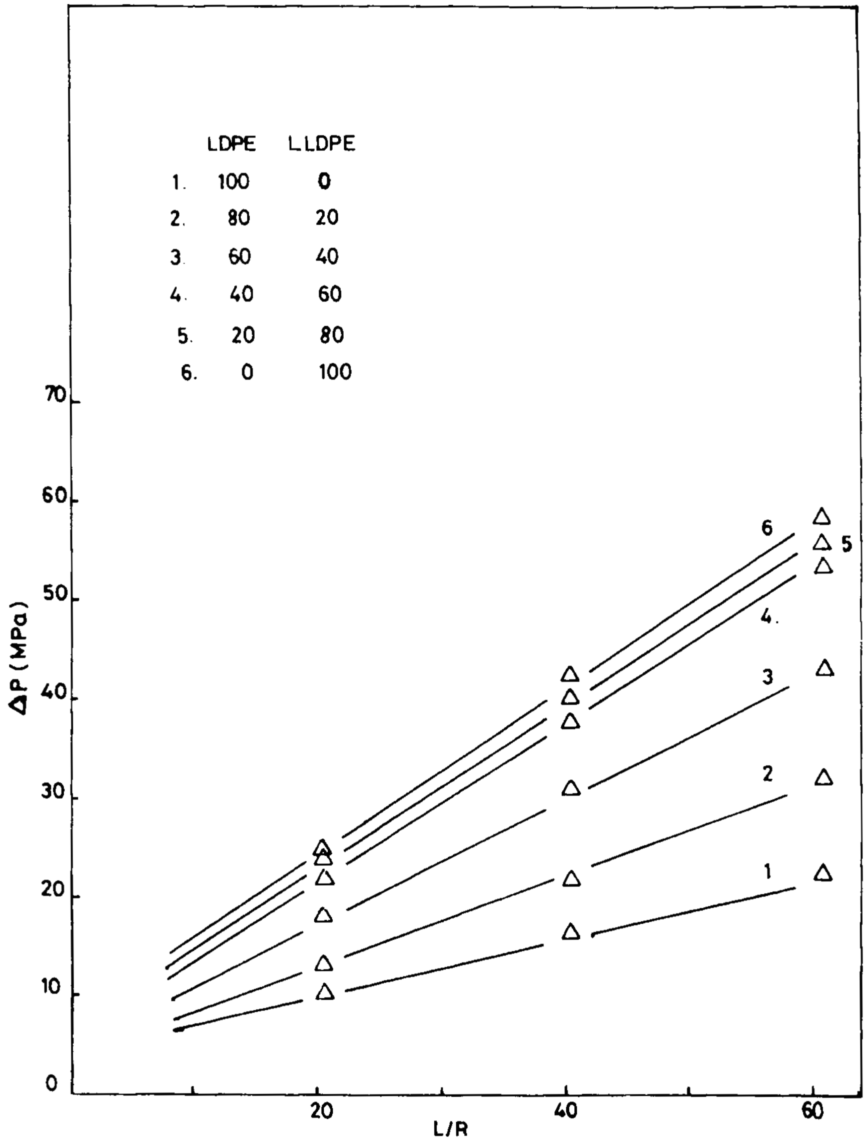

FIGURE 2 Variation of pressure drop ΔP with L/R ratio of the capillary for LDPE/LLDPE blends at a fixed shear rate (10^2 S $^{-1}$).

RESULTS AND DISCUSSION

Flow Curves

From volumetric flow rates at various applied pressures, values of apparent shear stress at wall, $\tau_{w_{app}}$, and apparent shear rate $\dot{\nu}_{w_{app}}$ were calculated using the following expressions.[22]

