Rheological Behaviour of the Crosslinkable Polyethylene and EPDM Rubber with the Help of a Torque Rheometry

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INTRODUCTION

It is very much unusual to find a single polymer is being used without being blended with other polymers. The rheological parameters of the blends dictate the processibility of the blend in the operating conditions. Rheology of the polymer blend differ widely from the single components depending on the type and phase composition. Coran[1,2] suggested that in the plastic/rubber blends the shear modulus depend on the viscosity of the rubber phase and crystallization of the plastic phase. According to Folt[3] the flow behaviour of the blend depend on the heat history and the morphology.

Considering the wide spread application of the crosslinkable polyethylene in cable industries and low loss elastomer like EPDM the rheological behaviour of four blends were studied with the help of Brabender Plasti-corder[4,5] at four shear rates and at three temperatures. Frictional heat development and the flow activation energies were also studied.

EXPERIMENTALS

Blending was done in a Brabender Plasti-corder (PLE-330) having 80 cc capacity as per formulations given in Table I. Crosslinked polyethylene used was of XLPE-11 variety of ACCI Ltd., India and EPDM was of Intolan-255 variety of International Synthetic Rubber Inc. The mixer temperature was first adjusted by circulation of oil. The crosslinked polyethylene was first put into the mixing chamber and allowed to melt at low shear rate, then the EPDM is added slowly. After the complete addition of EPDM the ram is closed and immediately the chart recorder was started. The blending was allowed to continue for 8 minutes. Then the ram is opened and the blend was taken out and sheeted in a two roll mill. The mixture temperature was adjusted to 100°C, 110°C and 120°C. At a particular mixer temperature, the blending was done at four different rotor speeds viz. 100 rpm, 80 rpm, 60 rpm and 40 rpm. In each case the chart gave the variation of mixing torque and stock temperature with the blending time. Equilibrium torque was taken at the 7th minute of the blending in each case. Activation energies of flow at a particular shear rate was determined from the knowledge of variation of the torque with the stock temperature as observed from the plastograph. n' and K' values for four blends were determined from log–log plot of torque (Nm) and rotor speed (rpm).

TABLE I

Blend formulations

Blend nos.	A	B	C	D
Crosslinked polyethylene (XLPE-11)	85%	70%	50%	30%
EPDM	15%	30%	50%	70%

RESULTS AND DISCUSSION

Mixing studies

The variation of the mixing torques with the mixing time for the four blends, were represented graphically in a plastogram (Figure
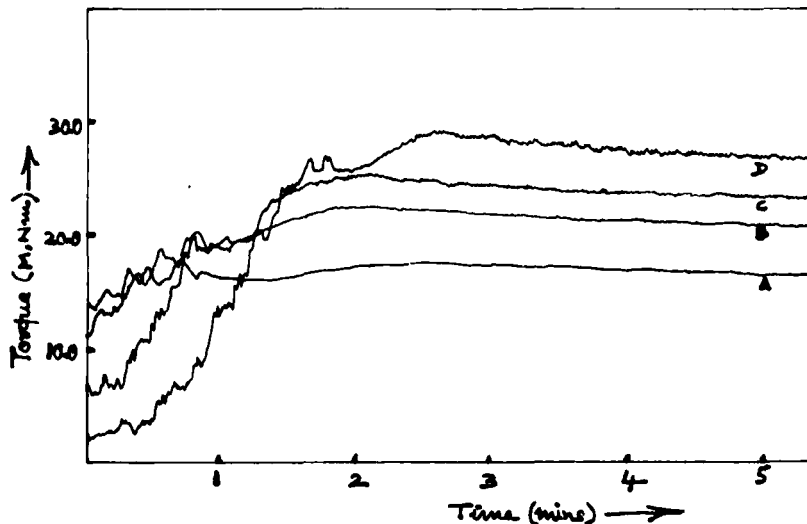
FIGURE 1   Variation of mixing torque with the time of mixing.

1). The mixing was done at 110°C and at 80 rpm. From the plastogram considerable amount of processing parameters can be obtained. As observed from Figure 1 the mixing torque increases rapidly within 1.0–1.5 min of mixing attains a maximum and then decreases slightly to a stationary level on prolonged mixing. The maximum torque attained is more as the XLPE is replaced by EPDM and consequently more mixing time is required to attain it. The rate of rise in the torque is more as the XLPE/EPDM ratio decreases and the difference between the initial and the maximum torque is higher for high EPDM content.

