

# Rheological and Mechanical Properties of Poly(ethylene acrylic acid) and Low Density Polyethylene Blends

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Received 22 January 2010; accepted 8 October 2010

DOI 10.1002/app.33957

Published online 31 March 2011 in Wiley Online Library (wileyonlinelibrary.com).

**ABSTRACT:** Rheological properties of poly (ethylene-acrylic acid) (PEA) and low density poly ethylene (LDPE) blends having varied amounts of LDPE from 0 to 100% have been evaluated at different temperatures (115, 120, and 130°C) and shear rates (61.33–613.30 s<sup>-1</sup>) using a Monsanto processability tester. A reduction in the melt viscosity of the PEA/LDPE blends was noticed with increasing the shear rate. The observed positive deviation in the experimental melt viscosities of the blends is an indication of the synergy present in the blends during melt processing. The activation energy ( $E_a$ ) of flow calculated using Arrhenius relation for PEA, LDPE, and their respective blends lies in the range 29.98–40.56 kJ mol<sup>-1</sup>. The experimental activation energy of

flow of the blends was higher than that obtained from the additivity rule. Highest activation energy was noticed for the blends containing 60–80% by weight of LDPE in PEA/LDPE blends, which is an indication for the miscibility of the blends at these ratios. The physicochemical properties such as density, tensile behavior, tear strength, and hardness (Shore A) of PEA, LDPE, and their blends have been evaluated as a function of varying amounts of LDPE. The obtained physicochemical properties of the PEA/LDPE blends lie in between that of pure polymers. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 121: 3070–3077, 2011

**Key words:** blend; PEA; LDPE; melt rheology; miscibility

## INTRODUCTION

During the last few decades, the commercial importance of polymer blends have been increased due to the possibility of attaining a wide range of properties by simple blending technique. Immiscible blends are preferred over miscible blends due to the advantage of obtaining a combination of the properties of the individual polymers. In miscible blends an average of the individual polymer properties can be expected.<sup>1</sup>

As the commercial importance of polymer blends is increasing, it is necessary to optimize the processing conditions for each blend. Generally, in the case of homopolymer, the flow behavior depends on the flow geometry and processing conditions such as the temperature, shear rate, time of flow, etc. In case of polymer blends, the flow behavior becomes more complex and it is influenced by additional factors like the miscibility of the system, the morphology, interfacial adhesion, and interfacial thickness.

A numerous research papers have been published in the past on the miscibility of polymer blends by different techniques.<sup>2–9</sup> The miscibility has been attrib-

uted to either specific interaction or chemical reaction between the blend constituents causing an increase in density of the blend above the theoretical density obtained 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. The complex rheological behavior of polymer blends have been investigated by several researchers.<sup>10–14</sup>

The mechanical properties such as tensile strength and static modulus of the blends exhibit a positive deviation from that of additivity rule. The thermal degradation studies<sup>15</sup> revealed the synergistic thermal stability of the blends. The blends also exhibit miscibility even after crosslinking by using a common curing agent such as dicumyl peroxide.<sup>16</sup> 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 Kamal<sup>17</sup> 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 to categorize the polymer blends into three groups, namely; (i) positively deviated blends (PDB); having higher value of experimental viscosity than that of

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theoretical or predicated one by the additivity rule (ii) negatively deviated blends (NDB); a lower value of viscosity than the one predicted by the additivity rule (iii) positive–negative deviation blends (PNDB); the presence of both positive and negative deviation from the theoretical one falls under this class. Later, Utracki<sup>18</sup> categorically correlated the rheological properties of these blends with their thermodynamic behavior and structure. He suggested that; (i) the positive deviation is a characteristic feature of a homogeneous polymer blend (miscible polymer blends) due to specific polymer-polymer interactions, (ii) the negative deviation and the positive–negative deviation may be correlated with the heterogeneous nature of the polymer blends. A heterophase polymer blend may show positive deviation if the interface interactions are due 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 ( $N$ ).

