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Rheological and mechanical properties of ABS/PC blends

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Abstract

Acrylonitrile-Butadiene-Styrene (ABS), polycarbonate (PC) and their alloys are an important class of engineering thermoplastics that are widely used for automotive industry, computer and equipment housings. For the process of recycling mixtures of ABS and PC, it is desirable to know how sensitive the blend properties are to changes in compositions. It was for this reason that blends of virgin ABS and virgin PC at five different compositions, namely, 15%, 30%, 50%, 70% and 85% by weight of ABS were prepared and characterised by rheological and mechanical measurements. Rheological properties of these blends in steady, oscillatory and transient step shear and mechanical properties, namely, tensile strength, elongation-at-break and Izod impact strength are reported. The results show that PC behaves in a relatively Newtonian manner, but ABS exhibits significant shear thinning. The ABS-rich blends show a trend that is similar to that of ABS, while PC-rich blends, namely 0% and 15%, exhibit a nearly Newtonian behaviour. However, at a fixed shear rate or frequency, the steady shear or the dynamic viscosity varied respectively in a non-monotonic manner with composition. Except for 15% blend, the viscosities of other blends fall into a narrow band indicating a wide-operation window of varying blend ratio. The blends exhibited a lower viscosity than either of the two pure components. The other noticeable feature was that the blends at 70% and 85% ABS content had a higher G' than pure ABS, indicating an enhancement of elastic effect. The tensile yield strength of the blends followed the 'rule of mixtures' showing a decreasing value with the increase of ABS content in PC. However, the elongation-at-break and the impact strength did not appear to obey this 'rule of mixtures,' which suggests that morphology of the blends also plays a significant role in determining the properties. Indeed, scanning electron micrographs of the fracture surfaces of the different blends validate this hypothesis, and the 15% blend is seen to have the most distinct morphology and correspondingly different behaviour and properties.

Keywords : ABS/PC blends, oscillatory and steady shear, heterogeneous morphology, mechanical properties

1. Introduction

ABS, PC and their blends are high impact resistant materials with good ductility, durability and high dimensional stability. They are widely used in the automotive industry and also for making computer and equipment housings. With continuous improvement in computer and electronic equipment, a large volume of plastic is discarded when the equipment is replaced, and this ends up in landfills. These materials are not biodegradable and since no viable recycling process appears to exist for these materials, they pose an environmental hazard. While recycling of single thermoplastic polymeric materials, particularly high density polyethylene (HDPE) used in milk containers, low density polyethylene (LDPE) used in garbage bags and polyethylene terephthalate (PET) used in beverage bottles, has developed to a reasonable level, recycling of ABS/PC thermoplastics has not progressed to the same extent and has remained somewhat unattractive. This is partly because the volume of these materials was not very large and partly because these materials are made from more than one polymer. The supply situation is now changing, but these plastics are still mixed with other polymers and materials. Generally, mixed recycled polymers exhibit poor mechanical properties compared to the pure components and show unpredictable rheological properties. They have, therefore, traditionally been used in low-value applications such as flower pots and park benches where mechanical property requirements are not very stringent. For a recycle process to be economically successful, though, the recycled materials need to be employed in high value applications or sold

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at a premium.

In their earlier work, Liang and Gupta (2000) studied the rheological and mechanical properties of a recycled PC blended with virgin PC; these authors reported that separated PC could be added to pure PC up to the 15% level without significantly altering the properties of pure PC.

This study is aimed at characterising alloys of ABS and PC with a view to developing commercially viable products through blending these two materials. The ultimate objective is to develop alloys containing as much recycled plastics as feasible. The study of the properties of the blends of virgin ABS/PC plastics provides a useful basis for realising and maximising product development from recycled materials.

2. Experimental procedure

2.1. Preparation of ABS/PC blends

A Brabender Twin Screw extruder was used for the blending. In terms of process conditions, though, one has to contend with the fact that PC has a glass transition temperature of 150°C while that for ABS is 95°C. Thus, the nominal extrusion temperatures for PC is 300°C but that for ABS is only 200°C. Getting the correct extrusion temperature for the preparation of ABS/PC blends between this range was important so that blends were neither underplasticised due to low temperature nor degraded due to high temperature. Consequently, different temperature profiles were used for extrusion followed by tests on the properties of the blends for thermal, mechanical and rheological stability. After a careful trial and error check, an optimised temperature profile was chosen for the preparation of the blends. Five different compositions of ABS/PC blends, namely, 15%, 30%, 50%, 70% and 85% by weight of ABS and virgin ABS (100%) and virgin PC (0%) were prepared for rheological and mechanical testing.

