A NEW IMPACT MODIFIER FOR TOUGHENING CLEAR APET

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Abstract

Polymers, as well as glasses, exhibit physical ‘aging’ which leads to embrittlement. Impact modifier additives counteract this embrittlement. In clear polymer systems, such as amorphous polyethylene terephthalate (APET), blends with typical commercial impact modifiers show a significant loss of optical clarity. The work presented here, based on a new impact modifier additive, shows that embrittlement of APET can be counteracted without significant loss of optical clarity.

Introduction

Physical ‘aging’ in polymers such as APET has been studied extensively (1-4). Brady (5) showed that impact additives can counteract the embrittlement in polymer systems caused by physical ‘aging’.

Typical in impact modifier additives such as MBS (methacrylate-butadiene-styrene), ABS (acrylonitrile-butadiene-styrene) and acrylic (butyl acrylate based) have refractive indices in the range of 1.52 to 1.54. When they are added to the higher refractive index APET resin (~1.575), they drastically reduce transparency and increase the % haze of the blend.

The present work demonstrates that a new, patented (6), core-shell impact additive, Paraloid® EXL-8619, can overcome the embrittlement effect of aging’ of APET articles and maintain the desirable optical properties of the articles.

Experimental

‘Impact additive’

A novel ‘impact additive’ based on core-shell chemistry was developed to eliminate the embrittlement effect of APET and maintain desirable optical properties. The ‘impact additive’ is a 100 micron powder with refractive index matching that of APET. It can be handled like other typical in impact modifiers, e.g., MBS, ABS. It is stable at APET processing conditions. It can be blended into APET without drying.

Blend of APET with ‘Impact additive’

The APET resin used for this work was a sheet-grade APET with 0.83 dl/gm intrinsic viscosity (IV). The APET pellets were dried under vacuum at 120°C for 16 hours prior to use. The ‘impact additive’ was not dried. The powder and pellets were blended in the correct ratio and then fed to a twin screw extruder. A 34 mm Leistritz co-rotating extruder with two counter-rotating, intermeshing screws was used. Barrel temperatures were 255-280-280-280°C and melt temperatures of 275-277°C were observed. Screw rpm was set at 168.

An additional set of pellets were prepared by passing them through the Leistritz extruder five (5) times to simulate recycling.

All pellets obtained from the APET / ‘impact additive’ blends were re-crystallized at 120°C for 3-4 hrs in a tray.

Sheet extrusion

The re-crystallized pellets from the blend were dried at 120°C just prior to sheet extrusion. Clear sheet was extruded from a 38 mm Killion single screw machine with adjustable slit die and then cooled on a roll stack. Barrel temperatures were set at 238-243-243-246°C and the collar and die zones at 246°C. Rpm of the screw was 105. Melt temperatures of 266-267°C were observed. The roll stack was set at 16°C. These conditions gave sheets of 0.8 mm [0.035”] thickness.

Additional sheet samples were prepared from pellets with ‘five passes’ through the extruder.

Testing

‘Impact additive’

Falling weight impact tests on the sheet materials were run using a DYNATUP instrument. Five tests per sample were measured. Average values for total energy (Joules) and % ductility were reported. A 25.4 mm (1”) diameter tup was used as the striking head.

‘Aging’ of the sheet samples was done in a 60°C air oven for up to 10 days. Samples were allowed to cool to room temperature before testing.

Optical properties of the sheet were measured on a Hunterlab Colorimeter - Illuminant
Results and Discussion

Impact after 'aging'

Sheet samples of neat APET resin and APET with 5% and 10% 'impact additive' were tested for heat aging resistance. The sheet was 'aged' in a 60°C oven for 1 to 10 days, re-ovend and cooled to room temperature. Table 1 shows the total energy and % ductility measured for APET sheets with and without the new impact additive. A little as 5% of the new impact additive maintains the impact even after 10 days heat 'aging'.

Recycling is a fact of life for plastics packaging. Them ofum ess recycle their trim ed sheet and consumers recycle their food packaging. In order to assess how the m odified APET would handle several recycle passes, a 'worst case' expen ment was done. 100% regrind. A additional sheet samples were made with 0, 5 and 10% of the new impact additive, but the pellet compound was re-extroded 5 times in the compounding extruder before m ake sheet in the Killion extruder. The sheet was 'aged' in a 60°C oven for 1 to 10 days, re-ovend and cooled to room temperature.

Table 2 shows the total energy and % ductility measured for APET sheets with and without the new impact additive. As expected, all these sheet samples show lower total energy and % ductility for '5 pass' recycled sheet compared to sheet made in one pass through the extruder. A PET chain scission is the likely cause of the loss of impact.

The neat APET sheet with '5 passes' through the extruder dropped to 0% ductility at 3 days of heat 'aging'. A PET sheet made with 5% and 10% of the new impact additive survived 10 days heat 'aging' with impact and ductility.