Gupta et al.<sup>19</sup> observed a 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. Bhattacharya et al.<sup>20</sup> and Santra et al.<sup>21</sup> have demonstrated that, the blends of ethylene methacrylic acid (EMA)/polydimethyl siloxane (PDMS) are miscible throughout the composition range. This miscibility has been assigned due to the chemical reaction between PDMS and EMA. Blends of LDPE have been extensively used in packaging industries because of easy availability, processability, and excellent optical and physicochemical properties. Lee et al.<sup>22</sup> have investigated the miscibility of LDPE/LLDPE blends. The *in situ* compatibilization of PS/PE blends was reported by Song and Baker.<sup>23</sup> The effect of PP on the rheological behavior of PS/SEBS blends were reported by Raha et al.<sup>24</sup> Several researchers have investigated the miscibility of different polymer blends.<sup>25–31</sup>

Polyethylene acrylic acid (PEA) is well known for its use in conventional extrusion coating, coextrusion coating, and extrusion lamination. The benefits of PEA such as excellent adhesion to various substrates such as foils, paper, films, etc., have received considerable attention to use it along with other polymers which requires the aforementioned properties for specific applications. PEA/LDPE blends have many industrial uses because of their good mechanical strength, processability, impact strength, etc. A thorough literature survey revealed the lack of information related to the evaluation of rheological and mechanical properties of PEA/LDPE blends. The present research investigation concentrated on the rheological and mechanical properties of PEA/LDPE blends as a function of varying amounts of LDPE.

## EXPERIMENTAL

### Materials

Polyethylene acrylic acid (Escor 5001) was supplied by M/s B.F. Good Rich Chemical Company, UK, with the following specifications: acrylic acid content—6.2% by weight, density—0.931 g cm<sup>-3</sup>, melt flow index (MFI)—2 g/10 min, and mp—99–100°C. Low-density polyethylene (LDPE) having the melting point of 105–110°C and density of 0.92 g cm<sup>-3</sup> was obtained from M/s Polyolefin Industries, India.

### Preparation of the blend

Melt blending of PEA/LDPE blends having different compositions of PEA and LDPE was carried out in a Brabender Plasticorder (mold PLE-330) at 150°C with a rotor speed of 80 rpm for 6 min. PEA was melted first for 2 min, then LDPE was added and mixed for additional 6 min. The total mixing time was 8 min in all the cases. The molten mass was then taken out from the plasticorder and sheeted out immediately in a laboratory open two-roll mixing mill (150 mm × 300 mm) at room temperature.

### Rheological measurement

The rheological characteristics of PEA/LDPE blends were measured on a Monsanto processability tester (MPT), which is a micro-processor-controlled programmable capillary rheometer having an  $L/D$  ratio of 20. The extrusion studies were carried out at three different temperatures of 115, 120, and 130°C, and at four different shear rates (61.33, 122.66, 245.32, and 613.3 s<sup>-1</sup>).

The apparent shear rate ( $\gamma_a$ ), apparent shear stress ( $\tau_a$ ), and apparent viscosity ( $\eta_a$ ) were calculated using the following mathematical expressions;

$$\gamma_a = 32Q/\pi d_c^3 \quad (1)$$

$$\tau_a = \Delta P/4(L_c/d_c) \quad (2)$$

$$\eta_a = \tau_a/\gamma_a \quad (3)$$

where  $Q$  = volumetric flow rate of melt in m<sup>3</sup> s<sup>-1</sup> (Barrel cross section area × plunger velocity).  $d_c$  = capillary diameter in meter,  $L_c$  = capillary length in meter and  $\Delta P$  is pressure drop across the capillary in pascals. The flow behavior or pseudoplasticity index " $n$ " and consistency index " $K$ " were determined from the power law expression:

$$\tau_a = K(\gamma_a)^n \quad (4)$$

Logarithmic plots of  $\tau_a$  and  $\gamma_a$  were found to be linear over the experimental shear rate range, from the slope and intercept of which the parameters  $n$  and  $K$  were estimated.

