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Behavior and melt viscosity of polycarbonate/low density polyethylene blend in capillary flow

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Abstract : A knowledge of the variation of melt viscosity of thermoplastic polymers with both with both shear rate and temperature is considerable importance to plastics engineers as well as to polymer rheologist. The actual measurements of the melt viscosity at a large number of the temperatures and shear rates are frequently a tedious and time-consuming task. A technique has been developed, based upon the applicability of shear rate – temperature superposition, for predicting the flow curves for PC/LDPE (50/50) blend at different temperatures for experimental form data obtained at one temperature for the material in question. The experimental validity for superimposing flow curves at different temperatures along the log shear rate axis has been established for PC/LDPE (50/50) blend. the temperature dependence of the resultant shift factors has been determined for the system, and the method of utilizing this information to predict viscosities as a function of temperature and shear rate is discussed.

Keywords: Rheology, PC, LDPE, Shifting factor, Activation energy.

Introduction

A knowledge of the melt viscosity of thermoplastic polymers at various temperatures and shear rates is important to plastics engineers as well as to polymer rheologist who must design polymer processing equipment or who must specify the processing conditions for the fabrication plastics materials, such information is equally important to producers of polymeric materials and to the applied polymer rheologist. While considerable data may be found in the literature relating melt viscosity either to shear conditions or to temperature, it is frequently difficult or impossible to apply these data to predicting the flow behavior of the specific resin polymers of interest to given engineers or rheologists.

Moreover, the actual measurements of the melt viscosity at a large number of the temperature and shear rates is normally at tedious and time – consuming task.

Because of its optical clarity, excellent thermal resistance and high impact strength, a very useful engineering thermoplastic, polycarbonate (PC), is used in a wide range of industrials such as automotive and transportation, building and construction, packaging, medical, optical and ophthalmic, and optical media [1-7]. Recently, it is in widespread use in the audio, video and interactive software media. The PC capacities of the global main markets in 2001, including North America, South America, Europe, and Asia, was increased from

1.5 million m.t./yr in 1999 (Modern Plastics International, 2000) to 1.73 million m.t./yr (Chemical Week, 2000). The world capacity of PC is continuing to increase to 2.62 million m.t./yr in 2002 and the capacity is projected to grow to about 3.1 million m.t./yr by 2007 (Chemical Week, 2002). According to the same report, the end uses of PC in 2001 were 1.1 billion lb, and 20% of PC was used in optical media whose proportion was equal to automotives and window glazing. Form another side, the higher price and melt viscosity of PC limited its application in some applications, such problems could be solved using polymer blending technique in the molten state.

The aim of this work is to prepare PC/LDPE (50/50) in the molten state using the extrusion process and studying the rheological properties of this blend using capillary rheometer (Melt indexer type) and to generalize a method of predicting the flow curves of PC/LDPE (50/50) blend at any desired temperature from any experimental data for this system.

Experimental

Materials

Low density polyethylene (LDPE) (SABIC$^{®}$LDPE2308TN00) [density = 0.924 g/cm^3, *MFI* =7.5 g/10 min (190 °C/2.16 Kg)] was supplied by Sabic (KSA). Polycarbonate (PC) (Lexan OQ1026 resin – ***CD producing grade***) [density = 1.2 g/cm^3, *MVFR* = 11.0 cm^3/10min (250 °C/1.2 Kg)] was supplied by Sabic (KSA).

Blend preparation

Blends of PC/LDPE (50/50) was compounded using a laboratory scale single screw extruder (SSE) (*D* = 20 mm, *L/D* = 25). The screw speed was set at 50 rpm in the blends preparation, and the extruder temperature profile along the barrel was 180, 190, 200, 210 °C (from feed zone to die).

Melt rheology measurements

Rheological properties of the blends were studied using a constant pressure circular capillary rheometer. The melt is extruded by the use of dead weights (i.e. constant pressure) rather than constant plunger speed. This instrument, popularly known as the Melt Flow Indexer (*MFI*), is very popular in the thermoplastics industry due to its ease of operation and low cost, which more than compensates for its lack of sophistication. The parameter measured through the melt flow indexer contains mixed information of the elastic and viscous effects of the polymer. The rheological experiments were carried out at 210, 215, 220, 225 and 230 °C, and by using *L/R* = 15. The apparent shear rate (γ_a) is given by:

$$\gamma_a = \frac{4Q}{\pi R^3} \tag{1}$$

where R is the capillary radius, and *Q* is the volumetric flow rate.

The apparent shear stress (τ_a) is given by:

$$\tau_a = \frac{RP}{2L} \tag{2}$$

where *P* is the pressure at the capillary entrance, and *L* is the capillary length.