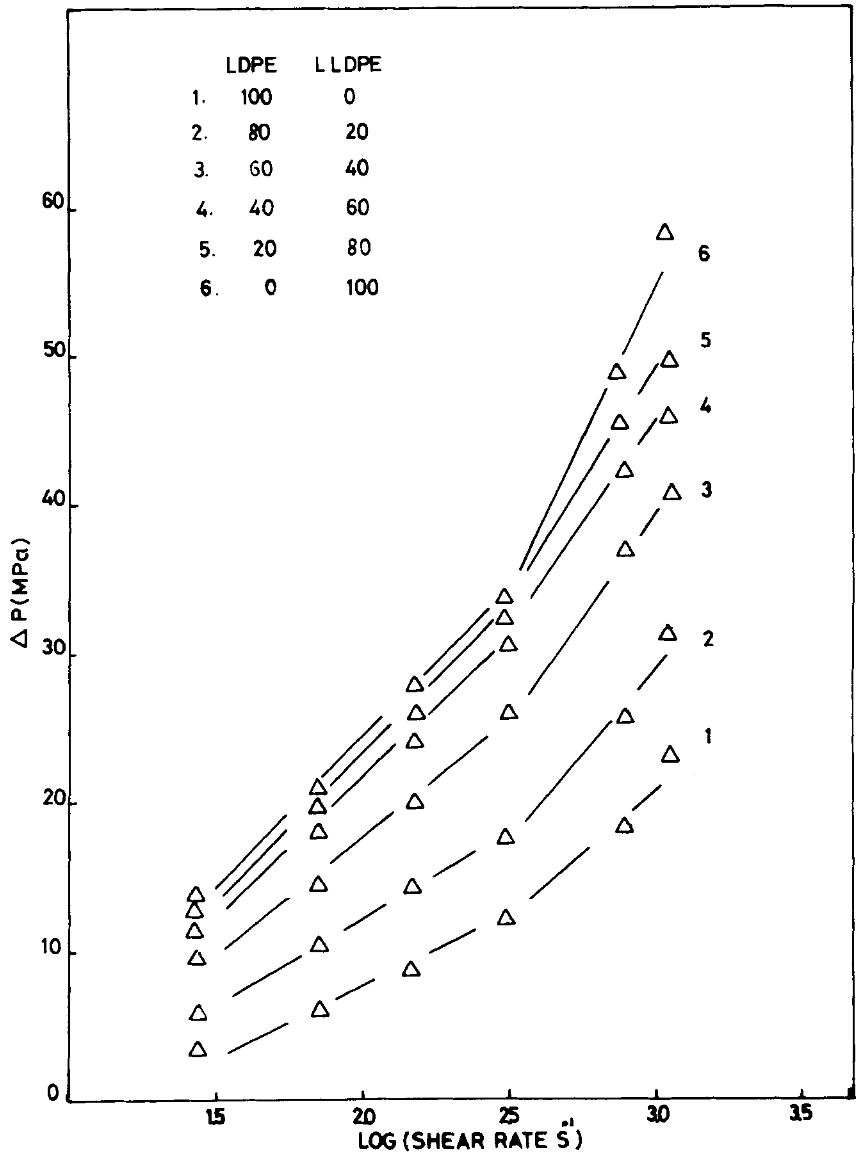

FIGURE 3 Variation of pressure drop ΔP with shear rate at 180°C (L/R = 60) for LDPE/LLDPE blends.

$$\tau_{w_{app}} = \frac{\Delta P}{2.L/R} \tag{1}$$

$$\dot{\nu}_{w_{app}} = \frac{4Q}{\pi R^3} \tag{2}$$

where ΔP is pressure difference between the entrance and exit regions of the

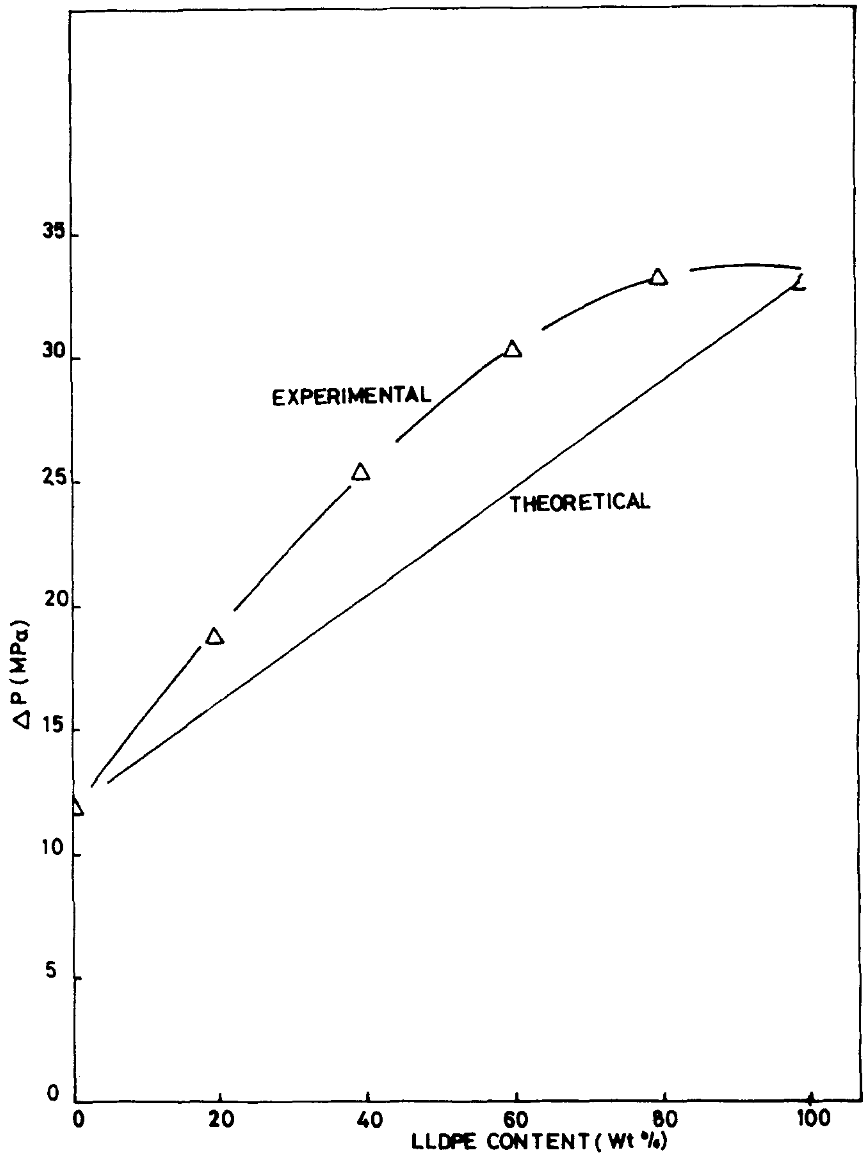

FIGURE 4 Variation of pressure drop ΔP with blend composition of LDPE/LLDPE blend at 180°C, ($L/R = 60$) and shear rate $10^2\ S^{-1}$.

capillary die, Q is the volumetric flow rate, and L and R are respectively the length and radius of the die.