After about 2.5 min of mixing the mixing torque reaches the equilibrium stage. The equilibrium torque is higher as the XLPE is being replaced by the EPDM. The extent of decrease in the mixing torque from the maximum torque to the equilibrium torque is more or less independent of the blend composition. The plastogram as in Figure 1 suggest more or less identical processibility for all the blends studied. However slightly rough and wider band of the blend D suggest comparatively poor processibility may be due to the higher elastic character of the blend because of high EPDM content. A smooth and uniform band for higher XLPE/EPDM ratio

probably reflect the good blending and better processing characteristics.

Rheological studies

The rheological interpretation of the torque rheometry measurements are based on the equation for the non-newtonian flow behaviour as described earlier[5-8]

$$M = C_n K' S^{n'} \tag{1}$$

where M is the mixing torque and S is the rotor speed, n' and K' are the parameter of power law equation. C_n is the instrumental constant. Eq. (1) can be rewritten as[5]

$$M = K_n S^{n'} \tag{2}$$

The blending was done at three temperatures and at four rotor speeds viz. 100 rpm, 80 rpm, 60 rpm and 40 rpm. The equilibrium torque was determined for all the shear rates at a particular temperature of blending. The blending temperatures considered were 100°C, 110°C and 120°C. A representative example of the variation of stress with the shear rate were shown graphically in Figure 2 as log–log plot of M vs S. The plots are found to follow a
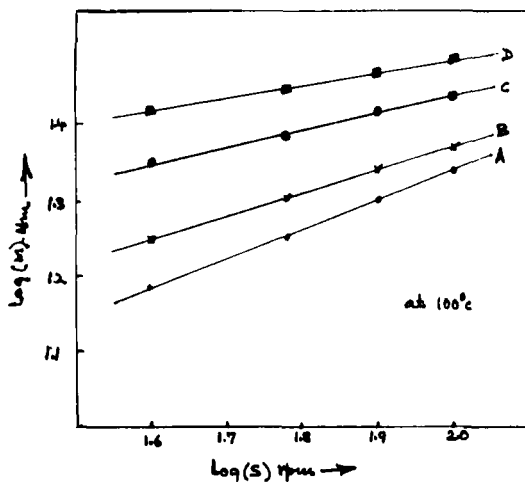

FIGURE 2   Variation of equilibrium torque with the rotor speed.

TABLE II

Rheological parameters (n' and C_nK')

Blend nos.	Mixer temperature (°C)	n'	K_n
A	100	0.47	2.85
B	100	0.36	5.46
C	100	0.24	10.70
D	100	0.195	13.70
A	110	0.52	2.16
B	110	0.41	4.31
C	110	0.30	8.19
D	110	0.235	11.30
A	120	0.56	1.31
B	120	0.49	2.89
C	120	0.38	5.85
D	120	0.27	9.16

straight line path for all the blends the slope of which determine the values of non-newtonian index 'n'''. The K_n values were determined by regression analysis and tabulated in Table II. Although the blends containing curative the maximum temperature considered is quite safe for processing.

As the EPDM content becomes more in the blends with XLPE the non-newtonian index decreases. This trend remains the same for all the temperature of blending studied. The increase in blending temperature increases the n' values. The value of non-newtonian index n' dictates the pseudoplastic flow behaviour of the blend system. Higher n' value for high XLPE/EPDM ratio is due to less pseudoplastic nature of XLPE. K_n is being the measure of consistancy index increases with the decreasing XLPE/EPDM ratio. The temperature of blending decreases the K_n values.

The increase in the rotor speed is always accompanied by the increase in the torque developed during blending for all the blend system. But the rate of rise in the torque with the shear rates depend on the blend ratio. At a particular blending temperature the rate of rise in torque with rotor speed decreases as the XLPE/EPDM ratio decreases. At a particular shear rate the mixing torque increases as the EPDM content increases in the blend, may be due to increasing frictional forces exerted during mixing. The differences

between the torque values at constant rotor speed with the blend ratio become wider as the temperature of blending increases.

