

Studies on Miscibility of Blends of Poly(ethylene-co-methyl acrylate) and Poly(dimethyl siloxane) Rubber by Melt Rheology

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SYNOPSIS

Rheological studies of the blends of poly(ethylene-co-methyl acrylate) (EMA) and poly(dimethyl siloxane) (PDMS) rubber have been carried out at various temperatures and different shear rates by a Monsanto processability tester. The melt viscosities of the blends are found to be higher than that calculated as per the additivity rule, showing a positive deviation, an indication of synergism present in the blends during melt processing. This confirms our earlier finding that the blends are miscible throughout the composition range. The activation energy of flow of the blends are always higher than that calculated as per the additivity rule. The comparatively higher activation energy for the 30 : 70 EMA-PDMS rubber blend among the systems studied confirms our earlier finding that extent of reaction between EMA and PDMS rubber is significantly higher at this proportion of the blends studied. This substantiates the miscibility between blends of EMA and PDMS rubber throughout the composition range. © 1995 John Wiley & Sons, Inc.

INTRODUCTION

Extensive studies on the miscibility of polymer blends by different techniques have been reported earlier by several authors.¹⁻⁸ The miscibility has been attributed to either specific interaction or chemical reaction between the blend constituents causing an increase in density of the blend above that calculated from the additivity rule. In most of the cases, this has been reflected in the rheological response of the blends showing a higher melt viscosity as compared to that obtained by the log-additivity rule.

Recently, the authors have demonstrated that blends of ethylene methyl acrylate copolymer (EMA) and polydimethyl siloxane (PDMS) rubber are miscible throughout the composition range. This miscibility has been assigned as due to the chemical reaction between vinyl groups attached to the silicone atom in the PDMS rubber chain and the α -carbon to the ester group in the EMA copolymer.^{9,10}

The reaction has been confirmed from IR and ^1H as well as ^{13}C solid-state MAS NMR spectroscopic studies and the miscibility has been confirmed from dynamic mechanical analysis (DMA), which show a single T_g of the blends above that calculated by using the Fox equation in all proportions of the blend composition.⁹ A positive deviation of the glass transition temperature (T_g) also indicates the synergistic effect prevalent in the blends which has been further confirmed from mechanical property measurements.¹¹ The mechanical properties such as static modulus and tensile strength exhibit positive deviation from that of the additivity rule. The thermal stability of the blends also improve exhibiting synergism as is evident from thermal degradation studies.¹² The blends also exhibit miscibility even after crosslinking by using a common curing agent such as dicumyl peroxide.¹¹ However, the rheological properties of such blends are rare, but they are easy to measure and relatively simple to interpret as they behave almost as a single phase melt.

Utracki and Kamal¹³ have reviewed in detail the subject of melt rheology of polymer blends and have compared the rheological properties of polymer blends with those of emulsions, block polymers, and homologous polymer blends. They have been able

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to categorize the polymer blends into three groups, namely (i) those having higher value of viscosity than that predicted by the additivity rule designated as positively deviated blends (PDB), (ii) those having a lower value of viscosity than that predicted by the additivity rule designated as negatively deviated blends (NDB), and (iii) those where both these characteristics are present, termed as positive-negative deviation blends (PNDB). Later Utracki¹⁴ categorically correlated the rheological properties of these blends with their thermodynamic behavior and structure. He suggested that (i) the positive deviation is a characteristic of a homogeneous polymer blend (miscible polymer blends) due to specific polymer-polymer interactions and also of heterogeneous emulsionlike immiscible blends; (ii) the negative deviation and the positive-negative deviation may be correlated with the heterogeneous nature of the polymer blends. However, the reverse trend is not true. A heterophase polymer blend may show positive deviation if the interphase interactions are due largely to compatibilization, shear grafting, or partial specific interactions. When the interaction between the phases is very little, the viscosity of the blend will show a negative deviation and normal stress difference (N) as positive deviation, due to slippage. When there is a change in the structure on the basis of composition, i.e., phase inversion or composite drop structure, both positive and negative deviation will be exhibited.

Gupta et al.¹⁵ observed the positive deviation with the binary miscible blends of PP (polypropylene)/SEBS (styrene-*b*-ethylene butylene-*b*-styrene) than the immiscible PP/PS (polystyrene) and PP/HDPE (high density polyethylene) blends. However, introduction of a compatibilizer such as SEBS into the immiscible pairs of PP/PS and PP/HDPE increased the melt viscosity of the ternary blends above that expected on the basis of log-additivity rule.