Lexan 101, a commercial grade virgin PC, and *Cycolac GPM 5500-1000*, a virgin ABS, both supplied by GE Plastics were used for this study. The relative densities of PC and ABS were 1.2 and 1.05 respectively. The PC had a molecular weight (MW) of 33050 and a polydispersity index (P.I.) of 2.3. Before extrusion, the PC and ABS pellets were dried at 90°C in a vacuum oven overnight.

It should be noted that PC is a homogeneous, single-phase polymer while ABS is a heterogeneous, two-phase terpolymer consisting of a dispersed rubbery phase made up of polybutadiene (PB) rubber grafted with styrene-acrylonitrile (SAN) and then dispersed in a continuous plastic phase of more SAN. Blending PC into ABS changes the ratio of plastic and rubber phase components, altering the structural morphology of the system and yielding different rheological and mechanical properties.

2.2. Test specimen preparations

The blends were extruded into a water bath, and the extruded strands were pelletized by a C.W. Brabender pelletizer; the pellets were then dried in the vacuum oven overnight. They were compression moulded into sheet specimens by a hydraulic press at a temperature of 200°C under a load of 15 tons for 2.5 minutes. Circular disks of 25 mm diameter and 1 mm thickness were cut from the sheets for rheological tests.

The appropriate specimens for mechanical testing, namely tensile test and impact tests, were also compression moulded using the hydraulic press. Moulds for the test sample were designed and fabricated as per the standard specimen dimensions. Dried pellets were pressed under a load of 15 tons for 5 minutes for tensile specimens and 7 minutes for the impact test specimens.

The moulded specimen for tensile tests was a dog-bone shaped with a total length of 110 mm, an effective length of 35.56 mm with an end width of 25 mm and test width of 6.5 mm. The impact specimen had a length of 63.5 mm with a width of 4.8 mm, a thickness of 12.7 mm and a notch at the one side centre in the direction of width with a residual thickness of 10.1 mm at the notch.

2.3. Scanning electron microscopy

Moulded specimens for impact testing were notched and then fractured at room temperature by the impact tester. The fractured surface was gold coated using a sputterer. A Hitachi S 4700 model scanning electron microscope was then used to study the fractured surfaces.

3. Rheological characterisation

Rheological measurements on these blends were carried out on a Rheometrics Mechanical Spectrometer, model RMS800 at a temperature of 225°C. The test modes included strain sweep at a 1 rad/s frequency to establish the linear strain range; dynamic frequency sweep at a strain of 10% to determine dynamic linear viscoelastic moduli G' G"; steady shear rate and transient (unsteady) step shear rate to evaluate viscous and elastic response; and dynamic time sweep in the linear range (at a frequency of 1 rad/s and a strain rate of 10%) to determine the build up or breakdown of network structure. Prior to testing, the samples were heated to 225°C for duration of 15 to 20 minutes until the residual stress was released and sample thermal equilibrium was attained.

3.1. Linear viscoelasticity

The response from the strain sweep test showed that all the blends exhibited a linear behaviour up to a strain magnitude of 100%; so both the storage modulus and the loss modulus and the dynamic viscosity are independent of strain amplitude in this region.

The linear viscoelastic data are viewed to be important as they provide much useful information to help understand the more difficult non-linear properties. Besides, it should not be forgotten that for any type or magnitude of deformation all materials should satisfy the linear viscoelastic response in the limit of small deformation.

The properties of pure PC or ABS depend on the molecular weight and their structures. The properties of the blends however are likely to be different, and we focus on their behaviour and attempt to explain how these properties depend on or differ from those of the pure materials.