From the log $\tau_{w_{app}}$ vs. log $\dot{\nu}_{w_{app}}$ plots (shown in Figure 1) values of power law exponent 'n' were calculated in accordance with the following power law:

$$\tau_{w_{app}} = K\dot{\nu}^{n}_{w_{app}} \tag{3}$$

where K is a constant. Values of n shown in Table I, are much smaller than unity,

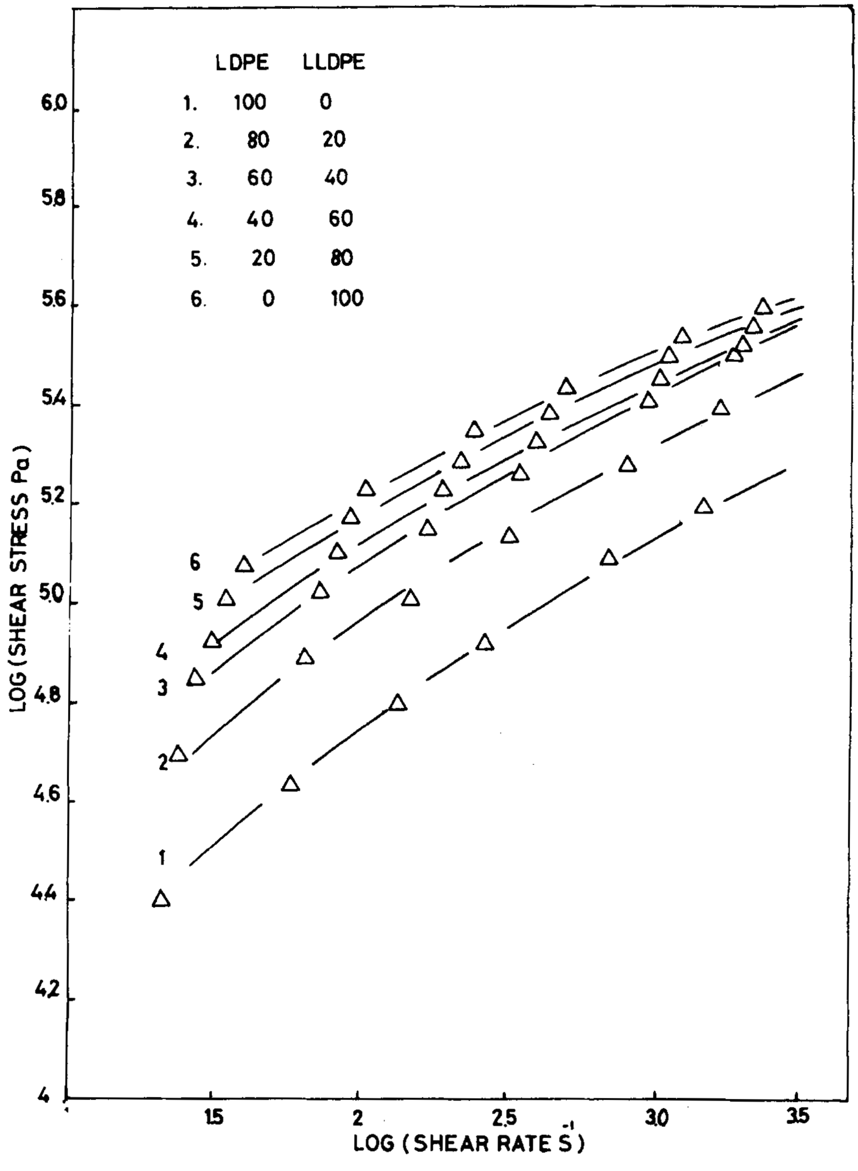

FIGURE 5 Flow curves in terms of shear stress as a function of shear rate of LDPE/LLDPE blends at 180°C.

