Properties of compatibilized nylon 6/ABS blends
Part I. Effect of ABS type

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Abstract

Blends of nylon 6 with four acrylonitrile–butadiene–styrene (ABS) materials were investigated over a range of compositions using an imidized acrylic (IA) polymer as the compatibilizing agent. The IA material is miscible with the SAN matrix and is able to react with the amine end groups of nylon 6 during melt processing. The effects of ABS type and blend composition on the morphological, rheological and mechanical behavior of these blends were explored at a fixed (5 wt.%) compatibilizer content. In general, incorporation of the IA polymer can lead to super tough blends using a broad range of ABS materials. Of the ABS materials used, those with a monodisperse population of butadiene rubber particles and low melt viscosity were found to generate blends with superior low temperature toughness compared to those with broad particle size distributions and higher viscosity. In several compatibilized blends, increasing the ratio of nylon 6 to ABS improved the modulus and yield strength and did not significantly affect the room temperature impact strength up to a certain level; however, the ductile-to-brittle transition temperatures of these blends were quite sensitive to the amount of rubber in the blend, and increased steadily with increasing nylon 6/ABS ratio. The effect of compatibilizer content on blend properties was determined at a fixed ratio of nylon 6 to ABS. Super tough materials can be generated using very small (0.5 wt.%) amounts of the imidized acrylic polymer. Further increases in compatibilizer content did not significantly affect the room temperature impact strength, but did improve the low temperature toughness. When processing characteristics are considered, there is no justification to use more than about 2 wt.% of this compatibilizer. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Nylon; Acrylonitrile–butadiene–styrene; Imidized acrylic polymer

1. Introduction

Polyamides, or nylons, are an attractive class of engineering polymers due to their excellent strength and stiffness, low friction, and chemical and wear resistance [1]. However, they are highly notch sensitive, that is, they are often ductile in the unnotched state, but fail in a brittle manner when notched. In addition, polyamides tend to be brittle at low temperatures and under severe loading conditions. Fortunately, the inherent chemical functionality of nylons makes them an attractive candidate for modification. Numerous articles describe the approaches that improve the toughness of these materials by reacting a polymer containing an appropriate chemical functionality with the acid or amine end groups of the polyamide during melt processing [2–36].

There is a large body of literature, which describes the use of elastomers to toughen polyamides. There are numerous examples of functionalized hydrocarbon elastomers [2–18] and core-shell impact modifiers [19–21], which have been used as toughening agents. The morphology of the blend is a primary issue in the extent of toughening achieved and is influenced by several factors, including the functionality of the compatibilizer, rheological characteristics, processing conditions, etc.

Blends of polyamide and acrylonitrile–butadiene–styrene (ABS) materials are of significant commercial interest. ABS materials consist of a butadiene rubber embedded within a matrix of styrene–acrylonitrile copolymer (SAN), some of which is chemically grafted to the rubber and are generally noted for their excellent toughness, dimensional stability and relatively low cost. Incorporation of ABS is somewhat similar to independently dispersing rubber and rigid phases in the nylon 6 matrix; the rubber phase can impart low temperature toughness to the blend, while the rigid phase provides stiffness. Recent studies from this laboratory has shown that incorporating such combinations of rubber and rigid phases in a nylon 6 matrix can generate blends with an excellent balance of stiffness and toughness.
<table>
<thead>
<tr>
<th>Designation used here</th>
<th>Supplier designation</th>
<th>Rubber content (wt.%)</th>
<th>Mol. Weight (g/mol)</th>
<th>AN content (wt.%)</th>
<th>Graft ratio</th>
<th>Rubber particle size (μm)</th>
<th>Brabender torque (N m)</th>
<th>Izod impact strength (J/m)</th>
<th>Source</th>
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<td>ABS-45-a&lt;sup&gt;c&lt;/sup&gt;</td>
<td>Starex</td>
<td>45</td>
<td>$M_n = 35,000$&lt;sup&gt;d&lt;/sup&gt;</td>
<td>25</td>
<td>0.45</td>
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<td>433</td>
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<td>393</td>
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<td>BL-65</td>
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<td>0.40</td>
<td>$0.05, 0.40$</td>
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<td>Capron 820&lt;sup&gt;f&lt;/sup&gt;</td>
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<td>EXL 4140</td>
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<td></td>
<td>9.8</td>
<td>30</td>
<td>Rohm and Haas</td>
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</table>

<sup>a</sup> Values taken at 240°C and 60 rpm after 10 min.