Fig. 1 presents the response of storage and loss moduli against frequency. As far as the loss modulus G'' is concerned, it is seen that pure PC (0%) has a much higher modulus indicating higher energy dissipation compared to the various blends. With the addition of 15% ABS, the blend exhibits the lowest modulus at lower frequencies but rises above the corresponding data for the other blends to attain the highest magnitude at higher frequencies. Both the 15% and 30% blends deviate from the ABS-rich samples at high frequencies and show a higher modulus. The responses for 50%, 70% and 85% are ABS dominant and similar to pure ABS (100%), and they converge into a narrow band in the frequency range of 0.2 to 2 rad/s.

Fig. 1 also shows the storage modulus response. Pure PC (0%) appears to be much more elastic than pure ABS,

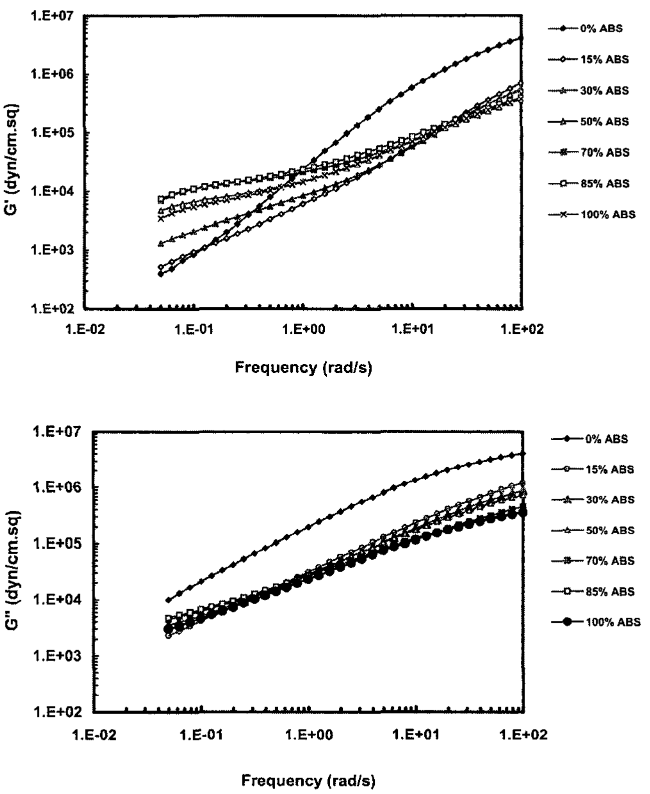

Fig. 1. Storage (G') and loss (G'') moduli vs. Frequency at 10% strain and 225°C.

showing the lowest storage modulus at low frequencies but then reaching the highest value at higher frequencies. All other blends behave similar to pure ABS showing a near plateau at low frequencies and falling into a narrow band at high frequencies. The plateau height is higher than the G'' value at the same frequency and is seen to increase with increasing ABS content. However, the 70% and 85% blends show the highest modulus at low frequencies indicating that they are capable of storing more energy than pure ABS. Exhibition of this solid-like behaviour at low frequency is not uncommon for heterogeneous systems although such behaviour has not been completely explained (Marie-Pierre Bertin, 1995).

These dynamic mechanical responses of ABS/PC blends do not appear to be systematic deviations from the behaviour of either pure PC or pure ABS. In other words, the behaviour does not follow the 'rule of mixtures', which should show a predictable increasing or decreasing trend with increasing or decreasing blend content. Quite clearly, the addition of PC into ABS changes the ratio of the plastic phase component (PS, AS) to the rubbery phase component (PB, BS, BA), altering the interaction between these phases. This, in turn, changes the structural morphology of the system, resulting in varying rheological and mechanical properties. Previous works by Yuji Aoki (1986) and Marie-Pierre Bertin *et al.* (1995) indicate that the viscoelastic behaviour of ABS depends strongly on the rubber phase or more precisely on the degree of grafting of the rubber particles. Therefore, the responses of the ABS/PC blends are likely to be influenced by the changed rubber phase as well as the plastic phase of the blends.

In order to verify the above hypothesis, the morphology of the ABS/PC blends was examined with the help of a scanning electron microscope. Fig. 2 shows micrographs of the fractured surfaces with respect to composition of the various blends. The difference between the ABS rich phases and PC rich phases is clear. For blends containing 15% ABS up to 50% ABS, we can see that the ABS phase appears as spherical inclusions in the PC phase matrix. Also, at the fracture surface, there seems to be slippage between the PC and ABS phases as these two phases are not bonded together, especially at low ABS content. However, ABS-rich blends look more uniform and the micrographs resemble the fracture surface of pure ABS. Thus, it is not surprising that the 15% ABS blend seems to stand out the most from the other blends.