implying a strong pseudo-plastic character of the melts of these binary blends. It can also be seen that the flow behavior of the blends does not reflect large differences from those of the individual components. Rabinowitsch correction was applied using values of n determined from Equation (3), to obtain true shear rate $\dot{\nu}_w$ according to the following expression.[22]

$$\dot{\nu}_w = [(3n + 1)/4n]\dot{\nu}_{w_{app}} \tag{4}$$

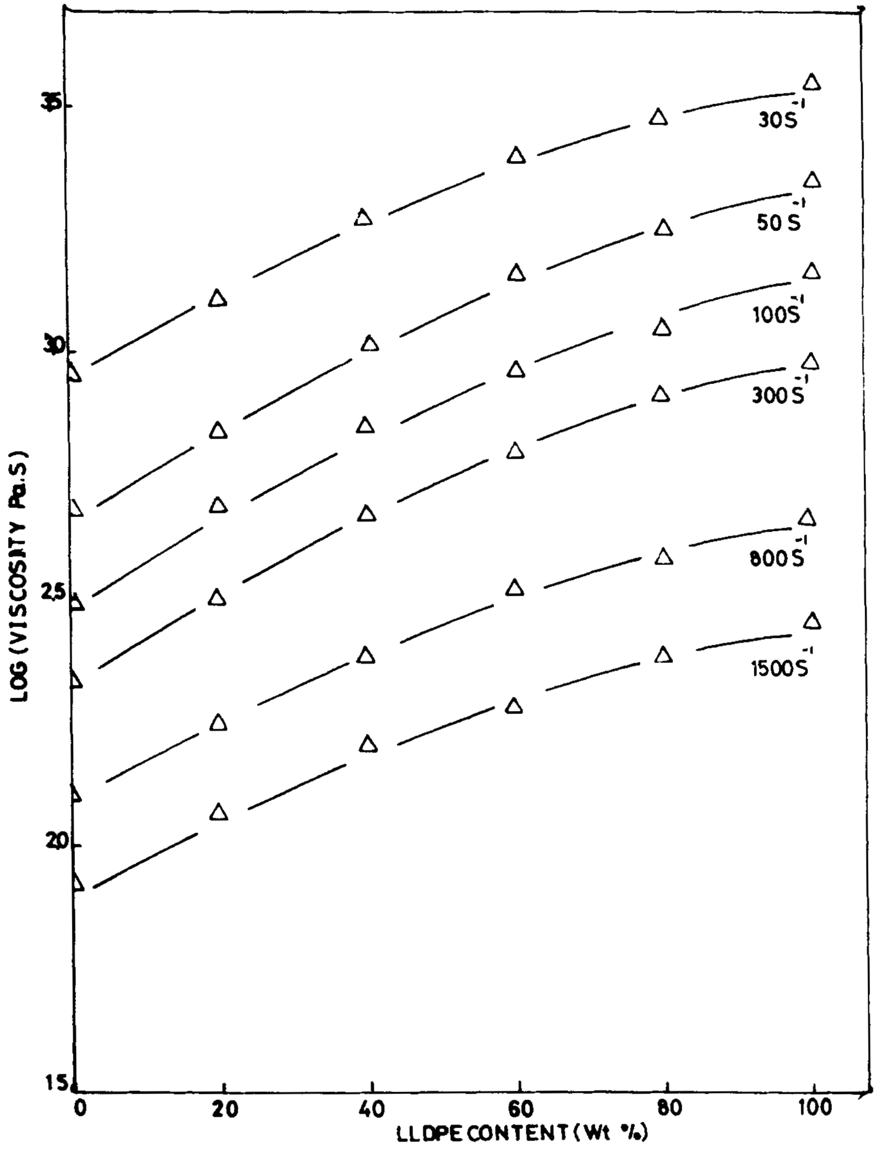

FIGURE 6 Variation of melt viscosity with blend composition at 180°C for LDPE/LLDPE blends.

Similarly, true shear stress at wall τ_w was calculated by applying Bagley correction according to the following expression.[22]

$$\tau_w = \frac{\Delta P - P_c}{2.L/R} \tag{5}$$

where the Bagley correction factor P_c was evaluated from the flow data recorded with three different dies of L/R varying from 20 to 60. Bagley plots, ΔP as a function of L/R for LDPE/LLDPE blend at various compositions, are quite linear