**TABLE I**  
**Flow Behavior Index ( $\eta$ ) and Consistency Index ( $K$ ) of PEA, LDPE, and Their Blends at Different Temperatures**

PEA/LDPE blend (wt %)	Temperature (°C)	$\eta$	$R^2$	$K$ (Pa s <sup>-1n</sup> )	$R^2$
100/0	115	0.32	0.991	1.72	0.992
	120	0.35	0.992	1.58	0.993
	130	0.37	0.982	1.47	0.983
80/20	115	0.39	0.990	1.57	0.978
	120	0.35	0.981	1.55	0.989
	130	0.37	0.986	1.42	0.899
60/40	115	0.43	0.978	1.42	0.956
	120	0.42	0.982	1.37	0.987
	130	0.38	0.989	1.36	0.968
40/60	115	0.43	0.967	1.46	0.987
	120	0.35	0.976	1.38	0.982
	130	0.37	0.989	1.34	0.988
20/80	115	0.42	0.969	1.47	0.965
	120	0.33	0.989	1.39	0.955
	130	0.37	0.988	1.32	0.966
0/100	115	0.36	0.977	1.41	0.989
	120	0.36	0.983	1.38	0.999
	130	0.36	0.987	1.33	0.988

## RESULTS AND DISCUSSION

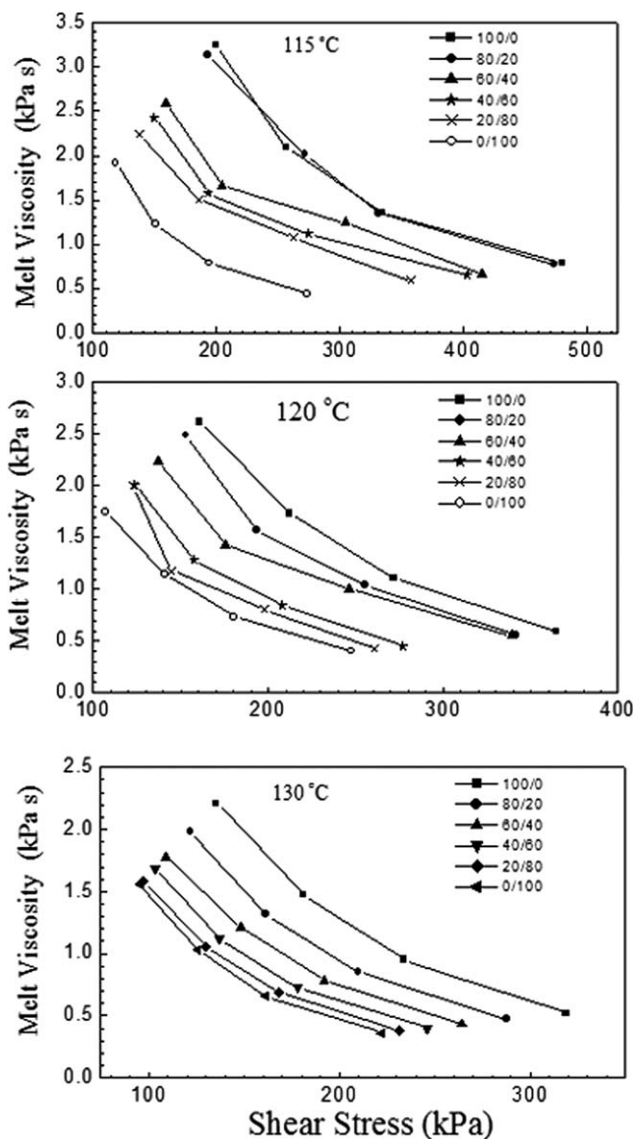
### Flow behavior

The flow behavior index " $n$ " and the consistency index " $K$ " were obtained from the power law fluid model<sup>31</sup> can be used to understand the rheology of polymer materials. The " $n$ " and " $K$ " of PEA/LDPE blends and their respective pure components at three different flow temperatures are given in Table I. The effect of different blend composition and temperatures did not show any systematic variation in " $n$ " values. It can be observed from the Table I that, the values of " $n$ " lies in the range 0.3231–0.4377. The value of  $n < 1$  is an indication for the characteristic non-Newtonian behavior (pseudoplastic or shear thinning) of the pure components and blends.<sup>31</sup> The " $K$ " value found to be higher for PEA as compared to LDPE. It can be noticed that, the incorporation of LDPE into PEA increased the melting temperature and reduced the " $K$ " values. The " $n$ " and " $K$ " values of the blends found to be intermediate between that of pure components.