The ratio of the loss modulus to the storage modulus, known as the loss tangent value, provides a useful measure of the relative magnitudes of energy storage and dissipation of energy. From Fig. 3, it can be determined that PC has the highest loss tangent at nearly all frequencies; this is not surprising considering that this polymer is known for its excellent impact strength. Conversely, the 70% and 85 % blends have the lowest loss tangent, and thus the lowest impact strength.

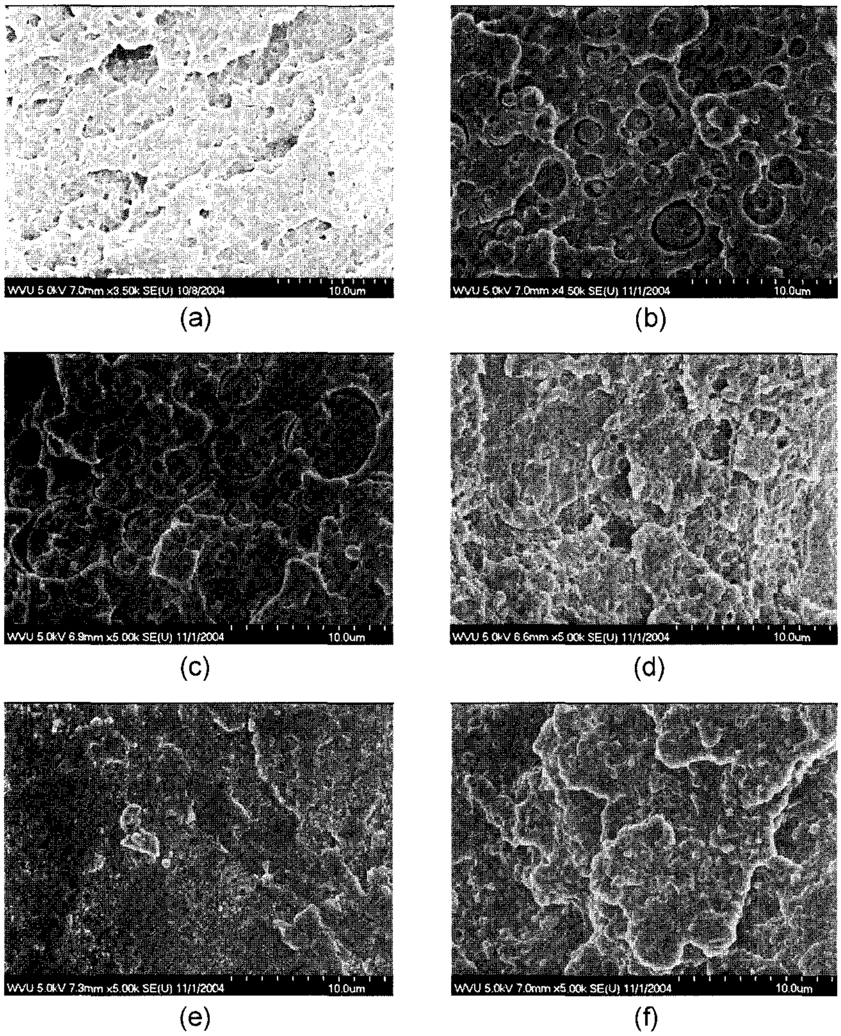

Fig. 2. Scanning electron micrographs of fracture surfaces of PC/ABS blends: (a) ABS (b) 15% ABS (c) 30% ABS (d) 50% ABS (e) 70% ABS and (f) 85% ABS.