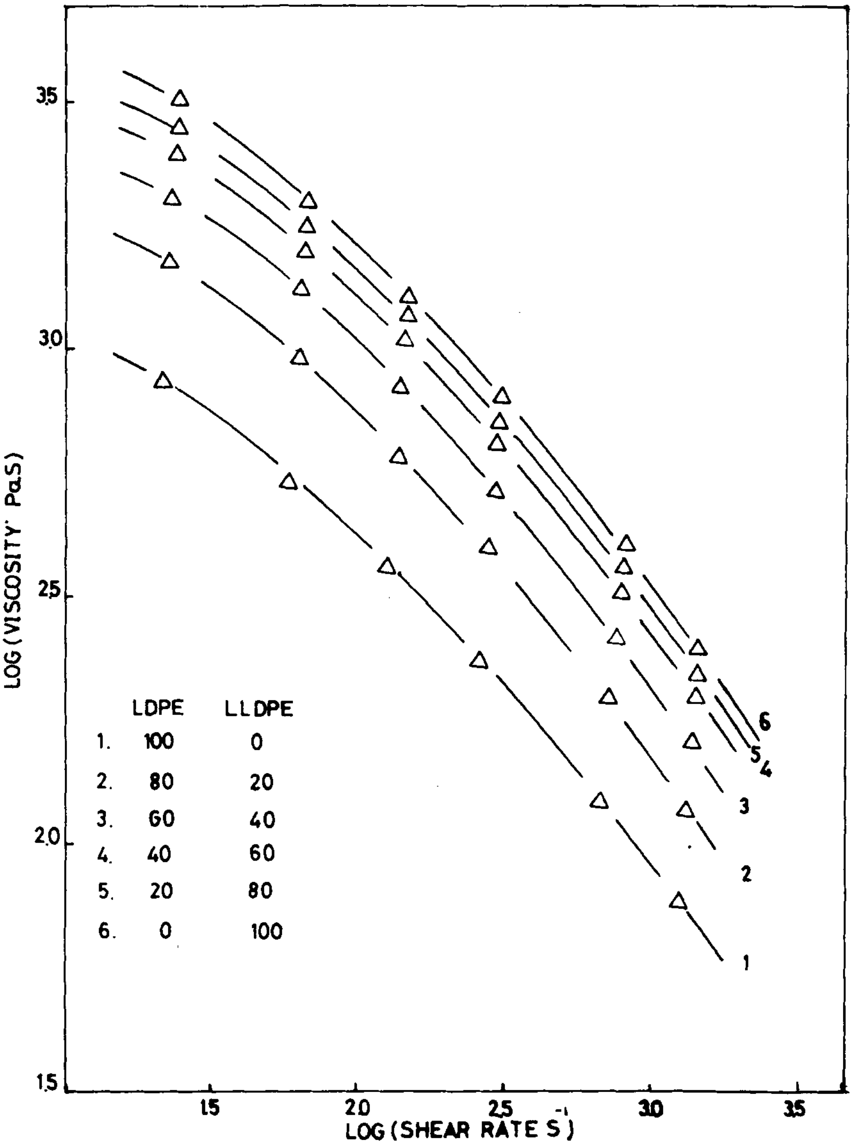

FIGURE 7 Variation of melt viscosity with shear rate at 180°C for LDPE/LLDPE blends.

as shown in Figure 2. This linearity of Bagley plot confirms the absence of slippage at the capillary wall and at the interface boundaries where the slippage is believed[23] to cause curvature in Bagley plot.

Variation of ΔP with log $\dot{v}_w$, shown in Figure 3 is quite linear over the entire range of measurements for the blend at various compositions as well as the blend components. The slope of the lines increases gradually with increase in LLDPE content, at relatively low rates of shear and rapidly at relatively high rates of shear. Increase of slope implies greater difficulty of flow or higher viscosity, due to the occurrence of elongational flow. The observed increase of slope with the increase