Effect of mixing temperature on the stock temperature of the blend

The rotation of the rotors during mixing, put the mechanical energy into the mass of the material in the mixing chamber and this mechanical energy transforms into frictional heat energy and thereby increasing the stock temperature more than the mixer temperature. The extent of rise in the stock temperature depend on the viscosity of the material. At various rotor speeds and mixer temperature the equilibrium stock temperature of the various blends were measured with the help of a chart recorder. The variation of the stock temperature and the mixer temperature for all the blends at various shear rates have been shown as a straight line plot in Figures 3, 4, 5 and 6. It is quite evident from the figures that the slope of the straight lines decreases as the XLPE/EPDM ratio
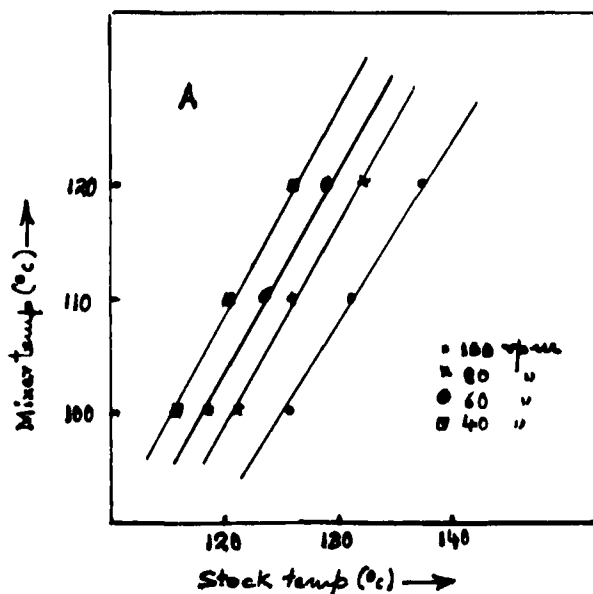

FIGURE 3   Variation of stock temperature with mixer temperature for blend A.
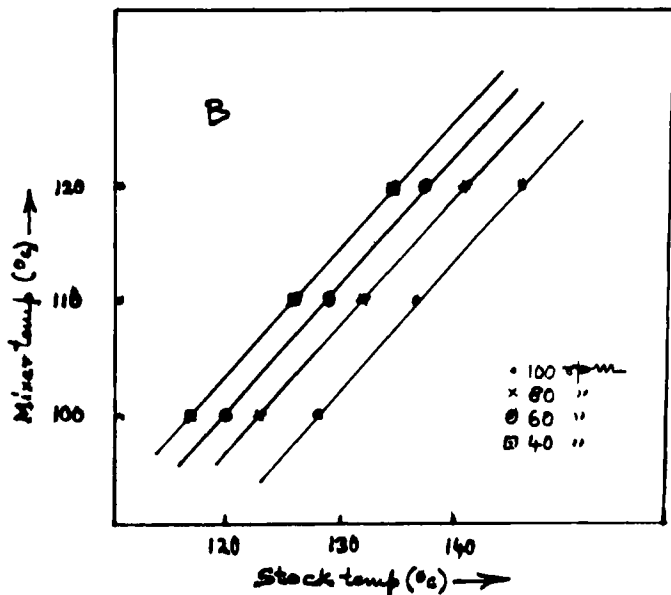
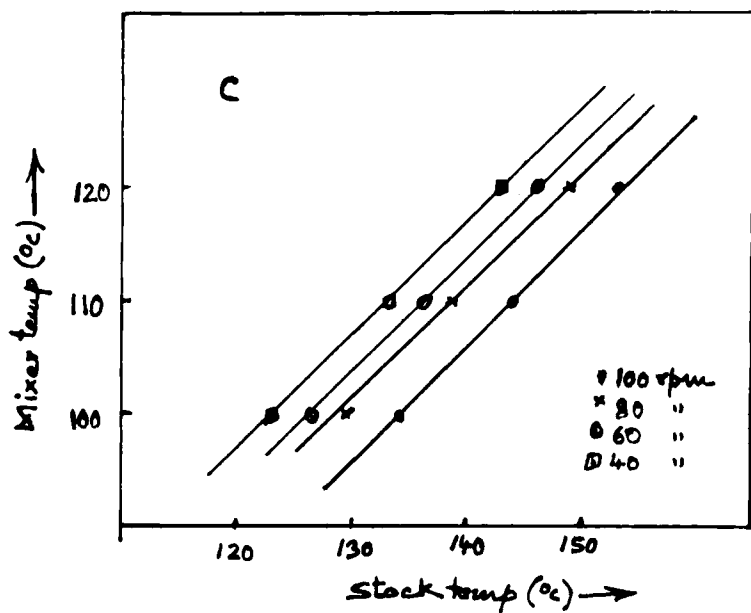FIGURE 4   Variation of stock temperature with mixer temperature for blend B.