The present work is an attempt to correlate the melt viscosity of the blends with its miscibility characteristics. Thus, it deals with the study of melt viscosity, activation energy of flow, and the flow behavior index of the blends at four different shear rates beginning from 12.28 to 2452 s⁻¹ and at three different temperatures, viz., 120, 135, and 150°C.

EXPERIMENTAL

Materials

Polydimethyl siloxane (PDMS) rubber containing a small percentage (3.8) of vinyl groups (Silastic WC-

50) was supplied by M/s Dow Corning Inc., USA, with the following specifications:

Specific gravity: 1.15.
Brittle temperature: -39°C.

EMA (OPTEMA TC-120) was supplied in kind by M/s. Exxon Chemicals Eastern Inc., Bombay, India, with the following specifications:

Methyl acrylate content, %: 21.
Melting point, °C: 81.
Density, g/cm³: 0.94.
 \bar{M}_n : 31,500.
 \bar{M}_w : 2,20,700.
Polydispersity (M_w/M_n): 7.

Preparation of the Blend

Melt blending of the constituents in various proportions was carried out in a Brabender Plasticorder (model PLE-330), at 180°C for 10 min at 100 rpm rotor speed. The molten mass was then taken out from the plasticorder and sheeted out in a laboratory open two-roll mill (150 × 300 mm) at room temperature. The blends have been designated EM₇₀, EM₅₀, and EM₃₀, subscripts denoting the proportion of EMA in the blend.

Measurement of Flow Properties

The melt flow properties of the pure components and the blends were carried out by means of a Monsanto processability tester (MPT), a capillary rheometer at three different temperatures (120, 135, and 150°C) and at four different shear rates (12.26, 122.6, 1226, and 2452 s⁻¹).

RESULTS AND DISCUSSION

While analyzing the data from the above experiments, several assumptions have been made to simplify the results. For example, a steady flow is assumed throughout the length of the die. It is assumed that the pressure drop over the length of the die is linear and that the exit pressure from the die is zero. The formula for shear rate assumes constant viscosity at all shear rates, which is clearly not so for polymers. As a result the data from MPT tests is expressed as apparent values. The apparent shear rate and shear stress values can be calculated by using the following equations:

Apparent shear rate at the wall:

$$\dot{\nu}_{wa} = \frac{32 \text{ (Barrel area)(Ram rate)}}{\pi(\text{capillary die diameter})^3} \quad (1)$$

Apparent shear stress at the wall:

$$\tau_{wa} = \frac{\text{Barrel pressure}}{4(\text{capillary die length/diameter})} \quad (2)$$

By introducing correction factors, the true values can be obtained as,

True shear stress at the wall (τ_w):

$$\tau_w = \frac{\text{MPT barrel pressure}}{4(L/D + L/D \text{ correction})} \quad (3)$$

where L/D correction is called Bagley correction factor.

In the present case, the melt was allowed to enter into the round capillary die having L/D ratio of 20 : 1 at multiple entry angle of 45° and 60° , which minimize the pressure drop and, hence, introduction of the correction factor, is not needed. Therefore, the apparent shear stress is taken approximately as the true shear stress.

True shear rate ($\dot{\nu}_w$) at the wall is obtained by applying the Robinowitz correction¹⁶ as,

$$\dot{\nu}_w = \frac{3n' + 1}{4n'} \times \dot{\nu}_{wa} \quad (4)$$

where $n' = d \ln \dot{\nu}_{wa} / d \ln \tau_{wa}$.

If the plot of log apparent shear stress versus log apparent shear rate is a straight line, then the slope n' is constant and the materials obey power law model, with $n = n'$. Thus,

$$\tau_w = K \dot{\nu}_w^n / n \quad (5)$$

where K is in consistency index, τ_w is true shear stress at the wall, $\dot{\nu}_w$ is true shear rate at the wall, and n in the flow behavior index.

When $n = 1$, the material is Newtonian; when $n < 1$, the material is pseudoplastic; and when $n > 1$, the material is dilatant. Taking log of the above equation,

$$\log \tau_w = \log K + n \log \dot{\nu}_w \quad (6)$$

The log-log plots of τ_w vs. $\dot{\nu}_w$ yield straight lines. The slope (n) and the intercept (K) of the blends

and the pure components at three different flow temperatures are given in Table I.