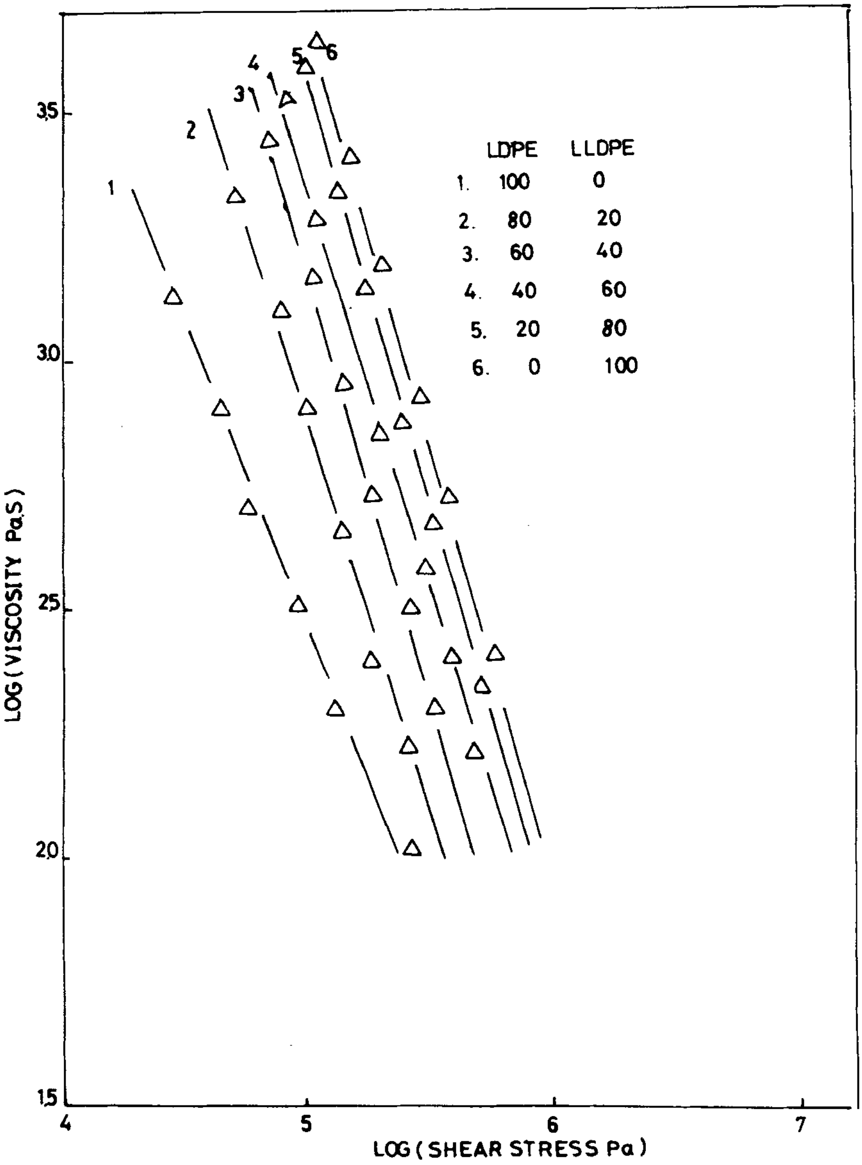

FIGURE 8 Variation of melt viscosity with shear stress at 180°C for LDPE/LLDPE blends.

of shear rate supports this view, since the elongational flow predominates at higher shear rates. The two step variation of slope in the case of the blends might correspond to the onset of elongational flow of the two components (LDPE and LLDPE) of the blend.

Variation of ΔP as a function of blend composition at a constant shear rate is shown in Figure 4. The value of ΔP increases gradually with LLDPE content.

Flow curves in terms of corrected values of shear stress and shear rate are presented in Figure 5, for the LDPE/LLDPE blend for various blend compositions.

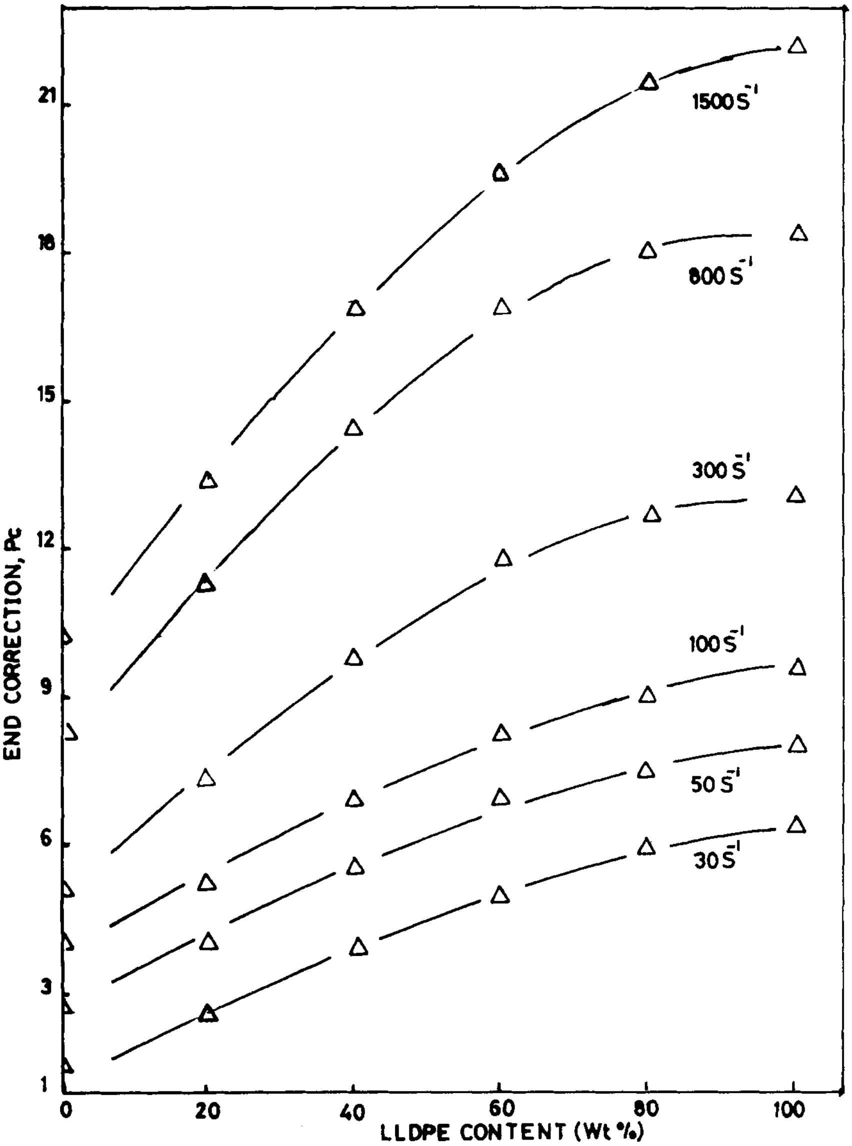

FIGURE 9 Variation of Bagley correction factor P_c as a function of blend composition at various shear rates.

Data on pure LDPE and LLDPE resins are also included in Figure 5 to illustrate the behavior of the blend in comparison with its two components. Viscosity of LLDPE is higher than that of LDPE. Flow curves for the blend at various compositions lie in between the flow curves of LDPE and LLDPE.

Variation of melt viscosity with blend composition at various shear rates (Figure 6) shows a 'positive deviation' with respect to linear extrapolation between LDPE and LLDPE extremes. This indicates stronger interface interaction in accordance with Utracki's[24] definition of a positive deviation blend.