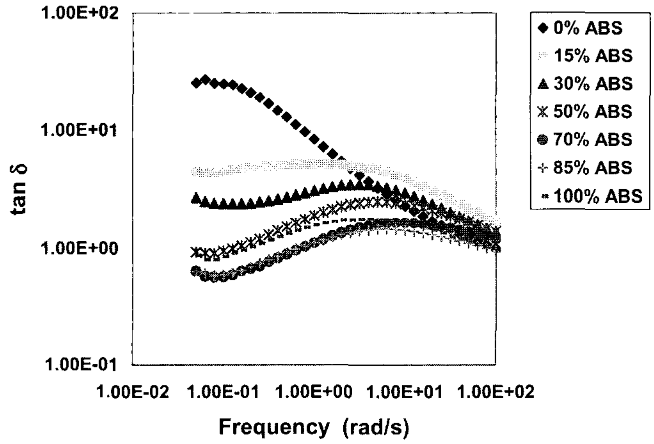

Fig. 3. tan δ vs. Frequency at 10% strain and 225°C.

3.2. Complex viscosity and shear viscosity

Results for the absolute value of the complex viscosity against frequency are presented in Fig. 4. Pure PC has the highest viscosity, and, as is known, it shows a weak shear thinning effect while ABS shows a significant shear thinning behaviour. The shear-thinning viscosity behaviour of ABS carries over to the blends, and their viscosity is seen to decrease substantially with the addition of ABS into PC. The only exception is the 15% sample, which shows a relatively constant viscosity behaviour that is similar to pure PC; however, the magnitude of the viscosity is significantly lower than that of pure PC, and, in fact, the 15% blend has the lowest zero shear viscosity among all the blends. The response of the 30% ABS sample is seen to represent a transition from weak shear thinning (PC) to strong shear thinning (ABS). The decrease in η_0 (zero shear rate viscosity) upon adding 30% and 50% ABS is about 38% and 27% respectively. Thus, processability would seem to improve leading to a power saving when ABS is added to PC. A possible reason for the decrease of viscosity with addition of ABS may be solvation of the

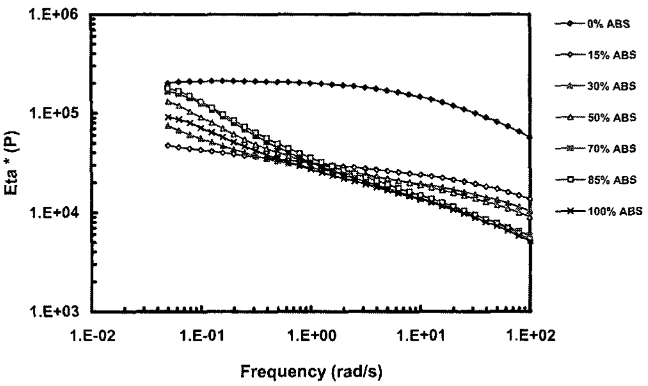

Fig. 4. Dynamic viscosity vs. Frequency.

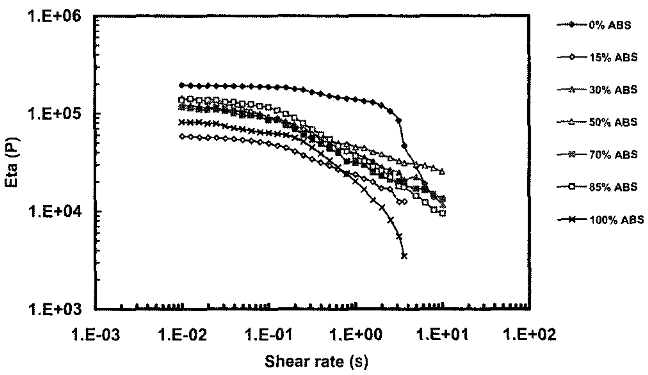

Fig. 5. Shear viscosity for ABS/PC blends at 225°C.

highly entangled structure of molecules of PC by the ABS molecules (Babbar and Mathur, 1994).

The steady shear viscosity responses are shown in Fig. 5. From these data and the data in Fig. 4, it is seen that the magnitude of the complex viscosity for some blends nearly equals the steady shear viscosity at corresponding values of low frequency and small shear rate. From continuum mechanics, it is known that both complex and shear viscosity approach η_0 as the frequency (ω) and the shear rate ($\dot{\gamma}$) approach zero. This is seen to be true for most blends, although some difference in their values is noticeable. It then can be said that the Cox-Merz rule ($\eta(\dot{\gamma}) = \eta^*(\omega)$) is loosely followed by nearly all the blends. It is also known that the rate of decrease in dynamic viscosity at large frequency is different from the rate of decrease of shear viscosity at large shear rates (Bird *et al.*, 1987). For example, unlike in dynamic tests, 15% sample exhibits a significant shear thinning behaviour in steady shear viscosity. This is, however, expected and plausible as these deformations appear to occur beyond the linear range and also these blends were found to show some time-dependent properties discussed later in Section 3.3. It is however not obvious if some discrepancy in magnitude observed can be attributed to this effect.