Flow Behavior

It is observed from Table I that the flow behavior index n increases gradually from 0.5 for EMA to 0.8 for PDMS rubber. Lower value of n , i.e., $n < 1$ is characteristic of non-Newtonian, i.e., pseudoplastic or shear thinning behavior of the pure components and the blends. The values for the blends lie in between them. With an increase in the PDMS content in the blend, n increases obviously at all three processing temperatures, indicating a reduction in the pseudoplastic character of the blends. Further, an increase in the processing temperature from 120 to 150°C lowers the n value marginally for all the blends and the pure components. The consistency index (K) increases with PDMS rubber but decreases with an increase in flow temperature, the variation being much less in the case of PDMS rubber than in the case of EMA, whereas blends occupy intermediate positions depending on the composition.

Melt Viscosity

The melt viscosity of the pure components and the blends are plotted against the shear stress, at three different temperatures, viz. 120, 135, and 150°C as

Table I Flow Behavior Index (n) and Consistency Index (K) of EMA, PDMS Rubber and Their Blends at Different Temperatures

Sample (EMA-PDMS)	Temperature ($^\circ\text{C}$)	n	K (Pa s^n)
100 : 0	120	0.593	16464
	135	0.556	10786
	150	0.536	7921
70 : 30	120	0.640	21862
	135	0.630	17448
	150	0.617	13214
50 : 50	120	0.708	27167
	135	0.683	20741
	150	0.658	14847
30 : 70	120	0.767	36240
	135	0.745	28140
	150	0.741	26275
0 : 100	120	0.84	42878
	135	0.833	39391
	150	0.829	35365

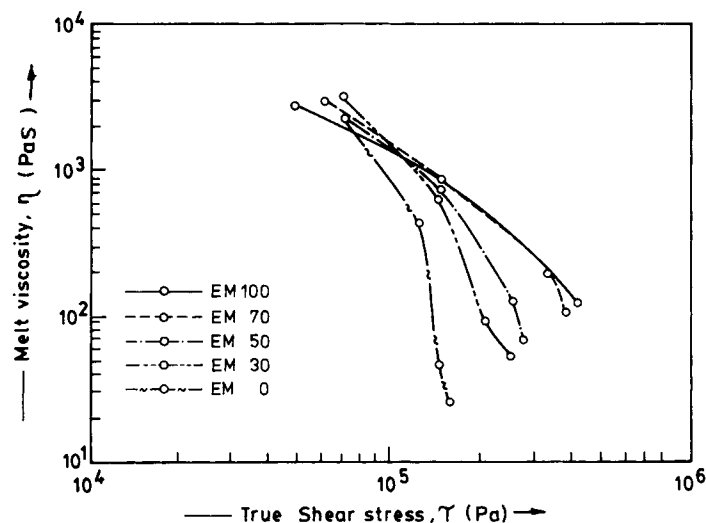


Figure 1 Plot of melt viscosity of the blends and the pure components against the true shear stress at 120°C. (Subscripts in the figures indicate the proportion of EMA in the blends.)

depicted in Figures 1-3. It is observed in general that at all temperatures of capillary flow, the shear viscosity decreases with an increase in shear stress, an indication of shear thinning effect of the blends, which is characteristic of their pseudoplastic nature. However, at all three different processing temperatures the reduction in melt viscosity of PDMS rubber is more drastic at higher shear stresses, i.e., beyond 10^5 than that observed for EMA in which case the reduction in melt viscosity is minimum at all three temperatures. The true shear stress (τ_w), the

true shear rate ($\dot{\nu}_w$), and the true viscosity (η) of the blends and individual constituents at three processing temperatures are given in Tables II-IV.