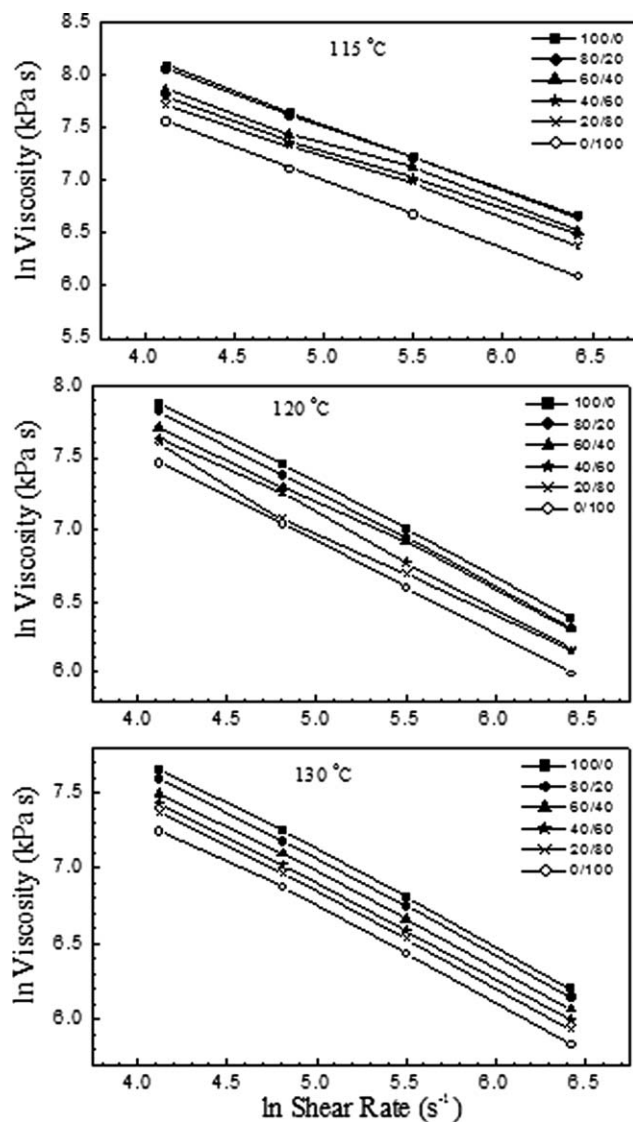
### Melt viscosity

The effect of different blend ratio, shear stress, and temperatures on the melt viscosity of PEA, LDPE, and their blends are shown in Figure 1. In general, it can be observed that, at all temperatures and blend ratios, the melt viscosity found to decrease as a function of increasing shear stress. This is an indication for the shear thinning of the blends and also the characteristic feature of pseudoplastic behavior of polymer blends.

The increased shear stress is expected to break the polymer–polymer interaction leading to loosely bound structures and easy orientation of molecules along the direction of stress together with the slippage of molecules. This structural breakdown is well supported by a drastic drop in viscosity as a function of increased shear stress. At practically zero shear stress, the randomly oriented and entangled molecules may be responsible for a high viscosity. From the Figure 1, it can be noticed that, the viscosity of PEA was higher than that of the LDPE at a given processing temperature. A reduction trend in the melt viscosity of PEA/LDPE blends can be observed with increasing the LDPE content over the entire shear stress values. On the other hand, the melt viscosity of PEA reduces steadily with an increase in shear stress. This can be attributed to



**Figure 1** Plot of melt viscosity of PEA, LDPE and their blends as a function of shear stress.



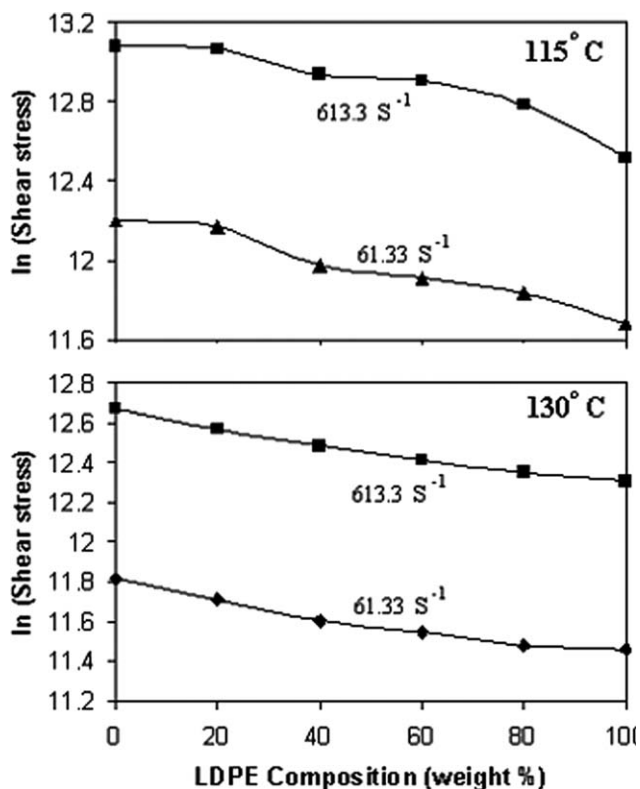
**Figure 2** Plot of melt viscosity of PEA, LDPE and their blends as a function of shear rate.

