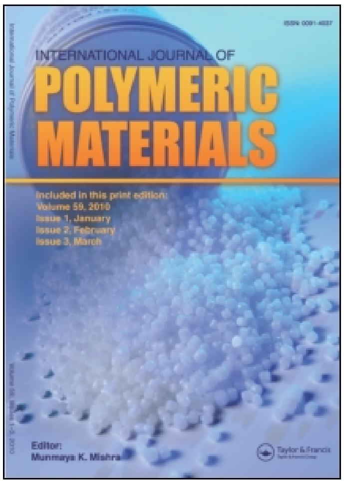

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Effect of Hexadiene and Vinyl Acetate on the Flow Behaviour of EPDM and EVA Blends

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Effect of Hexadiene and Vinyl Acetate on the Flow Behaviour of EPDM and EVA Blends

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Flow behaviour of melt-mixed blends of EPDM and EVA with special reference to the change of their HD and VA content are studied. Viscosity decreases linearly with the increase in shear rate for all the blends which is more prominent in case of low VA content EVA. Low HD shows more pseudoplastic behavior. Stored elastic energy increases as the EPDM/EVA ratio decreases. Relaxation time increases for high HD and VA content. Circumferential wrinkles on the extrudate surface with surface fractures was observed at high shear rate for high HD containing EPDM.

KEY WORDS Flow behaviour, EPDM, EVA, blends

1. INTRODUCTION

Rubber/thermoplastic blends are a type of thermoplastic composite materials that are processable as a 'melt' at an elevated temperature and has been considered as active field in polymer processing research. Viscosity of individual polymer plays a decisive role in the flow and forming of the compounds. Several attempts have been made to predict the complicated rheological behavior of polymer blends in view of its relevance to processing.[1–8] Rheology and morphology of XLPE/EPDM and XLPE/EVA blends[9–11] have been studied following a model developed by Das *et al.*[12,13]

Here, we have studied the flow behavior of polyblends of EPDM and EVA with special reference to the change in HD and VA content. The effect of shearing on melt-viscosity, extrudate swell as well as other rheological parameters related to extrudate morphology have been reported.

2. EXPERIMENTAL

The blend formulations are given in the Table I. EPDMs are of Nordel-1040 and Nordel-1440 variety of Du Pont, UK and EVAs are of Elvax-460 and Levaprene-450 variety of Du Pont, USA and Bayer, Germany respectively. Blending was done in a Brabender plasticorder at 60 rpm and 120°C for plastic EVA and at 40°C for elastomeric EVA. Instron capillary rheometer ($L/D \approx 101$) was used to study the melt flow behavior of the blends. Four shear rates and one temperature were selected to study the rheological parameters. Non-Newtonian Index (n) and consistency index (k) were determined from shear stress and shear rate data by regression analysis.

Extrudate samples were used to determine the swelling ratio (α) with the help of microscope fitted with a micrometer. Viscoelastic parameters like stored elastic energy (W), recoverable deformation (γ_m), shear modulus (G), relaxation time (t_R), modified Weissenburg number (W'_N) were determined as earlier[12,13] as follows:

$$\gamma_m = \sqrt{(1/2C)(\alpha^4 + 2\alpha^{-2} - 3)}$$

where $C = (3n + 1)/4(5n + 1)$ and γ_m is recoverable deformation

$$W = C\gamma_m\tau$$

$$t_R = [1/(\tau(1 - n)/n)][(n.\ k^{1/n})/G(1 - n)]\{\exp[(1 - n)/n] - 1\}$$

Extrudate morphology and melt fracture were studied with the help of M45 WILD Autophotomat. The morphology of the blends was also studied with the help of scanning electron microscopy.