It is evident from the tables that an increase in the shear rate decreases the melt viscosity of all the blends and the pure components invariably at all the temperatures studied because of shear thinning effect of the materials. But with an increase in concentration of PDMS rubber in the EMA matrix, the shear viscosity increases above those of the pure components in all the proportions with a minimum

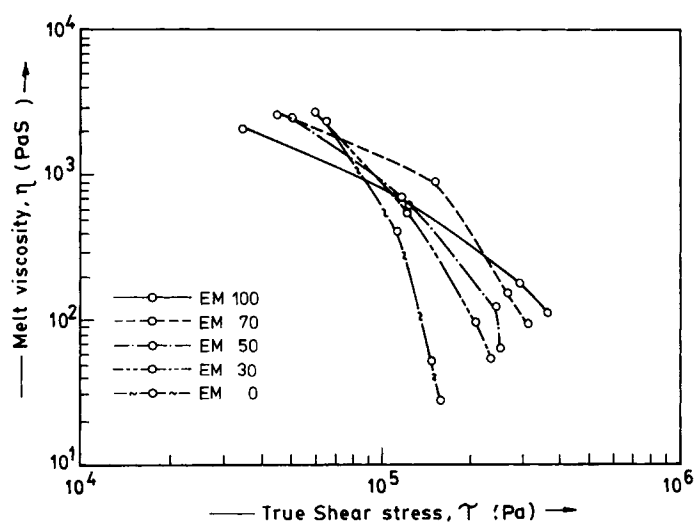


Figure 2 Plot of melt viscosity of the blends and the pure components against the true shear stress at 135°C. (Subscripts in the figures indicate the proportion of EMA in the blends.)

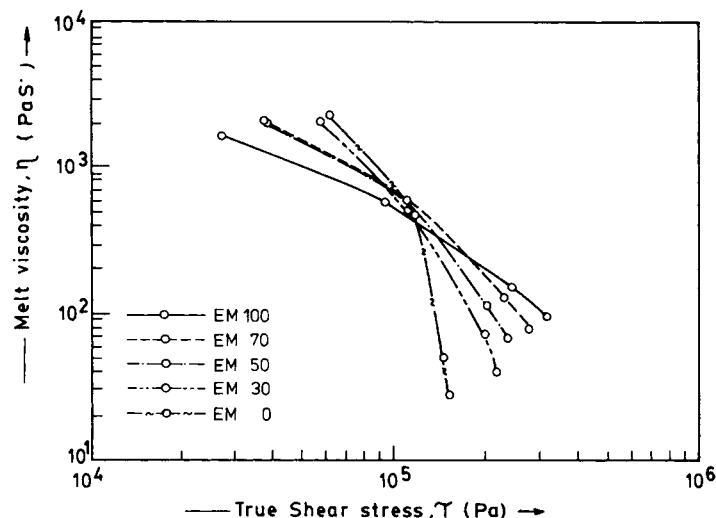


Figure 3 Plot of melt viscosity of the blends and the pure components against the true shear stress at 150°C. (Subscripts in the figures indicate the proportion of EMA in the blends.)

value for the PDMS rubber. This is particularly more prominent at lower shear rates ($< 160 \text{ s}^{-1}$) as seen from Tables II–IV, whereas at higher shear

Table II Melt Viscosity of the EMA–PDMS Rubber Blends and Its Pure Components at Various Shear Rates and at 120°C

Blends (EMA–PDMS)	True Shear Rate (s^{-1})	True Shear Stress (Pa)	True Viscosity (Pa s)
100 : 0	16.76	49002	2923.6
	167.6	147013	877.1
	1676	341000	203.5
	3352	418000	130
70 : 30	17.75	59502	3352.3
	177.5	148346	835.7
	1775	337150	190.0
	3550	364964	106
50 : 50	19.91	61459	3086.8
	199.1	146384	735.3
	1991	257460	129.3
	3982	276585	072
30 : 70	22.75	71554	3145.3
	227.5	146911	645.7
	2275	206581	90.8
	4550	250839	52
0 : 100	30.48	71000	2329.4
	304.8	128000	420.0
	3048	147000	48.2
	6096	159000	28.5

rates ($> 160 \text{ s}^{-1}$), the viscosity values of the blends lie in between those of the pure components. This may be explained as due to higher wall slip at higher shear rates as suggested by Turner and Moore.¹⁷ It has also been observed that on increasing the temperature of shear flow from 120 to 150°C that the shear viscosity decreases nonuniformly for all the blends and pure components, only varying in degree. The reduction in viscosity is more drastic for EMA copolymer and marginal for PDMS rubber at lower shear rates, whereas in case of the blends, the reduction is intermediate depending upon the blend composition. Also at higher shear rates ($> 3550 \text{ s}^{-1}$), the decrease in viscosity is marginal for the blends and PDMS rubber. This may be explained as due to increase in the free volume at higher temperatures.