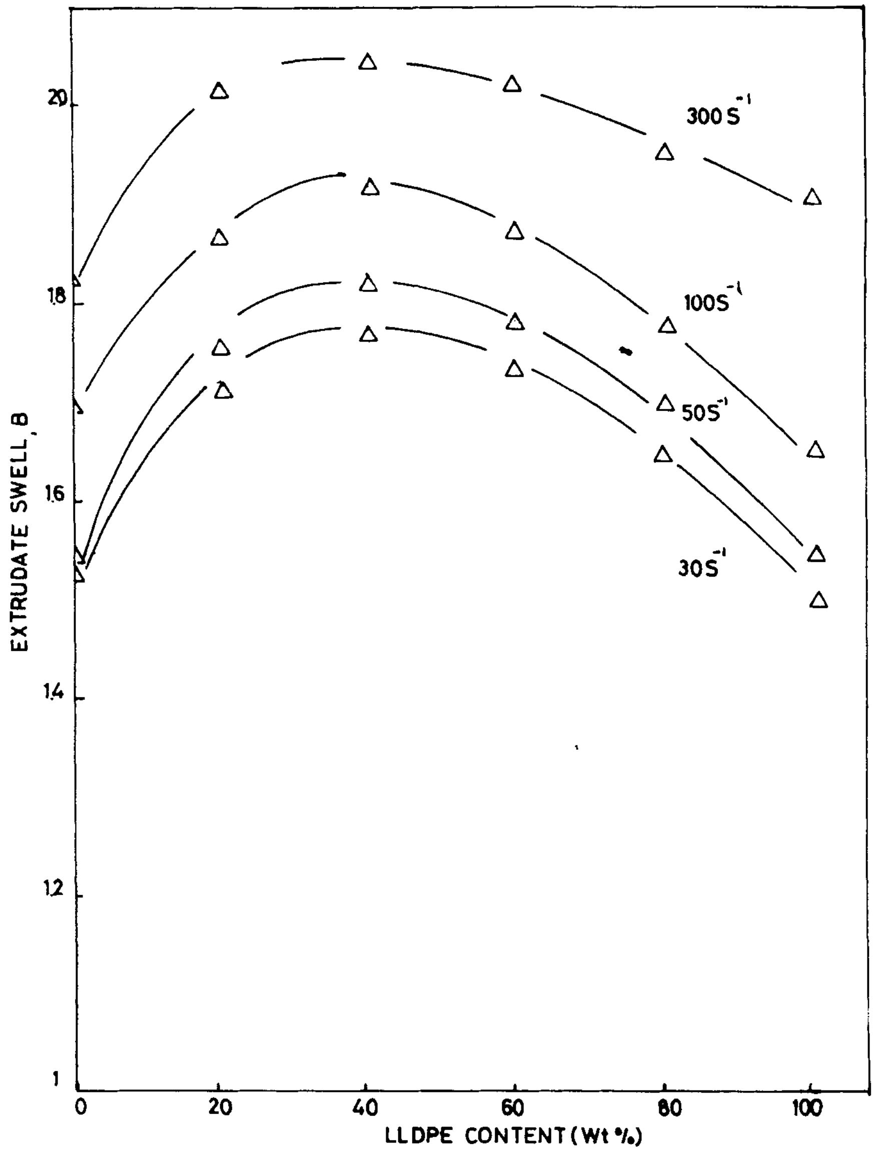

FIGURE 10 Variation of extrudate swell (*B*) with blend composition of LDPE/LLDPE blends at 180°C, at various shear rates.

The power law fitting these flow curves (Figure 5) may be written as follows:

$$\tau_w = K' \dot{\gamma}_w^{n'} \qquad (6)$$

where K' and n' are used to distinguish them from K and n of Equation (3). These corrected values of power law exponent n' are slightly smaller than the values of n corresponding to Equation (3). Furthermore the flow behavior of blends does not reflect large differences in the viscosity of the individual components.