high intermolecular force of interactions between the chain segments in PEA due to the presence of polar acrylic units and comparatively lesser orientation under high shear stress. The melt viscosity of all the blends and their pure components found to reduce with increase in the shear rate at all the temperatures. This may be due to shear thinning effects of the materials. A linear reduction in the melt viscosity of PEA, LDPE and their blends as a function of shear rate can be observed from Figure 2. The melt viscosity values of the blends lies in between that of pure components. The reduction in melt viscosity as a function of increased shear stress may be due to the higher wall slippage (suggested by Turner and Moore<sup>32</sup>). The observed reduction in viscosity was more drastic for PEA and marginal for LDPE at lower shear rates, whereas in case of the blends, the reduction is intermediate depending on the blend composition.

The effect of low ( $61.33 \text{ S}^{-1}$ ) and high ( $613.3 \text{ S}^{-1}$ ) shear rate on the shear stress as a function of varying amount of LDPE in PEA/LDPE blends at 115 and  $130^\circ\text{C}$  is shown in Figure 3. A nonlinear relationship between the shear stress and LDPE content in PEA/LDPE blends can be noticed at both low and high shear rate. The observed higher shear stress incase of PEA may be due to the polar nature of PEA.

The interaction (strong or weak) between the two phases of a blend can be known by measuring the deviation of measured (experimental) viscosity from the calculated viscosity using additivity rule.<sup>33</sup> Both miscible and immiscible blends have the tendency to exhibit positive or negative deviation depending on the nature of polymer, blend type, composition, processing conditions, etc. In general, the negative deviation is expected due to the slippage of inter-layer due to the formation of weak interface.<sup>28,34</sup> Table II shows the experimental ( $\ln \eta_{\text{exptl}}$ ) and theoretical melt viscosity ( $\ln \eta_{\text{theo}}$ ) of all the blends at three different temperatures and four different shear stresses calculated using the Utracki equation<sup>18</sup> given below:

$$\ln(\eta)_{\text{blend}} = \sum W_i \ln(\eta)_i \quad (5)$$



**Figure 3** Plot of LDPE composition verses ln shear stress at 115 and  $130^\circ\text{C}$ .

TABLE II  
Experimental and Theoretical Viscosity of PEA, LDPE, and Their Blends at Various Temperatures and Shear Rates

Blends (PEA/LDPE) (wt/wt, %)	Shear rate (s <sup>-1</sup> )	115°C			120°C			130°C		
		ln η (theor)	ln η (exptl)	Error (%)	ln η (theor)	ln η (exptl)	Error (%)	ln η (theor)	ln η (exptl)	Error (%)
100/0	61.33	-	-	-	-	-	-	-	-	-
	122.66	-	8.087	-	-	7.871	-	-	7.651	-
	245.32	-	7.644	-	-	7.458	-	-	7.257	-
	613.3	-	7.217	-	-	7.010	-	-	6.810	-
80/20	61.33	-	6.662	-	-	6.389	-	-	6.204	-
	122.66	7.982	8.052	0.87	7.790	7.821	0.396	7.571	7.594	0.303
	245.32	7.537	7.608	0.93	7.375	7.381	0.081	7.161	7.182	0.292
	613.3	7.109	7.208	1.37	6.928	6.947	0.273	6.735	6.751	0.237
60/40	61.33	6.549	6.648	1.49	6.311	6.321	0.158	6.129	6.151	0.358
	122.66	7.876	7.858	0.23	7.708	7.714	0.078	7.491	7.494	0.040
	245.32	7.431	7.436	0.07	7.294	7.295	0.013	7.106	7.098	0.111
	613.3	7.000	7.123	1.73	6.846	7.122	3.875	6.661	6.663	0.030
40/60	61.33	6.436	6.518	1.26	6.233	6.315	1.298	6.055	6.066	0.181
	122.66	7.772	7.795	0.3	7.626	7.628	0.026	7.409	7.430	0.283
	245.32	7.295	7.361	0.9	7.211	7.258	0.648	7.031	7.021	0.140
	613.3	6.892	7.019	1.81	6.765	6.773	0.118	6.587	6.588	0.015
20/80	61.33	6.324	6.488	2.53	6.155	6.164	0.146	5.981	5.994	0.217
	122.66	7.666	7.714	0.62	7.544	7.600	0.737	7.329	7.367	0.516
	245.32	7.219	7.319	1.36	7.130	7.073	0.812	6.955	6.965	0.144
	613.3	6.783	6.974	2.74	6.683	6.693	0.149	6.513	6.532	0.291
0/100	61.33	6.211	6.368	2.47	6.132	6.152	0.325	5.906	5.932	0.438
	122.66	-	7.561	-	-	7.463	-	-	7.249	-
	245.32	-	7.112	-	-	7.047	-	-	6.880	-
	613.3	-	6.675	-	-	6.601	-	-	6.439	-