<sup>b</sup> Room temperature notched Izod impact.

<sup>c</sup> All the ABS grades were designated by their weight percent rubber.

<sup>d</sup> Molecular weights are for free SAN.

<sup>e</sup> Bimodal particle size distribution.

<sup>f</sup> The designation of this material has recently been changed to B73WP.
There is significant interest in developing nylon/ABS blends, with the goal being to combine the desirable properties of the blend constituents. However, simple blends of ABS with polyamides have poor mechanical properties unless a compatibilizing agent is used, unlike blends of ABS with polycarbonate [37] and with poly (butylene terephthalate) [38].

Several approaches to the reactive compatibilization of nylon/ABS blends have been reported in the recent literature [23–36]. The preferred compatibilization strategy has been to incorporate a functional polymer capable of reacting with the amine end groups of the polyamide that is also miscible with the SAN phase of ABS. Recent work by Majumdar et al. has demonstrated the utility of imidized acrylic (IA) polymers as compatibilizers for nylon 6/ABS blends [31,32]. These IA materials, which can be miscible with SAN [39], also contain acid and anhydride functionalities capable of reacting with nylon 6. The focus of this article is to further investigate the properties of nylon 6 blends with a number of emulsion-made ABS materials (grafted rubber concentrates) using an imidized acrylic polymer as a compatibilizer.

The effectiveness of ABS as an impact modifier can be governed by several variables such as concentration, rheology, rubber phase characteristics (particle size and distribution, crosslink density), as well as the amount of SAN grafted to the rubber, AN content in the SAN, and the molecular weight of the free SAN. In this study, a range of ABS materials is used to explore the effect of some of these variables on blends with nylon 6. Many of the materials used here lead to blends with far superior low temperature toughness than observed previously [31]. The current study focuses primarily on blends with a fixed compatibilizer (imidized acrylic polymer = IA) content of 5 wt.% and a standard processing history (one extrusion step and one injection molding step). An accompanying article addresses the effects of compatibilizer type and content, as well as some effects of processing history on the properties of nylon 6/ABS blends.

2. Experimental

Table 1 summarizes pertinent information about the materials used in this study including four ABS materials with different characteristics, designated by their rubber content expressed as a weight percent. All of the latter are emulsion-made SAN grafted rubber concentrates, i.e. high rubber contents, that are usually blended with additional SAN to form the final ABS product. Several previous reports from this laboratory have employed the material designated here as ABS-50, which was previously designated as BL-65 in some other reports [30,31,36,40,41]. The “ABS-45” series of materials became available to this laboratory more recently. Two of these (ABS-45-a and ABS-45-b) contain nearly monodisperse rubber particles, while two others (ABS-45-c and ABS-50) contain a broad range of rubber particle sizes. The graft ratio, which is related to the ratio of grafted to ungrafted SAN, ranges from 0.40 to 0.72, and was determined by the supplier using procedures described elsewhere [38]. An SAN with an AN content typical of commercial ABS products (25 wt.%) was used to dilute the ABS-50 material such that the mixture contained 45 wt.% butadiene rubber. All the ABS-50 blends used for mechanical testing were diluted in this manner, such that all 4 ABS materials could be compared at a constant rubber content. The nylon 6 used is a commercially available material with \( M_n = 22\,000 \). The imidized acrylic polymer, synthesized by reactive extrusion of PMMA with methyl amine, contained 1.08 wt.% glutaric anhydride and has been described in more detail elsewhere [39]. Previous studies from this laboratory has investigated polyamide/ABS blends containing 5 wt.% of this imidized acrylic polymer [31]. The current study primarily uses the same compatibilizer content to facilitate comparison; however, limited studies presented later indicate that using lower amounts of compatibilizer may be more desirable for commercial use.