The apparent viscosity (η_a) is given by:

$$\eta_a = \frac{\tau_a}{\gamma_a} \tag{3}$$

The values of flow activation energy at a constant shear stress (E_τ) were determined by using Arrhenius equation form:

$$\eta_a = A.e^{\frac{E_\tau}{RT}} \tag{4}$$

where *A* is the consistency related to structure and formulation and *R* is the gas constant (8.314 J/mol. K).

Results and discussion

Rheological curves

Figure 1 represent the flow curves of the blend at the studied temperatures (210, 215, 220, 225, 230 °C), these curves are very important in determining the processing conditions of the material in the different processing techniques such as injection, extrusion and blow molding.

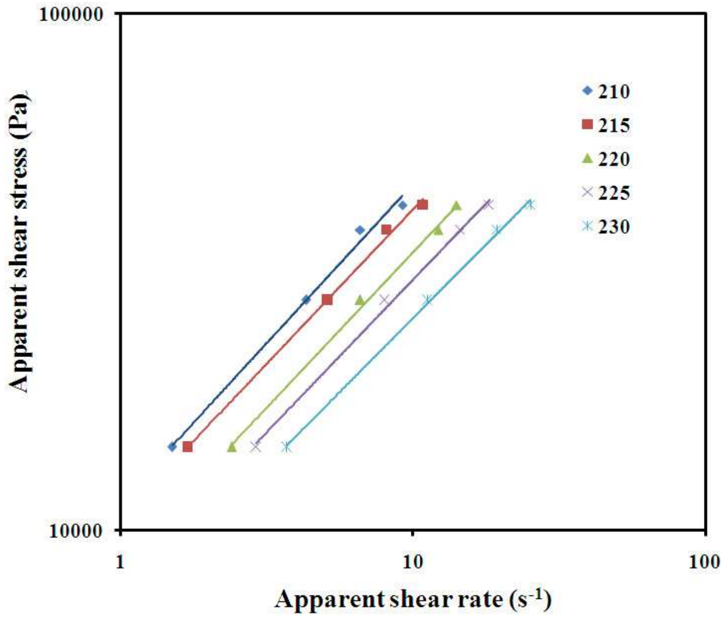

Fig. 1 Flow curves of PC/LDPE (50/50) at various temperatures and *L/R*=15

It can be seen that these curves apparently deviate from linear relationship inclining to the axis of shear rate, which means that the blend is pseudo plastic non-Newton fluids similar to most polymeric melts [8], and they obey the power law:

$$\tau = K.\gamma^{n} \tag{5}$$

where K is the consistency index and n is the non-Newtonian index, which can be calculated from the slope of the lines in Fig. 1.

$$n = \frac{d \log \tau}{d \log \gamma} \tag{6}$$

Figure 2 shows the viscosity curves of PC/LDPE (50/50) blend at various temperature and shear rates. It could be noted form Fig. 2, that the melt viscosity of the blend decreases with increasing shear rate at the various temperatures; this behavior confirms the last finding about the pseudo plastic manner of the blend.

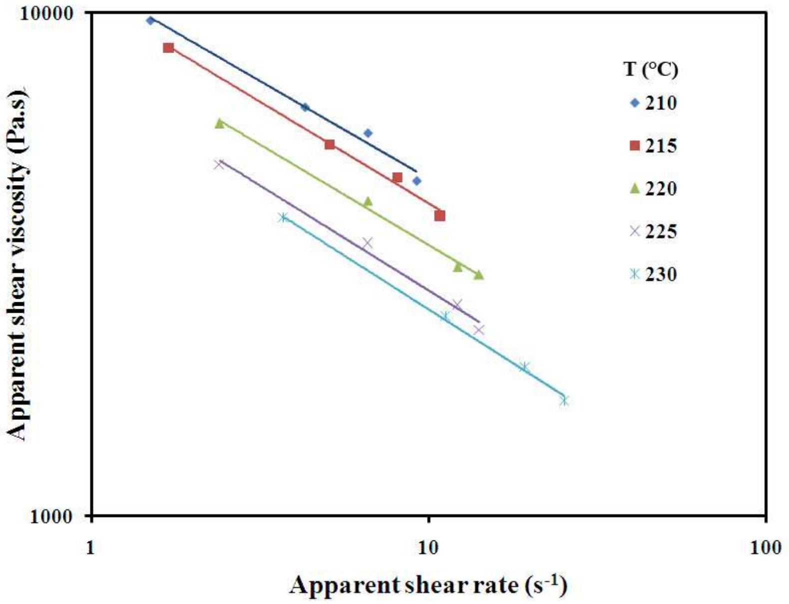

Fig. 2 Viscosity curves of PC/LDPE (50/50) blend at various temperatures and *L/R*=15

Shift factor determination

The flow curves may be shifted along the shear rate axis, *i. e.* at a constant shear stress for each point. The curve of superimpose on the single master cure corresponding to an arbitrarily was chosen as a reference temperature. The arbitrary reference temperature used here for the blend was 220 °C. The horizontal shift factor (a_T) were obtained by choosing the shear stresses (14475, 27900, 38125, 42600 Pa).