FIGURE 5   Variation of stock temperature with mixer temperature of blend C.
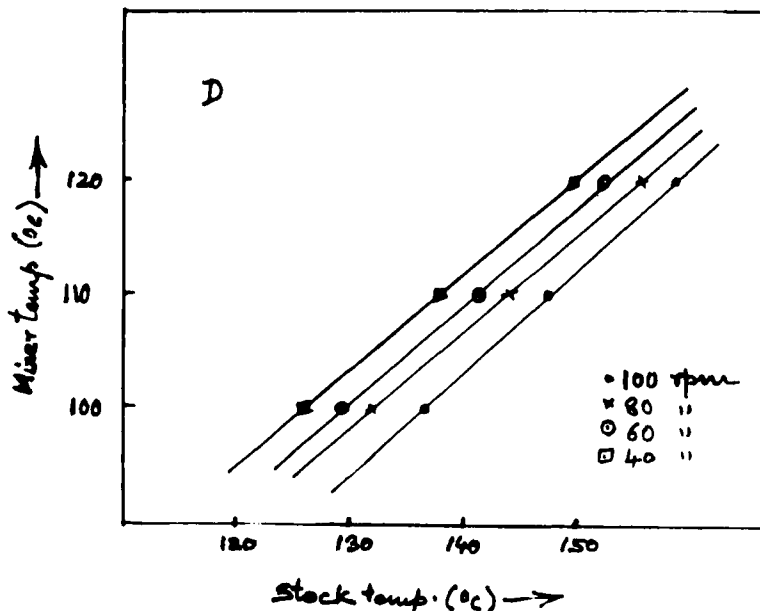
FIGURE 6   Variation of stock temperature with mixer temperature of blend D.

decreases. They are approximately 1.66, 1.15, 1.05 and 0.85 for the blends A, B, C and D respectively. This is due to the more frictional forces experienced by the presence of higher amount of rubber (EPDM) in the blend resulting more frictional heat build up. The rate of rise in stock temperature for a particular blend, is more or less independent of rotor speed. However high shear rate is accompanied by higher stock temperature for a particular mixer temperature.

Flow activation energy

The plot of $\log M$ against T^{-1} (stock temp. °C) shows linearity for all the blends at a particular shear rate. This linear relationship confirms the Arrhenious type behaviour from which the flow activation energy can be found out as a function of rotor speed. A representative plot of $\log M$ vs T^{-1} as a function of rotor speed for
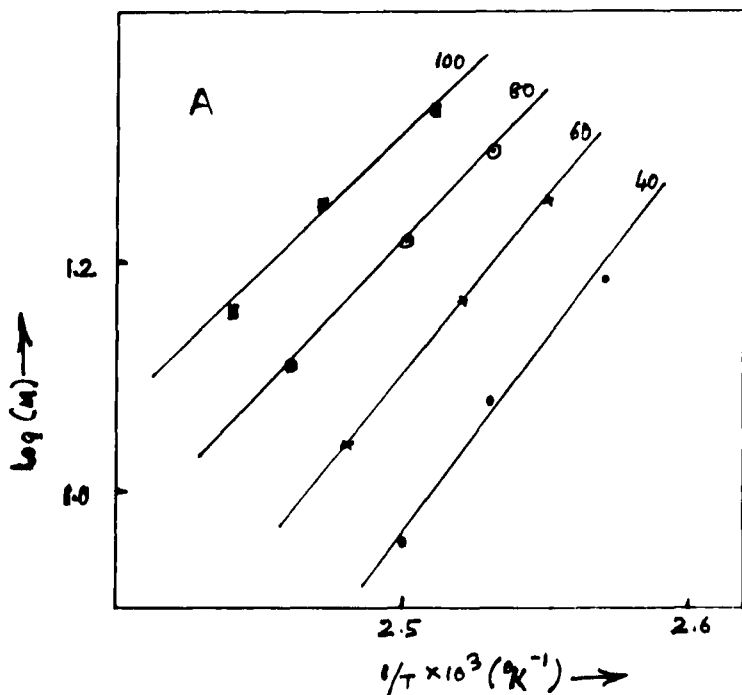
FIGURE 7   Determination of activation energy of flow for blend A.

blend A is shown in Figure 7. Flow activation energies as calculated from the slopes of the straight lines are given in Table III. As observed the activation energy does not change appreciably with shear rate. However slightly higher values are observed for lower rotor speed for blend A and B. Activation energy decreases as the EPDM content increases in the blend up to 50:50 level beyond which it remains more or less constant.

CONCLUSION

Mixing torque increases as the XLPE is replaced by EPDM with simultaneous increase in stock temperature due to increase in frictional force. High shear rate increases the torque and stock

TABLE III

Activation energy of flow at constant shear rate

Blend nos.	Shear rate (rpm)	ΔE_r (K, cal/mole)
A	100	13.4
	80	13.6
	60	14.2
	40	14.0
B	100	5.8
	80	6.0
	60	7.0
	40	7.3
C	100	5.0
	80	4.8
	60	4.5
	40	4.7
D	100	4.9
	80	4.6
	60	4.2
	40	4.4

temperature. Non-newtonian index decreases and consistency index increases with the increase in EPDM in the blend. Increase in mixer temperatures increasing effect on non-newtonian index. Flow activation energy decreases initially but after certain blend ratio it remains more or less constant and independent of shear rate.

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