3. RESULTS AND DISCUSSION

3.1. Effect of Blend Ratio, HD and VA Content on Viscosity of the Blends

The variation of apparent viscosity with the shear rates for various blends are shown in Figure 1. Viscosity decreases linearly with the shear rate for all the blends

TABLE I

Blend Formulations

Blend Nos.	A	B	C	D	E	F	G	H
EPDM Rubber (NORDEL-1040)	60	40	60	40	—	—	—	—
EPDM Rubber (NORDEL-1440)	—	—	—	—	60	40	60	40
Ethylene Vinyl Acetate Copolymer (ELVAX-460)	40	60	—	—	40	60	—	—
Ethylene Vinyl Acetate Copolymer (LEVAPRENE-450)	—	—	40	60	—	—	40	60

NORDEL-1040 (1,4 HD ≈ 4%).
NORDEL-1440 (1,4 HD ≈ 6%).
ELVAX-460 (18% VA; MFI = 2.8).
LEVAPRENE-450 (45% VA; ML_{1+4} = 22 ± 2).

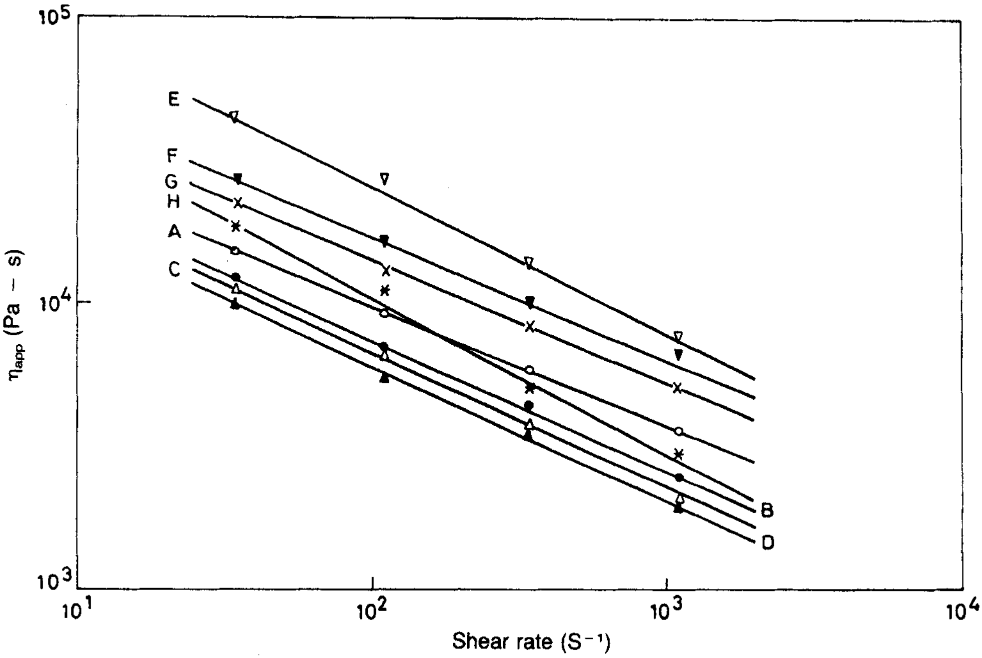

FIGURE 1 Variation of apparent viscosity with shear rate for the blends at 200°C.

studied, the rate of which depends on HD content of EPDM and VA content of EVA as determined from the n-values. As the EPDM-EVA ratio decreases viscosity also decreases for all the blends irrespective of VA content of EVA. However, it is more prominent in case of low VA containing EVA. The effect of EPDM-EVA ratio on the viscosity is not so well marked in case of high VA containing EVA and low HD containing EPDM. For high HD containing EPDM, viscosity is higher than the low HD containing EPDM-EVA blend. At low shear rate viscosity differences are comparatively wide, but tend to converge at high shear rate for low VA containing EVA-EPDM blends suggesting more shear sensitivity of low VA containing EVA. At all shear rates low viscosity differences are observed in case of low HD and high VA containing EPDM-EVA blend. The decrease in viscosity at high shear rate region for EPDM rich blends irrespective of HD content, particularly for low VA containing EVA may be due to breaking up elastomeric EPDM microgels at high shear rate region. However, this may not be a predominant factor in case of high VA containing EVA/EPDM blend.