Flow curves and shifting the corresponding points on the flow curves at the different temperatures were coincided at these shear stresses, the values of a_T were calculated using the following equation [9-12]:

$$a_T = \left(\frac{\gamma_{a(220)}}{\gamma_{a(T)}}\right)_\tau \tag{7}$$

where $\gamma_{a(220)}$ is the reference shear rate (at 220 ºC), and $\gamma_{a(T)}$ is the shear rate at T. a_T values were calculated at each shear stress. It could be noted that the last equation may be rewritten as following:

$$a_T = \left(\frac{\eta_{a(T)}}{\eta_{a(220)}}\right)_\tau \tag{8}$$

where $\eta_{a(220)}$ is the reference shear viscosity (at 220 ºC), and $\eta_{a(T)}$ is the shear viscosity at T. The values of the shifted factors are summarized in Table 1. In order to test the applicability of these shear rate-temperature superposition shift factors. The a_T values presented in table 1 were used to contrast the master flow curve at 220 ºC utilizing equation (8).The resultant master curve shown in Fig. 3 clearly illustrates the validity of the superposition method.

Table 1 Experimental rheolgical data

T (°C)	τ_a (Pa)	γ_a (s^{-1})	$\gamma_{a(200)}$ (s^{-1})	a_T	$a_T \cdot \gamma_a$ (s^{-1})	η_a (Pa.s)
210	14475	1.5	2.4	1.6	2.4	9650
	27900	4.3	6.6	1.534884	6.6	6488.372
	38125	6.6	12.2	1.848485	12.2	5776.515
	42600	9.2	14.1	1.532609	14.1	4630.435
215	14475	1.7	2.4	1.411765	2.4	8514.706
	27900	5.1	6.6	1.294118	6.6	5470.588
	38125	8.1	12.2	1.506173	12.2	4706.79
	42600	10.8	14.1	1.305556	14.1	3944.444
220	14475	2.4	2.4	1	2.4	6031.25
	27900	6.6	6.6	1	6.6	4227.273
	38125	12.2	12.2	1	12.2	3125
	42600	14.1	14.1	1	14.1	3021.277
225	14475	2.9	2.4	0.827586	2.4	4991.379
	27900	8	6.6	0.825	6.6	3487.5
	38125	14.5	12.2	0.841379	12.2	2629.31
	42600	18.2	14.1	0.774725	14.1	2340.659
230	14475	3.7	2.4	0.648649	2.4	3912.162
	27900	11.2	6.6	0.589286	6.6	2491.071
	38125	19.3	12.2	0.632124	12.2	1975.389
	42600	25.2	14.1	0.559524	14.1	1690.476

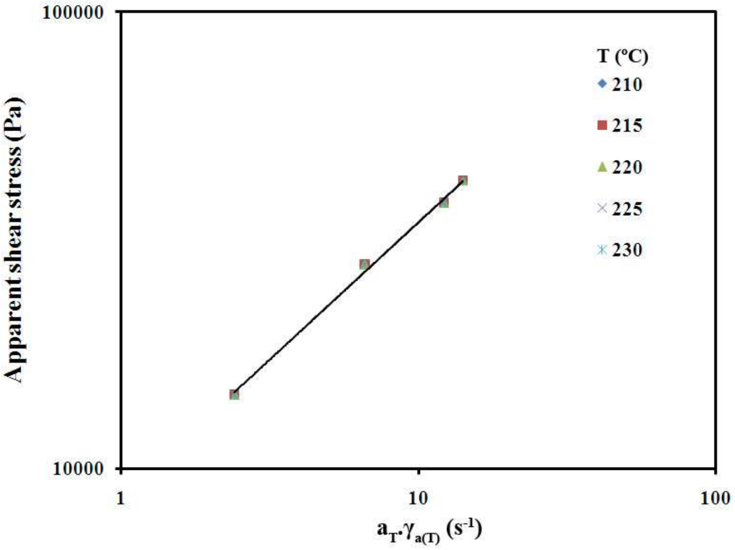

Fig. 3 The master curve

Moreover, it is not even necessary that the known data set be available at the reference temperature since, assuming data to be known at T_1 and desiring data at T_2, elimination of γ_a(ref) from equation 7

$$\gamma_a(T_2) = a_{T1}\gamma_a(T_1) / a_{T2} \tag{9}$$

The temperature dependence on the shift factors was next investigated together with a simple exponential or Arrhenius type equation of the form:

$$a_T = B.e^{\frac{E}{R.T}} \tag{10}$$

Where T is the absolute temperature and E is the shift factor activation energy and R is the general gas constant. The last equation could be rewritten as following:

$$Loga_T = LogB + \frac{E}{2.303R.T} \tag{11}$$

This could be also simplified as following:

$$Loga_T = B' + \frac{C}{T} \tag{12}$$

Where:

$$C = \frac{E}{2.303R} \tag{13}$$

Figure 4 shows the linear plots of $Loga_T$ versus *1/T* at constant shear stresses. The slop of these lines gives the shift factor activation energy.