The viscosity data from Fig. 5 can be used to further

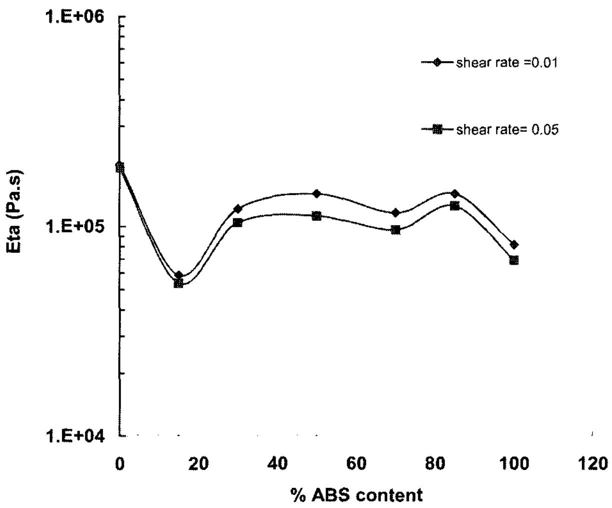

Fig. 6. Steady shear vs. ABS content at different shear rate.

analyse the shear steady state behaviour of the complex ABS/PC blends. In particular, steady state viscosity data at lower shear rate were plotted against ABS content as shown in Fig. 6. The shear viscosity data exhibit a wave-like response as the ABS content increases showing two maximum peaks at 50% and 85% composition. The viscosity of the blends falls with the addition of ABS composition reaching a minimum at 15% composition. It then increases monotonically with the increase of ABS content showing a maximum peak at 85%. This inconsistent feature is not totally unexpected and may be attributed to the complex ABS/PC system and their multiphase morphology.

3.3. Dynamic time sweep

The dynamic viscosity tests were conducted against time for sample stability. It was found that pure PC and pure ABS appear quite stable over a test period of more than one hour. With an increase of the ratio of the mix (PC or ABS), the blends exhibited material softening over time. This effect was seen to be the most pronounced in 30% and 50% blends. This feature illustrates some potential difficulty in conducting valid rheological tests over an extended period of time.

3.4. Step shear rate transient response

A transient step-shear-rate test was conducted to determine the viscoelastic linear and non-linear responses over time from a start up deformation. Tests were conducted for a range of shear rates. The most important feature of these data was the existence of stress overshoot followed by an eventual plateau, which would indicate a steady state value. It is however, noted that for some blends the steady value appeared to give way to a decaying trend which can be attributed to sample distortion. Pearson and Kiss (1987) suggested that the overshoot occurs because polymer strands are stretched during brief period after start-up of shearing. The maximum overshoot appeared to occur at a roughly constant value of the imposed strain, $\dot{\gamma}t$ of about 2. This appears to satisfy the Doi-Edwards equation (Larson, 1988). The magnitude of the overshoot was seen to be increasing with the increase of ABS content in pure PC. This can be attributed to the fact that ABS exhibits more elastic than viscous behaviour (Marie-Pierre Bertin, 1995).

4. Mechanical properties

The mechanical properties evaluated were tensile strength, elongation-at-break and Izod impact strength. Tensile tests were conducted on an Instron machine model 8501 at room temperature. The specimen was stretched at a constant rate of 0.2 in/min. The results for yield strength and fracture strength are presented in Fig. 7, and results for elongation-at-break are given in Fig. 8 as a function of ABS content. The results in Fig. 7 show that the strength properties nearly follow the rule of mixtures, with strength decreasing with increasing ABS content in PC. However, Fig. 8 demonstrates that the elongation-at-break is non-