3.2. Effect of HD and VA Content on the Rheological Parameters

The rheological parameters (n and k) are shown in Table II and Table III. The non-newtonian index (n) increases generally as the EVA dosage increases more in the blend irrespective of HD content of EPDM and VA content of EVA. 'n' values also suggest the more pseudoplastic behavior of low HD containing EPDM at fixed level of VA content. The consistency index (k) decreases with the increase in EVA content in the blend, however, it decreases more drastically in case of low VA

TABLE II

Values of Non-Newtonian Index (n), Consistency Index (k), γ_w, W, G and t_R for the Blends at 200°C

Blend Nos.	$\gamma_w (s^{-1})$	$W(10^5\ J/m^3)$	$G(10^5\ J/m^3)$	t_R(s)	n	$k \times 10^{-4}$
A	42.95	0.038	1.79	0.128	0.48	9.549
	143.22	0.051	5.25	0.020		
	429.54	0.166	4.43	0.013		
	1432.22	0.759	2.71	0.013		
B	41.6	0.008	3.59	0.101	0.52	6.309
	138.7	0.024	4.40	0.015		
	416.0	0.153	2.61	0.009		
	1387.0	0.557	2.41	0.009		
C	41.29	0.041	0.657	0.258	0.53	5.75
	137.6	0.068	1.349	0.073		
	412.9	0.245	1.212	0.048		
	1376.0	0.358	2.85	0.012		
D	40.43	0.021	0.960	0.161	0.56	4.79
	134.8	0.045	1.49	0.065		
	404.3	0.152	1.66	0.035		
	1348.3	0.401	2.12	0.017		

TABLE III

Values of Non-Newtonian Index (n), Consistency Index (k), γ_w, W, G and t_R for the Blends at 200°C

Blend No.	$\gamma_w (s^{-1})$	$W(10^5\ J/m^3)$	$G(10^5\ J/m^3)$	t_R(s)	n	$k \times 10^{-4}$
E	40.71	0.494	0.738	0.537	0.55	18.2
	135.75	0.861	1.55	0.036		
	407.1	1.380	3.84	0.008		
	1357.5	0.847	22.58	0.008		
F	39.91	0.271	0.523	0.444	0.58	10.5
	133.1	0.521	1.15	0.007		
	399.1	0.819	2.93	0.024		
	1331.0	1.023	10.44	0.007		
G	39.67	0.156	0.614	0.461	0.59	8.71
	132.27	0.310	1.22	0.145		
	396.72	0.423	3.24	0.0034		
	1322.79	0.116	49.60	0.0012		
H	38.97	0.122	0.369	0.611	0.62	6.02
	129.96	0.239	0.786	0.755		
	389.7	0.323	2.18	0.044		
	1299.6	0.083	35.31	0.001		

containing EVA which is once again more prominent in case of high HD containing EPDM. Non-newtonian index values of the blends suggest their pseudoplastic behavior. However, pseudoplasticity is more for low VA containing EVA at fixed level of HD in EPDM.

A representative example of die-swell of the blends is shown in Figure 2 as a plot of swelling ratio against shear rate. A sharp decreasing trend in swelling with the shear rate beyond the shear rate of $10^2\ s^{-1}$ is observed for all the blends except for low HD containing EPDM and low VA containing EVA blends where it increases sharply. The effect of blend ratio on the die-swell is more pronounced in