It is interesting to note that at lower shear stresses the viscosity of PDMS rubber is higher than that of EMA, whereas at higher shear stresses, the reverse is true, i.e., viscosity of EMA is higher than that of the PDMS rubber at the same processing temperature. This is attributed to the fact that PDMS rubber contains reinforcing fumed silica as a filler up to 35% as determined by thermogravimetric analysis (TGA).¹² Thus there are two types of interactions possible, i.e., polymer–polymer interaction and polymer–filler interaction. At low shear stresses both interactions are not affected, hence the viscosity of PDMS rubber is very high at low shear stresses. But as the shear stress increases, both the interactions are broken, leading to loosely bound structures and easy orientation of molecules along the direction of

Table III Melt Viscosity Data of the Pure Components and Blends of EMA and PDMS Rubber at 135°C at Various Shear Rates

Sample (EMA-PDMS)	True Shear Rate (s ⁻¹)	True Shear Stress (Pa)	True Viscosity (Pa s)
100 : 0	16.14	35000	2168.5
	161.4	116000	718.7
	1614	290000	180.0
	3228	364000	112.8
70 : 30	16.7	45369	2716.7
	167.0	151570	907.6
	1670	263222	157.6
	3340	314591	94.2
50 : 50	19.41	50299	2591.4
	194.1	124120	640.0
	1941	242993	125.2
	3882	250349	64.5
30 : 70	21.12	59821	2832.5
	211.2	122428	580.0
	2112	206703	98.0
	4224	233675	50.3
0 : 100	28.32	66000	2330.5
	283.2	113000	400.0
	2832	146000	51.6
	5664	157000	27.8

stress, together with the slippage of molecules. This structural breakdown is well supported by a drastic drop in viscosity as shear stress increases. On the other hand, melt viscosity of EMA reduces steadily with the increase in shear stress, which may be due to higher intermolecular force of interactions between the chain segments due to the presence of polar methyl acrylate units and comparatively lesser orientation under high shear stress. The viscosities of the blends follow an intermediate path depending on the composition. The shear thinning characteristics of the blends and the pure components at different temperatures are depicted in Figures 1-3. It is interesting to note that at intermediate shear stresses, the viscosities of both EMA and PDMS rubber come closer to each other, implying that a homogeneous dispersion of the blends is possible in the intermediate shear stress of 10⁵. This may be treated as the critical shear stress. Another interesting phenomenon observed is that the melt viscosities of the blends always lie in between those of the pure components at all shear stresses, except at the critical stress region.

Table V shows the experimental melt viscosity ($\ln \eta$)_{expt1} and theoretical melt viscosity ($\ln \eta$)_{theo} of all the blends at three different temperatures and four different shear stresses, calculated by using the Utracki equation¹⁴ as given below:

$$\ln(\eta)_{\text{blend}} = \sum W_i \ln(\eta)_i \quad (7)$$

where W_i is the weight fraction of the component in the blend. The values are expressed in log scale. The experimental melt viscosities are found to be higher than the theoretical ones for the blends showing a positive deviation from the additivity rule. This trend of positive deviation in melt viscosity is observed at all the processing temperatures for all the blends, which is a characteristic property of miscible and homologous polymer blends. Therefore it is inferred that the blends of EMA and PDMS rubber are miscible throughout the composition range as has already been reported earlier,⁹ to be due to chemical reaction between the blend constituents.

Table IV Melt Viscosity of Blends of EMA and PDMS Rubber at Various Shear Rates and at 150°C

Sample (EMA-PDMS)	True Shear Rate (s ⁻¹)	True Shear Stress (Pa)	True Viscosity (Pa s)
100 : 0	15.84	27000	1704.5
	158.4	93600	591.0
	1584	245300	154.8
	3168	313100	90.0
70 : 30	17.30	36932	2134.8
	173.0	110008	635.8
	1730	229262	132.5
	3460	278792	83
50 : 50	18.13	38162	2104.9
	181.3	112914	622.8
	1813	202472	116.8
	3626	240296	58.0
30 : 70	20.9	57989	2774.1
	209.0	120429	576.2
	2090	199102	95.3
	4180	218228	45.0
0 : 100	27.43	61300	2234.8
	274.3	116060	423.1
	2743	145300	52.9
	5486	151700	27.3