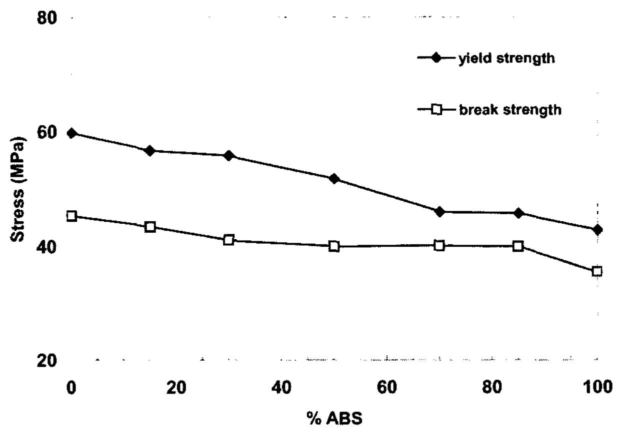

Fig. 7. Yield strength and break strength as a function of ABS content.

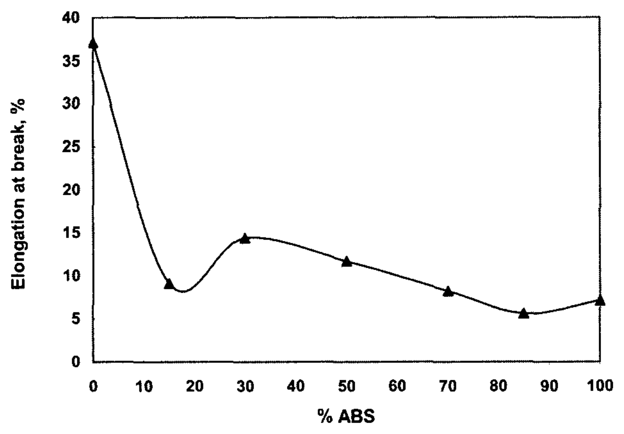

Fig. 8. Effect of ABS content on elongation at break.

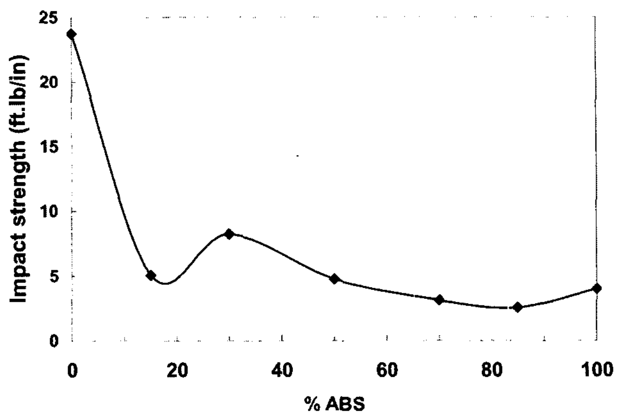

Fig. 9. Effect of ABS content on impact strength.

monotonic with ABS content, and, overall, ABS/PC blends have a much smaller elongation compared to pure PC.

The Izod impact test was conducted on Satec Impact Testing Machine equipped with a 2 fl-Ib Izod pendulum and the specimen holder. The test calculates the energy required to break a specimen when the pendulum hits the specimen on impact. The impact tests were carried at room temperature of 25°C, and their results are presented in Fig. 9.

The result of the Izod impact testing shows the same features as shown by elongation-at-break. While PC and blends with lower ABS content exhibit high impact strength, ABS rich blends have much smaller impact strength than PC.

5. Conclusions

Rheological and mechanical properties of the ABS/PC blends are evaluated here to developing a strategy of recycling these materials. A recycling process is viable if these materials can be employed in high value applications or in new product development. The results of this study show that the processability of PC can be improved by the addition of ABS. The shear thinning effect of ABS rich blends provides a significant power saving in the processing of these blends. Furthermore, except for the 15% blend, the viscosities of other blends fall into a narrow band which offers a wide range of operation window and room to mask possible variations in properties of recycled polymers. The results of the tensile and impact tests also indicate an improvement in the processability of PC by adding ABS. The tensile yield strength results for the blends follow the 'rule of mixtures' showing a decreasing value with the increase of ABS content. However, neither the impact strength nor the elongation-at-break obeys this rule, and they both show much lower magnitudes than that of virgin PC. These results are important as they indicate the possibility of reusing recycled polymers at a much higher percentage through blending ABS/PC.

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