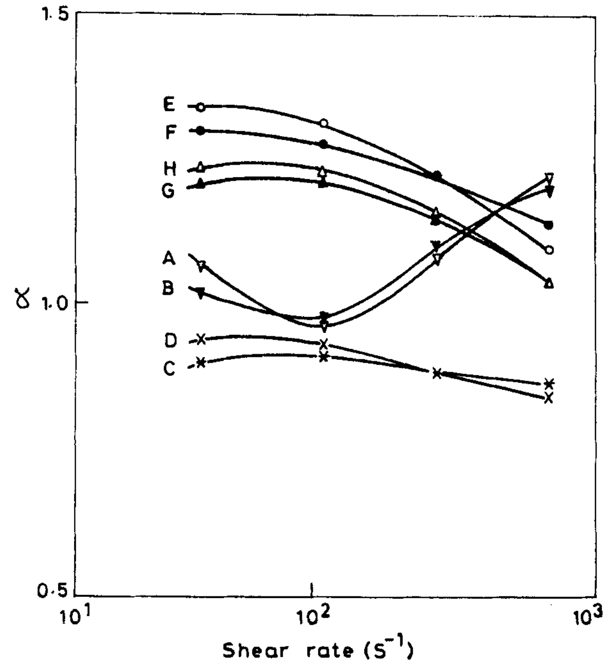

FIGURE 2 Variation of extrudate swell with shear rate at 200°C.

case of high HD containing EPDM irrespective of VA content of EVA. At low shear rate region, high EPDM/EVA ratio is accompanied by higher swelling when blended with low VA containing EVA. However, reverse trend is observed in case of high VA containing EVA blends. At fixed level of HD content high VA containing EVA gives rise to lower swelling.

Other calculated rheological parameters at 200°C are given in Table II and Table III.

The shear rate at wall (γ_w), stored elastic energy (W), shear modulus (G) and relaxation time (t_R) for all the blends studied at 200°C are shown in Table II and Table III. The variation of modified Weissenburg number (W'_N) is a monotonically increasing function of α and is shown in Figure 3. The recoverable deformation (γ_m) is plotted against the extrudate swell (α) and is shown in Figure 4, which also follows a straight line pattern.

Stored elastic energy (W) increases with the increase in shear rates for all the blends. However, at still higher wall shear rate (around 1300–1350 s^{-1}) slight decrease in 'W' was observed in case of high HD containing EPDM in the blend. As observed from the Table II and III, with the increase in EVA content in the blend "W" decreases irrespective of HD content of EPDM and VA content of EVA. With the increase in HD content in EPDM, W increases for both low and high VA containing EVA. From the tables it is also observed that as the shear rate increases the relaxation time (t_R) decreases. As the EPDM/EVA ratio decreases, relaxation time also decreases with an exception for the high HD containing EPDM and high VA containing EVA where it increases. For both low and high

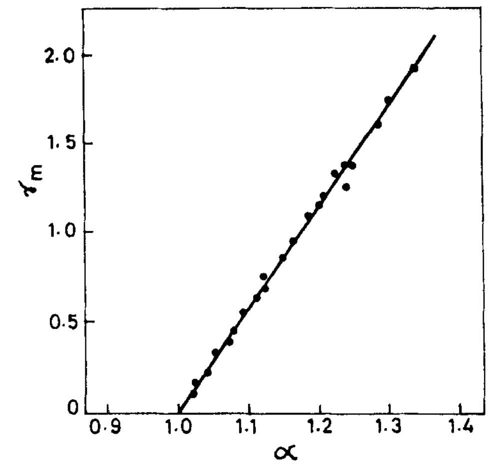

FIGURE 3 Variation of recoverable deformation with extrudate swell.

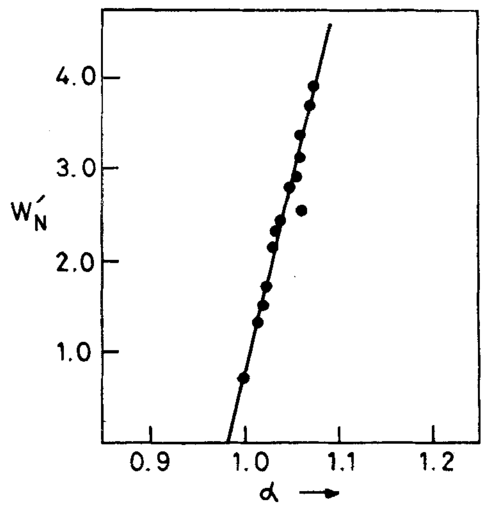

FIGURE 4 Variation of Weissenberg number with extrudate swell.