Blends in this study were prepared by simultaneous extrusion of all components in a Killion single screw extruder (L/D = 30, 2.54 cm diameter) at 240°C using a screw speed of 40 rpm. The extruded pellets were injection molded into standard 0.318 cm thick Izod (ASTM D256) and tensile bars (ASTM D638 type I) using an Arburg Allrounder injection molding machine. Prior to each processing step, all polyamide-containing materials were dried in a vacuum oven for at least 16 h at 80°C. Notched Izod impact measurements were made using a TMI pendulum-type impact tester equipped with an insulated chamber for heating and cooling the specimens. Tensile testing of dry as-molded specimens was done using an Instron at a crosshead speed of 0.508 cm/min for modulus and yield strength measurements and 5.08 cm/min for elongation at break measurements. For rheological characterization, the various polymers were tested in a Brabender torque rheometer outfitted with a 50 cm\(^3\) mixing head and standard rotors, operated at 240°C and 60 rpm.

Blend morphologies were determined using a JEOL 200CX transmission electron microscope operating at an accelerating voltage of 120 kV. Ultrathin sections (15–20 nm) were cut from Izod bars perpendicular to the flow direction under cryogenic conditions (−45°C) using a Reichert–Jung Ultracut E microtome. Various staining techniques were employed to induce phase contrast in the TEM. To stain the polyamide phase, the microtomed sections were exposed to a 2% aqueous solution of phosphotungstic acid (PTA). In other cases, osmium tetroxide (OsO\(_4\)) was used to stain the unsaturated rubber phase in ABS. This involved exposing the sections to a 2% aqueous solution of OsO\(_4\) for at least 12 h.
3. Mechanical properties, morphology, and rheology

The mechanical properties, morphology, and rheology of uncompatibilized and compatibilized nylon 6/ABS blends formed from various ABS materials are described here.

3.1. Binary and compatibilized blends containing equal parts of nylon 6 and ABS

The structure and properties of uncompatibilized (50/50 nylon 6/ABS) and compatibilized blends (47.5/47.5/5 nylon/ABS/IA) are described here. Fig. 1 shows the Izod impact strength versus temperature for 50/50 blends of nylon 6 with each of the four ABS materials. As mentioned previously, the material designated as ABS-50 was diluted with SAN such that all the blends have the same rubber content; the diluted ABS material is designated as ABS-50/SAN. Each of the binary blends is brittle at room temperature but becomes tough as the temperature is raised toward the \( T_g \) of nylon 6, about 50°C. Table 2 shows the mechanical properties of these binary blends, as well as compatibilized blends that will be discussed later. The blend based on ABS-50, which has the highest ductile–brittle transition temperature, also has the lowest elongation at break. Interestingly, the other three nylon 6/ABS blends are quite ductile during tensile testing at room temperature (as indicated by a high elongation at break) but are quite brittle as judged by notched Izod impact testing. This is no doubt a consequence of the notch sensitivity of these materials.

The TEM photomicrographs in Figs. 2 and 3 show the morphology of each blend; the polyamide phase is stained by phosphotungstic acid and the unsaturated rubber phase in ABS is stained by osmium tetroxide. Despite the differences in structural characteristics among these ABS materials, the morphological nature of their blends with nylon 6 is quite similar. The micrographs in Fig. 2 indicate that each blend contains rather large, elongated ABS domains, which have some degree of co-continuity. This leads to a poor distribution of the butadiene rubber particles throughout the sample, as demonstrated in Fig. 3. It is well documented that the rubber particles must be well-dispersed throughout the mixture to achieve effective toughening [42,43].

Previous study from this laboratory has shown that the imidized acrylic (IA) polymer referred to earlier can improve the impact properties of blends of nylon 6 with the material designated as ABS-50 [31]; similar blends based on the other ABS materials in Table 1 are explored here. Fig. 4(a) shows the Izod impact strength versus temperature for each compatibilized nylon 6/ABS blend; all blends contain a 1:1 ratio of nylon 6 to ABS by weight and 5 wt.% IA. It is evident that incorporation of the compatibilizer leads to dramatic improvements in impact strength for all of the blends relative to the uncompatibilized blends. The impact properties observed here for blends containing ABS-50 are consistent with those reported by Majumdar et al [31]. Each of the blends are quite tough at room temperature; however, there are distinct differences in their low temperature toughness that will be discussed in more detail later.