where  $W_i$  is the weight fraction of the component in the blend. The observed experimental melt viscosities of all the blends were higher than the theoretical ones. All the blends have showed a positive deviation from the additivity rule. The melt viscosity of all the blends at all processing temperatures showed a trend of positive deviation. Therefore it is inferred that, the blends of PEA and LDPE are miscible throughout the composition range. This is may be due to the fact that, both PEA and LDPE belongs to the same homologous series.

#### Activation energy of flow

The determination of activation energy of flow helps to decide the temperature of various processes such as injection molding, calendaring, extrusion, etc. The activation energy ( $E_a$ ) of viscous flow derived from the Arrhenius type of relation is valid for fluids obey the power law.<sup>35</sup> The  $E_a$  calculated using the following equation:

$$\eta = Ae^{E/RT} \quad (6)$$

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

Figure 4 shows the plots of melt viscosity versus reciprocal of temperature at low (61.33 s<sup>-1</sup>) and high (613.3 s<sup>-1</sup>) shear rates.

(613.30 s<sup>-1</sup>) shear rates for PEA, LDPE, and their blends having different compositions. The activation energy was calculated using the slope (from Fig. 4) which is equal to  $E/R$ . The  $E_a$  as a function of different LDPE content in PEA/LDPE blends for high and low shear rate is shown in Figure 5(a,b). The observed activation energy of flow was low at low shear rates and high at high shear rates for all the

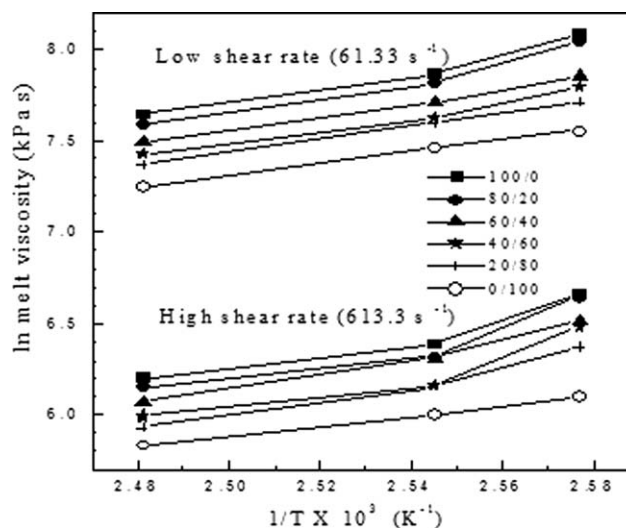
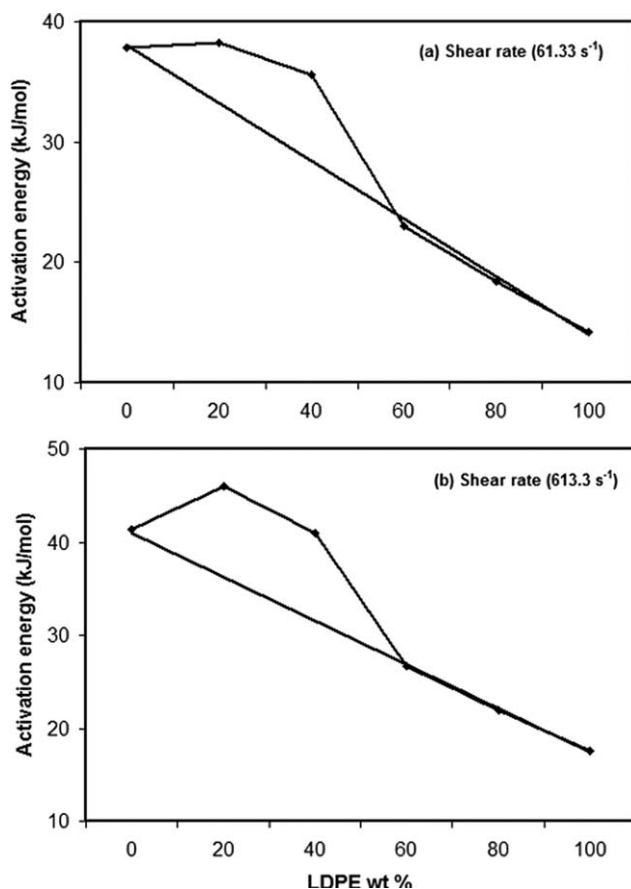