VA containing EVA, with the increase in HD content relaxation time increases. Shear modulus (G) of blends follow the same pattern as stored energy. However, at higher wall shear rate (around 1300–1357 s^{-1}) there is abrupt rise in shear modulus in case of high HD containing EPDM.

4. Morphology of Blends

Physical appearance of the extrudates of different blends are shown in Figures 5, 6, and 7 as a function of shear rate. As the shear rate is increased, the extrudate swell increases with slight wavy finishes for low HD containing EPDM (Figure 5(a)

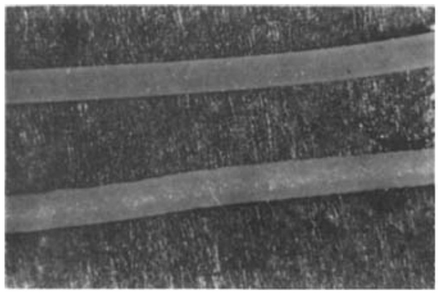

FIGURE 5 Photographs of the extrudates: (a) blend A at 33.8 s^{-1} and 200°C, (b) blend A at 338 s^{-1} and 200°C.

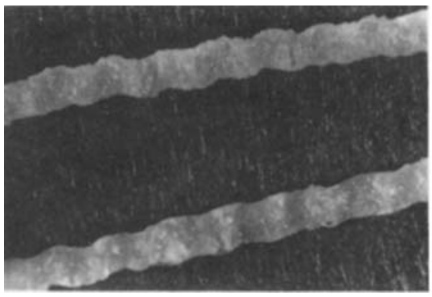

FIGURE 6 Photographs of the extrudates: (a) blend E at 33.8 s^{-1} and 200°C, (b) blend E at 338 s^{-1} and 200°C.

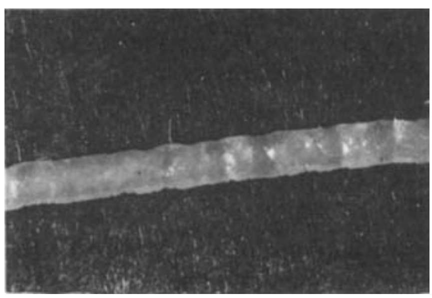

FIGURE 7 Photographs of the extrudates: blend G at 338 s^{-1} and 200°C.

and 5(b)). For high HD containing EPDM, however, circumferential wrinkles on the extrudate surfaces is observed with surface fractures at high shear rate (Figure 6(a) and 6(b)). The surface fracture appears to diminish when high VA containing EVA is used (Figure 7). More surface irregularities are observed in case of high HD containing EPDM.

Blend morphology after the solvent extraction in *n*-pentane are shown in Figures 8 and 9. As observed from the figure, at low EPDM:EVA ratio the EPDM remains in the discrete dispersed phase and high HD containing EPDM remains as a larger nodule than the low HD containing EPDM. The more uniformly distributed low HD containing EPDM phase probably makes the extrudate surface smoother. The

FIGURE 8 SEM photograph of the blend B (500×).

FIGURE 9 SEM photograph of the blend F (500×).

higher degree of melt fracture in case of high HD containing EPDM appears to be due to non-uniform distribution of EPDM phase having larger domain size. High VA containing EVA however, makes such distribution somewhat uniform thus reducing the extent of melt fracture.

5. CONCLUSION

Flow behavior and the morphology of the EPDM/EVA blends depend on the change of their HD and VA content and their relative response towards shear rate. It appears that the viscosity probably decreases at high shear rate region due to breaking up the elastomeric EPDM microgels. Pseudoplasticity is more for low VA content EVA at a fixed level of HD in EPDM. Die swell is more in case of high HD content EPDM. EPDM with high HD content exhibits higher melt fracture due to non-uniform distribution of EPDM phase having larger domain size.

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