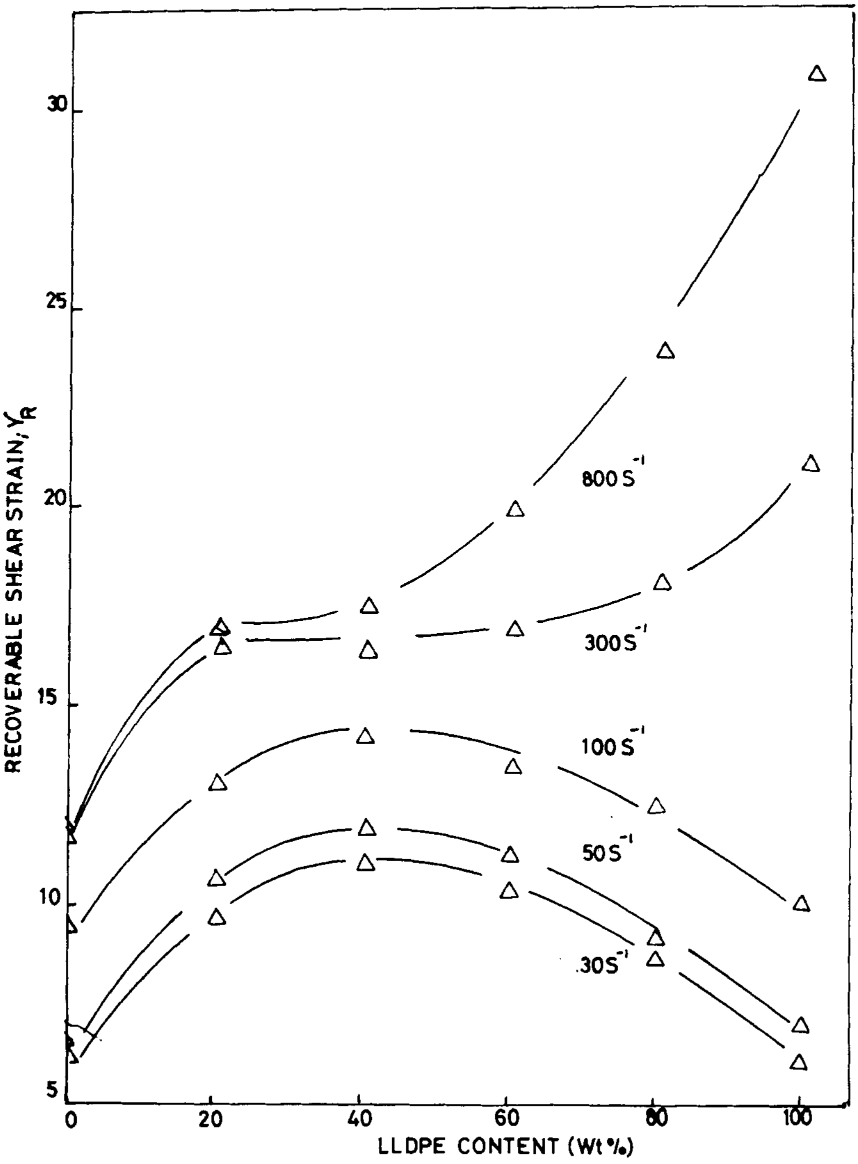

FIGURE 11 Variation of recoverable shear strain with blend composition of LDPE/LLDPE blends at 180°C, at various shear rates.

Melt Viscosity

Melt viscosity data of the blend at various compositions are presented in Figures 7 and 8 as the variations of melt viscosity ($\eta = T_w/\dot{v}_w$) with shear rate and shear stress. At any given shear rate or shear stress, melt viscosity is lowest for LDPE and increases smoothly with LLDPE content. Decrease of melt viscosity with increasing shear rate and shear stress is linear for all the compositions of the blend. These linear variations are consistent with the following power law relationships:

$$\eta = A\dot{\nu}_w^a \tag{7}$$

$$\tau = B\dot{\nu}_w^b \tag{8}$$

Elastic Parameters

Bagley Correction Factor P_c. Variation of Bagley correction factor P_c, as a function of blend composition at various shear rates is shown in Figure 9. P_c value increases with increasing shear rates at all compositions of the blend. At relatively low levels of shear, P_c value increases gradually with increase in LLDPE content and rapidly at relatively high shears. This effect may be attributed to the fact that at high shear rates, the normal stresses built up in the polymer by flow exceed the shear stresses by a larger order of magnitude and cause losses of pressure at the inlets and outlets of the capillaries and in extreme cases melt fracture and other mostly undesirable flow phenomena.[25]

Melt Elasticity

Elasticity of the melt results in expansion of the polymer fluid on its exit from the die. The extrudate swell ratio D_e/D, where D_e and D are diameters of the extrudate and the die, respectively, is a direct measure of melt elasticity. As a function of blend composition, extrudate swell (B) is shown in Figure 10. All the blends show larger B values than those of the constituent polymers. Bogue and White[26,27] suggested use of the parameter recoverable shear strain, ν_R for describing and distinguishing the fluid elasticity of different viscoelastic fluids as a function of shear stress. ν_R was calculated from the expression,[22]

$$\nu_R = 2[(D_e/D)^6 - 2]^{1/2} \tag{9}$$

As shown in Figure 11 ν_R increases with LLDPE content and then decreases at relatively low levels of shear. At high levels of shear (8×10^2 S^{-1}) ν_R increases rapidly with increase in LLDPE. Pure LLDPE has the highest value of ν_R which decreases substantially on blending with LDPE. The reduction of melt elasticity on blending with LDPE is a useful gain in property relevant to processing, since extrudate distortion tendency reduces with decreasing melt elasticity[22,28–30] thus enabling the processing at higher shear rates without loss of surface smoothness of the product.

CONCLUSIONS

1. Melt viscosity of LDPE/LLDPE blends shows positive deviation from the log additivity rule indicating strong interface interaction resulting in sufficient compatibility in the melt.
2. The pseudo-plasticity of the blends does not show large differences from those of the individual components.
3. Melt elasticity of LDPE/LLDPE blends increases with LLDPE content moderately, at low levels of shear and sharply at relatively high levels of shear.

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