Fig. 4(b) shows the Izod impact strength versus temperature for the blends represented in Fig. 4(a), which are shown as dashes, relative to those of a commercial nylon/ABS blend sold by Bayer [26] under the trade name Triax™ 1120. The Izod impact values for the commercial material are taken from data published by Majumdar et al. [30] Each of the blends investigated here has a higher room temperature impact strength than Triax™ 1120, and all have superior low temperature toughness except the blend based on ABS-50. It is important to note that there are several fundamental differences between the commercial material and the blends investigated here. The exact composition of Triax™ 1120 has not been published, which makes it difficult to make a direct and fully meaningful comparison, i.e.
Table 2  
Mechanical properties of nylon 6/ABS/IA blends

<table>
<thead>
<tr>
<th>Nylon 6/ABS/IA blend wt/wt/wt.</th>
<th>ABS type</th>
<th>wt.% rubber in blend</th>
<th>Izod impact strength (J/m)</th>
<th>Ductile-Brittle transition temp. (°C)</th>
<th>Modulus (GPa)</th>
<th>Yield strength (MPa)</th>
<th>Elongation at break (%)</th>
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</thead>
<tbody>
<tr>
<td>50/50/0</td>
<td>ABS-50°</td>
<td>22.5</td>
<td>127</td>
<td>50</td>
<td>1.69</td>
<td>42</td>
<td>51</td>
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<tr>
<td></td>
<td>ABS-45-a</td>
<td>22.5</td>
<td>141</td>
<td>35</td>
<td>1.51</td>
<td>38</td>
<td>82</td>
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<tr>
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<td>ABS-45-b</td>
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<td>106</td>
<td>45</td>
<td>1.47</td>
<td>38</td>
<td>103</td>
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<tr>
<td></td>
<td>ABS-45-c</td>
<td>22.5</td>
<td>134</td>
<td>35</td>
<td>1.56</td>
<td>40</td>
<td>212</td>
</tr>
<tr>
<td>47.5/47.5/5</td>
<td>ABS-50°</td>
<td>21.4</td>
<td>880</td>
<td>−7.5</td>
<td>1.73</td>
<td>43</td>
<td>65</td>
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<td>ABS-45-a</td>
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<td>974</td>
<td>−40</td>
<td>1.58</td>
<td>41</td>
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<tr>
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<td>861</td>
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<td>43</td>
<td>182</td>
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<td>55/40/5</td>
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<td>1.84</td>
<td>46</td>
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<td>15</td>
<td>1.85</td>
<td>52</td>
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<tr>
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<td>1.90</td>
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<td>81</td>
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<td>646</td>
<td>15</td>
<td>1.92</td>
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<td>188</td>
<td>30</td>
<td>2.28</td>
<td>56</td>
<td>130</td>
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</table>

*a Diluted to 45 wt% rubber with SAN.
fixed rubber and compatibilizer contents, etc. of this commercial material to the current blends. No attempt is made here to quantify how each of these variables mentioned earlier affects the impact properties of Trianex™ 1120 relative to the current blends; however, it is evident that many of the blends generated here have superior low temperature ductility compared to this commercially available material.

TEM photomicrographs of the compatibilized blends represented in Fig. 4(a) are shown in Fig. 5 (PTA stained) and Fig. 6 (OsO4 stained). A comparison of Figs. 2 and 5 clearly demonstrates that incorporation of the imidized acrylic leads to a significant reduction in the size of the ABS domains. As a result, the rubber particle dispersion throughout these materials is consequently improved dramatically, as seen in Fig. 6. The modulus and yield strength of the compatibilized blends are also higher than those of their uncompatibilized counterparts (see Table 2). This could be because of improved interfacial adhesion between the nylon 6 and ABS phases and the fact that the imidized acrylic is a very rigid material.