Table V Experimental and Theoretical Viscosities of the Blends of EMA and PDMS Rubber at Various Temperatures and Shear Rates

Blend Code (EMA-PDMS)	120°C		135°C		150°C	
	ln η (theor)	ln η (exptl)	ln η (theor)	ln η (exptl)	ln η (theor)	ln η (exptl)
100 : 0	—	7.98	—	7.68	—	7.44
		6.77		6.57		6.38
		5.31		5.19		5.04
		4.86		4.72		4.50
70 : 30	7.93	8.12	7.7	7.90	7.53	7.67
	6.56	6.72	6.4	6.81	6.28	6.46
	4.88	5.31	4.82	5.06	4.72	4.89
	4.40	4.86	4.30	4.55	4.14	4.42
50 : 50	7.88	8.03	7.72	7.86	7.58	7.65
	6.41	6.60	6.29	6.46	6.21	6.43
	4.60	4.86	4.57	4.83	4.5	4.76
	4.10	4.28	4.02	4.17	3.9	4.06
30 : 70	7.83	8.05	7.72	7.95	7.73	7.92
	6.26	6.47	6.17	6.36	6.14	6.35
	4.31	4.51	4.32	4.58	4.20	4.56
	3.81	3.91	3.74	3.91	3.66	3.95
0 : 100	—	7.75	—	7.75	—	7.71
		6.04		5.99		6.04
		3.87		3.94		3.96
		3.35		3.32		3.30

Activation Energy of Flow

The activation energy of viscous flow derived from the Arrhenius type of relation is valid for power law fluids¹⁸ and is calculated by using the following equation:

$$\eta = Ae^{E/RT} \quad (8)$$

where A is a constant, E is the activation energy of viscous flow, R is the gas constant, and T is the absolute temperature.

Plots of melt viscosity against reciprocal of temperature at low shear rate (12.26 s⁻¹) and at high shear rate (2452 s⁻¹) for all the blends and individual components are depicted in Figure 4. The plots are linear and their slopes are equal to E/R . It is observed that the activation energy of flow is less at high shear rates than that at lower shear rates for all the blends and pure components. However, at a higher shear rate (2452 s⁻¹) EMA has higher activation energy of flow than that of the PDMS rubber. This implies that PDMS rubber is less temperature sensitive. This is attributed to its typical structure

and lower intermolecular forces of attraction.¹⁹ The activation energies of the blends lie in between the pure components. It has been observed that the difference in activation energies between the blends and the pure constituents is less at low shear rates.

Plots of activation energy versus blend composition at both low and high shear rates, i.e., 12.26 and 2452 s⁻¹, respectively, are shown in Figures 5 and 6. Both figures show a reduction in the activation energy of flow with an increase in PDMS rubber concentration so as to exhibit a positive deviation from that obtained by the additivity rule. Thus, the blends require higher activation energy of flow than that calculated as per the additivity rule. Besides, the 30 : 70 EMA-PDMS rubber blend show maximum deviation from the additivity rule. This has been attributed to a very strong interaction via chemical reaction between the blend constituents as reported earlier forming a grafted copolymer, i.e., EMA-*g*-PDMS. This type of synergism is maximum for the blend proportion 30 : 70 at both the shear rates because of availability of more vinyl silicone groups in PDMS rubber to interact with EMA. This justifies for the highest activation energy of flow for