Figure 4 Plot of melt viscosity versus reciprocal of temperature for the pure components and PEA/LDPE blends at low (61.33 s<sup>-1</sup>) and high (613.3 s<sup>-1</sup>) shear rates.



**Figure 5** Plot of activation energy versus LDPE composition at: (a) low shear rate ( $61.33 \text{ s}^{-1}$ ) and (b) high shear rate ( $613.33 \text{ s}^{-1}$ ).

blends and their respective pure polymers. However, at both low and high shear rates ( $61.33$  and  $613.3 \text{ s}^{-1}$ ), PEA has showed higher activation energy of flow than that of LDPE. This phenomenon indicates that, PEA requires higher processing temperature than LDPE. The low activation energy in case of LDPE may be attributed to its typical structure and weak intermolecular forces of attraction.<sup>27–30,36</sup> The reduction in the activation energy of flow can be observed with increasing the amount of LDPE in the blends. The activation energies of the blends lie in between that of pure polymers. The

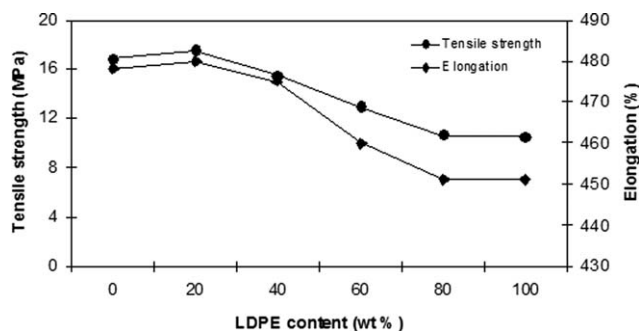
PEA/LDPE blends having 20 and 40 wt % LDPE has showed a positive deviation from the additive rule. The maximum synergy appears to exist with 80/20 and 60/40 (PEA/LDPE) blends at both low and high shear rates. The obtained data revealed that, the optimum composition of the blend fall in the range of 90/10–60/40 (PEA/LDPE).

The measured physico-mechanical properties of PEA, LDPE and their blends having different amounts of LDPE are given in Table III. The density of PEA ( $0.917 \text{ g cm}^{-3}$ ) was slightly higher than that of LDPE ( $0.909 \text{ g cm}^{-3}$ ). The densities of the blends lie between that of PEA and LDPE. The theoretical density of the blends were calculated using volume additive principle which states that,  $[d = w_1d_1 + w_2d_2]$  where “ $d$ ” is the density of the blend,  $w_1$  and  $w_2$  are the weight fractions of the constituents,  $d_1$  and  $d_2$  are the corresponding densities, respectively. The observed experimental densities of the blends containing 20 and 40% by weight LDPE was higher as compared to theoretical density. The tear strength of PEA and LDPE is  $103$  and  $88 \text{ kN m}^{-1}$  respectively. The tear strength values of PEA/LDPE blends as a function of varying LDPE content lies in the range of  $82$ – $95 \text{ kN m}^{-1}$ . The hardness of the blends lie in between that of PEA (98 Shore A) and LDPE (96 Shore A). The tensile strength of PEA, LDPE and their blends as a function of LDPE content is shown in Figure 6. The obtained tensile strength of PEA ( $16.8 \text{ MPa}$ ) was higher than that of LDPE ( $10.5 \text{ MPa}$ ). A reduction in tensile strength of the blends was noticed with increasing the LDPE content above 20% in the blends. The percentage elongation of the blend followed a similar trend as tensile strength. The percentage elongation of PEA and LDPE was 478 and 451%, respectively.