As mentioned earlier, the low temperature ductility of these blends is clearly a function of the particular ABS material used. Fig. 4(a) demonstrates that the blend based on ABS-45-a has the lowest ductile–brittle transition temperature, followed by ABS-45-b and ABS-45-c, while the blend containing ABS-50 has the highest ductile–brittle transition. Within this set of materials, the ductile–brittle transition temperature of these blends increases as the viscosity of the pure ABS phase increases (see Table 1). A high viscosity of the ABS phase makes it more difficult to disperse and could, therefore, reduce the capacity for low temperature toughness in these blends. In addition, increasing the viscosity of the ABS phase would tend to make it the less continuous phase [44]. The TEM photomicrographs in Fig. 5 suggest that the degree of phase co-continuity is the least for the blend based on ABS-50, i.e. the ABS with the highest viscosity. The rubber particles in the blend containing ABS-50 also appear to be less interconnected in nature than in each of the other blends, as shown in the micrographs in Fig. 6. The rubber particles in ABS-50 are generally larger than those in the other ABS materials, so the average interparticle distance for a fixed rubber volume fraction is, therefore, expected to be greater [45]. However,
what appears to be connectivity of the rubber particles could partly be an artifact of the thickness of the microtomed sections relative to the rubber particle size.

The distribution of rubber particle sizes in the ABS material may affect the low temperature toughness of its blends. Considerable attention has been paid to the effects of the size of simple rubber particles (when no encapsulating hard phase is present) on toughening polyamides; it has been demonstrated that there are upper and lower critical sizes beyond which the rubber particles are no longer effective for toughening [9,46]. It has been proposed that particles below a certain size limit are unable to cavitate, and thus do not participate in the toughening mechanism [47,48]. The ABS materials used here that have a fairly monodisperse population of particles (ABS-45-a and ABS-45-b) lead to blends with superior room temperature toughness and lower ductile–brittle transitions compared to blends containing ABS-45-c and ABS-50, both of which have a bimodal particle size distribution and contain a population of very small particles (0.12 and 0.05 μm, respectively). Previous study from this laboratory using compatibilized blends containing ABS-50 suggests that the smaller rubber particles in this material do not cavitate prior to fracture in low speed bending although the testing conditions employed in the former study differ from the ones used here [41]. If these smaller particles in ABS-45-c and ABS-50 are unable to cavitate, then these blends contain a lower effective rubber content for toughening.

Brabender torque rheometry was used to assess the rheological behavior of the compatibilized blends described earlier: the torque response of compatibilized blends is shown in Fig. 7. Each of the compatibilized blends has a much higher viscosity than predicted by mixture additivity (compare to torque values in Table 1), which indicates that a significant degree of grafting occurs during melt processing. Each of the ABS-45 type materials shows a continuous increase in torque with time, suggesting continued reaction of the imidized acrylic compatibilizer with nylon 6 over this time scale, as discussed in the accompanying article [49]. Other experiments will show that increasing the processing time (through the use of multiple extrusions) can induce changes in the morphology and mechanical properties of these blends. It is unusual that the torque for the blend containing the highest melt viscosity ABS material
(ABS-50) does not increase with mixing time, and thus has the lowest melt viscosity among all the blends after significant time in the Brabender. Recent evidence has shown that trace levels of impurities used in the manufacture of the ABS-50 material (acidic or ionic species, etc.) may cause degradation of polycarbonate [50] and possibly poly(butylene terephthalate) [38], thereby reducing the viscosity of their blends. One could postulate that the flat torque response of the current nylon 6/ABS-50 blend is due to simultaneous rheological effects of degradation of nylon 6 and its grafting reactions with the imidized acrylic polymer cancelling each other out. This issue was examined for the current nylon 6/ABS-50 blend; however, no evidence was found to support the claim that ABS-50 degrades nylon 6 [51].

### 3.2. Nylon 6/ABS ratio

This subsection explores the effect of the ratio of nylon 6 to ABS on the mechanical properties of compatibilized blends. Fig. 8 shows the room temperature Izod impact strength versus rubber content, achieved by varying the ratio of nylon 6 to ABS, for blends of nylon 6 with each of the ABS materials with the IA content fixed at 5 wt.%. All the blends are super tough when the rubber content is about 22%, this corresponds to the room temperature data shown in Fig. 4(a). Below 22 wt.% rubber, the blends based on ABS-50 are no longer super tough; the Izod impact strength is reduced by a factor of two when the rubber content is decreased to 16 wt.%. Each of the other blends retains their toughness at much lower rubber contents; blends based on ABS-45-a retain high levels of toughness down to 9 wt.% rubber.