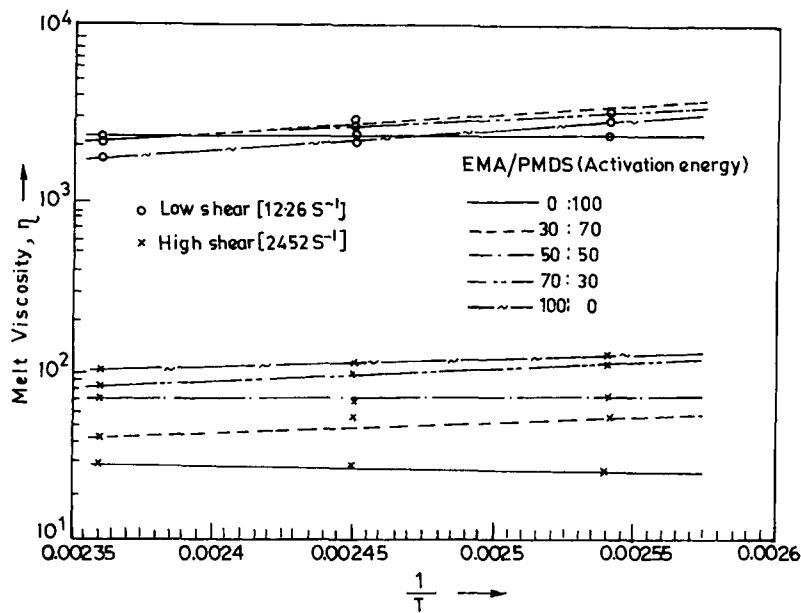


Figure 4 Plot of melt viscosity against reciprocal of temperature for the pure components and the blends at low (12.26 s^{-1}) and high (2452 s^{-1}) shear rates.

EM₃₀ blend. Similar observations have been made earlier from dynamic mechanical analysis.⁹

CONCLUSIONS

The following conclusions may be drawn from the present investigation:

1. On increasing shear rate and temperature of shear flow, the melt viscosity decreases for all the blends and pure components concurring with the shear thinning effect of the materials.
2. The melt viscosity of PDMS rubber is higher than that of EMA copolymer at low shear stresses except at 120°C , whereas the reverse is true at higher shear stresses.
3. The melt viscosity of the blends fall in between those of the pure components at high and low shear stresses whereas at intermediate shear stresses the viscosities are nearer to each other.
4. The melt viscosity of the blends at all shear stresses are found to be greater than that calculated as per additivity rule, implying that the blend viscosity show positive deviation from that of the additivity rule, a clear indication of miscibility of the polymer blends in the entire composition range.

5. The activation energy of flow decreases with PDMS rubber concentration in the blend, but 30:70 EMA-PDMS show highest activation energy of viscous flow than that exhibited by other blends, due to the synergistic effect a consequence of occurrence of chemical reaction between the blend constituents through grafting reactions.

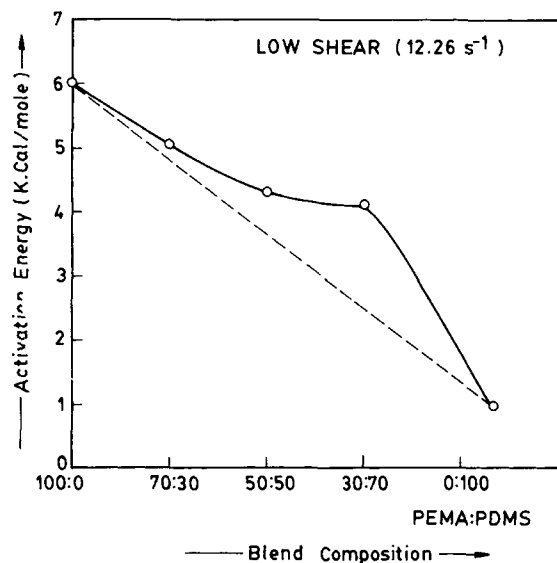


Figure 5 Plot of activation energy vs. blend composition at low shear rate (12.26 s^{-1}).

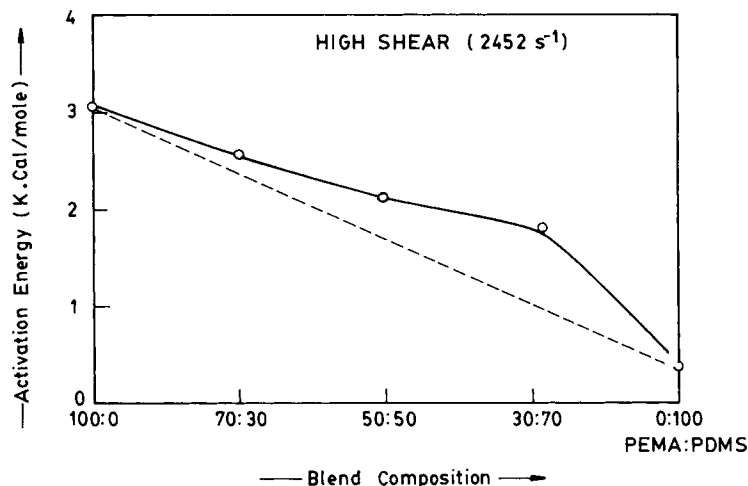


Figure 6 Plot of activation energy vs. blend composition at high shear rate (2452 s⁻¹).

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