The tensile modulus of PEA, LDPE, and their blends as a function of different amount of LDPE is shown in Figure 7. The modulus values for immiscible blends will follow incompatible behavior in which the blend modulus would be below the rule of mixtures. In the case of mechanically grafted or compatibilized blends, the blends would exhibit either parallel or series behavior (depending on

**TABLE III**  
Physico-Mechanical Properties of PEA/LDPE Blends

PEA/LDPE compositions (wt %)	Density ( $\text{g cm}^{-3}$ )			Tear strength ( $\text{kN m}^{-1}$ )	Surface hardness (Shore A)
	Expt	Theo.	Error (%)		
100/0	0.917	–	–	103	98
80/20	0.912	0.9154	0.37	95	98
60/40	0.914	0.9138	0.02	87	97
40/60	0.907	0.9122	0.57	84	96
20/80	0.903	0.9106	0.84	82	96
0/100	0.909	–	–	88	96



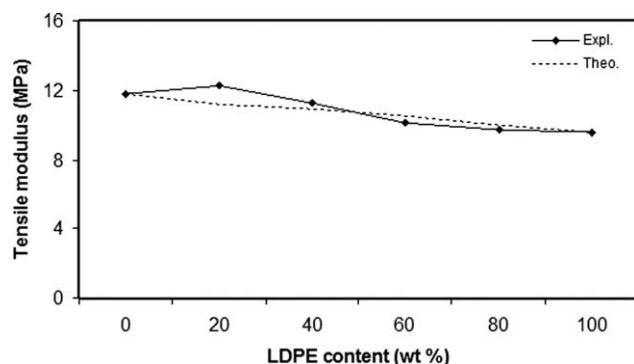
**Figure 6** Tensile strength and percentage elongation at break of PEA/LDPE blends as a function of LDPE content.

morphology) and will be quite close to the rule of mixtures. A positive deviation from the rule of mixture (or additive rule) will be possible unless orientation or other changes are induced in the polymers that produce increased stiffness in the test direction. PEA/LDPE blends approximately followed the rule of mixtures behavior, indicating good morphology development and a degree of mechanical grafting. The PEA/LDPE system, however, only reaches the rule of mixtures value at the cocontinuous composition ( $\sim 40\%$ ) and is below the proportional line to varying degrees at other compositions. The blends showed a negative deviation when the LDPE content is above 40%. The calculated percentage relative error between the experimental and theoretical values lies in the range from 0.84 to 4.0% for the blends containing different amount of LDPE from 0 to 100%, respectively.

## CONCLUSIONS

The following conclusions may be drawn from the present investigation:

1. The melt rheological behavior of PEA/LDPE blends has been investigated with reference to the effect of blend composition, shear rate, and temperature. On increasing shear rate and



**Figure 7** Tensile modulus of PEA/LDPE blends as a function of LDPE content.

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  $n < 1$  is an indication for characteristic pseudoplastic behavior of the materials.
3. The melt viscosity of PEA is higher than that of LDPE at all shear rates and temperatures. This is because of polar nature of PEA.
4. The activation energy ( $E_a$ ) of flow for PEA is higher than that of LDPE at all shear rates.  $E_a$  values increased with increasing in shear rate. Activation energy of PEA/LDPE blends decreases with increase in LDPE concentration in the blend. When LDPE content in the blend lies in between 20 and 40% by weight, a maximum activation energy of flow was observed. This may be due to maximum miscibility in this composition range.
5. The measured physicochemical properties of the PEA/LDPE blend lies between that of their pure polymer components.

The author (SR) would like to express his sincere gratitude to the Management of Indian Institute of Technology, Kharagpur, for awarding visiting Scientist Fellowship.

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