Fig. 9 shows the ductile–brittle transition temperature versus rubber content for the blends represented in Fig. 8. It is clear that the low temperature toughness of these blends is strongly dependent on the rubber content over the entire...
range studied. The ductile–brittle transition temperature increases steadily with decreasing rubber content for all the blends. Despite the differences in the molecular and structural characteristics of the ABS materials used here, each blend series shows a nearly linear relationship between ductile–brittle transition temperature and rubber content with nearly the same slope. When the ratio of nylon 6 to ABS is increased at a fixed IA content, the modulus and yield stress of each of the blends increases (see Table 2). Among the blends containing ABS-45 type materials, it is evident that the stiffness can be improved without compromising room temperature toughness by increasing the nylon 6/ABS ratio over a certain range. However, this benefit can no longer be realized once the ductile–brittle transition temperature of the blend is near room temperature. In general, ABS materials that lead to better low temperature ductility for blends containing 47.5 wt.% ABS, shown in Fig. 4(a), provide better low temperature toughness over the entire composition range when compared at a fixed ABS content.

3.3. Compatibilizer content

All the compatibilized blends discussed earlier contained 5 wt.% of the imidized acrylic polymer. Here, the effects of compatibilizer content on the mechanical properties and rheology of blends containing equal parts of nylon 6 and ABS–45-a are explored. This particular ABS material was chosen as it provided the best low temperature toughness among the compatibilized blends discussed previously.

Fig. 10(a) shows the room temperature Izod impact strength versus compatibilizer content for nylon 6/ABS-45-a/IA blends. A very small amount (0.5 wt.%) of IA is sufficient to generate blends that are super tough at room temperature. Further increases in compatibilizer content have no significant effect on the room temperature impact strength; however, the ductile–brittle transition temperature is quite sensitive to the compatibilizer content, as seen in Fig. 10(b). The ductile–brittle transition temperature drops below room temperature at less than 0.5 wt.% and decreases significantly until the compatibilizer content reaches about 2 wt.%, beyond which there is only a
Fig. 6. TEM photomicrographs of 47.5/47.5/5 nylon 6/ABS/IA blends containing the following ABS materials: (a) ABS-50/SAN; (b) ABS-45-a; (c) ABS-45-b; (d) ABS-45-c. The rubber phase of ABS is stained dark by OsO₄.

Fig. 7. Brabender torque versus time for 47.5/47.5/5 nylon 6/ABS/IA blends based on four ABS materials. Torque readings were taken at 240°C and 60 rev min⁻¹.
slight reduction out to about 10 wt.% IA where it seems to increase again.

Fig. 10(c) shows the Brabender torque of these blends as a function of compatibilizer content. The torque consistently increases with increasing compatibilizer content, which is no doubt due to the increased degree of grafting that occurs during melt blending. It is important to note that blends containing the lowest compatibilizer contents (0.5 and 1 wt.%) have Brabender torques that are only slightly higher than the uncompatibilized blend, whereas excellent room temperature impact strength is achieved (compare Figs. 10(a) and (c)). Such combinations of processability and impact strength are quite desirable for injection molded materials. When processing issues are also considered, there is no justification for using more than about 2 wt.% of this compatibilizer.

4. Conclusions

Blends of nylon 6 and ABS compatibilized with an imidized acrylic (IA) polymer were explored using a variety of ABS materials. Incorporation of the imidized acrylic polymer leads to considerably improved impact properties and a more efficient dispersion of the ABS domains and, thus, rubber particles, regardless of the ABS material used. Many of these blends possess much superior low temperature toughness than polyamide/ABS blends investigated previously. It appears that ABS materials with low melt viscosity and a monodisperse population of rubber particles lead to superior low temperature toughness. For some of these blends, increasing the nylon 6/ABS ratio over a certain range allowed for modest improvements in modulus and yield strength without compromising the room temperature impact strength.

The influence of compatibilizer content on blend properties was also investigated at a fixed ratio of nylon 6 to ABS. The room temperature impact strength of these blends reaches a maximum value at about 0.5 wt.% of compatibilizer; higher contents of compatibilizer significantly improve low temperature toughness up to about 2 wt.%. The melt viscosity (as judged by Brabender torque rheometry) of these blends continuously increases with the
addition of compatibilizer. When ease of processing by techniques like injection molding is considered, there is no justification for use of more than about 2 wt.% of the imidized acrylic polymer as a compatibilizer.

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