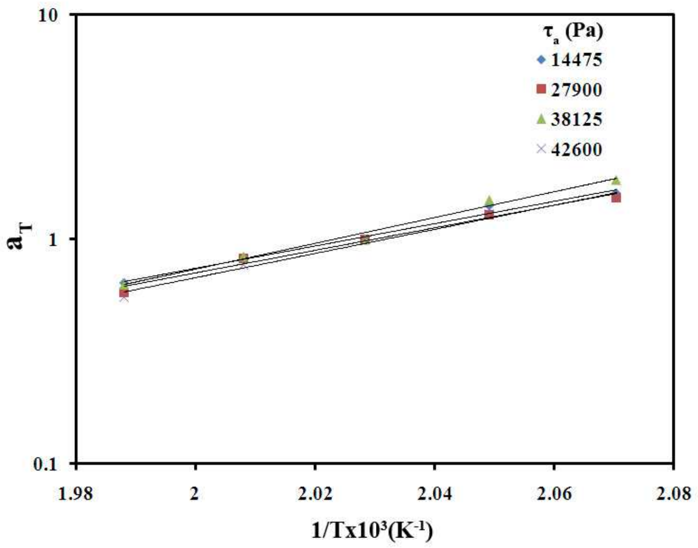

Fig. 4 shift factor versus *1/T* at constant shear stresses

The values of the shift factor activation energy at constant shear stresses are listed in Table 2:

Table 2 Shift factor activation energy at constant shear stresses (*E*)

Shear stress (Pa)	14475	27900	38125	42600
***E* (kJ/mol)**	95.86	96.52	109.99	103.59

Also, the flow activation energy at constant shear stresses from the apparent viscosity values (E_τ) was calculated using Arrhenius form equation (equation 4), Fig 5 shows the plots of apparent viscosity versus *1/T* at constant shear stresses:

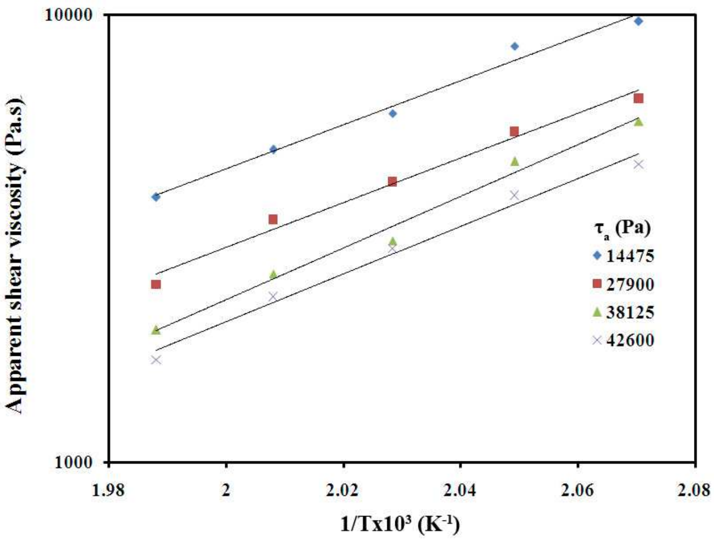

Fig. 5 Apparent shear viscosity versus *1/T* at constant shear stresses

It could be seen from Fig. 5, that the plots are linear and they obey the Arrhenius type equation (equation 4), the flow activation energy could be calculated from the slope of these line. The values of the flow activation energy at constant shear stresses (E_τ) are listed in Table 3:

Table 3 Flow activation energy at constant shear stresses (E_τ)

Shear stress (Pa)	14475	27900	38125	42600
E_τ (kJ/mol)	94.53	95.61	110.16	102.34

It is well known that the value of flow activation energy reflects the temperature-sensitivity of viscosity, so the more E_τ was the more sensitive the behavior of blends were to the temperature. It could be noted from Tables 1 and 2 that both flow activation energy (E_τ) and shift factor activation energy (E) has nearly the same values, which confirms the used prediction method.

Conclusion

The general validity of a shear rate- temperature superposition of log shear stress VS log shear rate flow curves have been demonstrated for PC/LDPE (50/50 wt%). The temperature dependent on the resultant shift factors has been shown to be capable of representation by Arrhenius type equation of the generic system. Moreover the technique of shear rate- temperature superposition and the shift factor temperature dependence to predict flow curve at various temperature allow us to determine the shear stress that is necessary for desired temperature at shear rate constant.

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