Optical properties

The new, patented, impact additive was designed to maintain the optical properties of APET while providing impact resistance. Sheets prepared for impact testing were used to measure optical properties such as % luminous transmission and % haze. Table 3 lists these data for APET sheets made from 'one pass' and 'five pass' (recycle study) pellets. A sheet made from a typical in impact modifier at 6% loading in the same APET is also shown for comparison.

The typical in impact modifier gives no luminous transmission and 100% haze even at the low addition level (6%). The new impact additive lowers the light transmission slightly and increases the haze slightly in comparison to this typical in impact additive.

Optical properties for sheets made after '5 passes' through the extruder show less haze than sheets with 'one pass' through the extruder.Dispersion of the impact additive in prouces with multiple passes through the extruder.

Effect of Moisture from the Impact Additive

It is widely known that APET and other polyester resins are sensitive to moisture during processing. Drying the APET to very low H2O levels (<50 ppm) is necessary to avoid property losses from chain scission. A separate study was run to evaluate the effect of moisture present in the new impact additive on the properties in APET. Table 4 shows that impact and optical properties of the APET/impact additive sheets are unaffected by the moisture in the new powder in impact additive.

Effect on other properties

This new impact additive has some additional benefits for cutting and de-nesting of modified APET sheets. These properties will be the subject of future papers.

Conclusions

From data in Tables 1 to 3, it is clear that a new, patented, impact additive allows APET sheet to retain its impact 'aging' while maintaining its desirable optical properties.

The residual moisture in the new impact additive does not affect the impact retention on aging or the optical properties in blends made from APET and the new impact additive.

Acknowledgements

This work was completed by several groups at our Bristol Research Laboratories. Much of it can be attributed to Evan Crook, recently retired, and his group. Thanks to them and to Rohm and Haas Co. for giving permission to publish this work.
References
5. J.M. Brady, POLYM ER, 33, 2981 (1992)
6. US patents # 5321056, 5409967

Table 1 Impact Properties of Sheet after Therm al Aging at 60°C

<table>
<thead>
<tr>
<th>Sample</th>
<th>0 days</th>
<th>1 day</th>
<th>10 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>APET</td>
<td>J (% D)</td>
<td>J (% D)</td>
<td>J (% D)</td>
</tr>
<tr>
<td></td>
<td>36 (100)</td>
<td>31 (80)</td>
<td>12 (60)</td>
</tr>
<tr>
<td></td>
<td>38 (100)</td>
<td>33 (100)</td>
<td>33 (100)</td>
</tr>
<tr>
<td></td>
<td>39 (100)</td>
<td>33 (100)</td>
<td>33 (100)</td>
</tr>
</tbody>
</table>

Conditions: 25.4 mm (1") tup used in DYNATUP Instrumented Falling Dart; sheet tested at room temperature; sheet thickness ~ 0.8 mm (0.032"), IA = new 'impact additive'

Table 2 Impact Properties of Sheet from 5 recycle passes before sheet extrusion Thermal Aging done at 60°C for 1 to 10 days

<table>
<thead>
<tr>
<th>Sample</th>
<th>0 days</th>
<th>3 day</th>
<th>10 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>APET</td>
<td>J (% D)</td>
<td>J (% D)</td>
<td>J (% D)</td>
</tr>
<tr>
<td></td>
<td>31 (100)</td>
<td>6 (0)</td>
<td>4 (0)</td>
</tr>
<tr>
<td></td>
<td>24 (80)</td>
<td>15 (40)</td>
<td>16 (40)</td>
</tr>
<tr>
<td></td>
<td>28 (100)</td>
<td>25 (100)</td>
<td>22 (80)</td>
</tr>
</tbody>
</table>

Conditions: 25.4 mm (1") tup used in DYNATUP Instrumented Falling Dart; sheet tested at room temperature; sheet thickness ~ 0.8 mm (0.032"), IA = new 'impact additive'

Table 3 Optical Properties of Sheet

<table>
<thead>
<tr>
<th>Sample</th>
<th>% LT</th>
<th>% haze</th>
</tr>
</thead>
<tbody>
<tr>
<td>APET</td>
<td>89.4</td>
<td>1.1</td>
</tr>
<tr>
<td></td>
<td>88.6</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>87.1</td>
<td>1.7</td>
</tr>
<tr>
<td></td>
<td>87.4</td>
<td>2.8</td>
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</table>

Table 4 Impact and Optical Properties of APET Sheet made from 'dry' and 'wet' Impact additive

<table>
<thead>
<tr>
<th>Sample</th>
<th>10 days</th>
<th>% LT/ % haze</th>
</tr>
</thead>
<tbody>
<tr>
<td>APET</td>
<td>10 (0)</td>
<td>88.9/0.6</td>
</tr>
<tr>
<td></td>
<td>+5% IA</td>
<td>19 (80) 88.6/3.7</td>
</tr>
<tr>
<td></td>
<td>+10% IA</td>
<td>21 (100) 87.7/6.5</td>
</tr>
</tbody>
</table>

Conditions: 25.4 mm (1") tup used in DYNATUP Instrumented Falling Dart; sheet tested at room temperature; sheet thickness ~ 0.8 mm (0.032"), IA = new 'impact additive', 'DRY' = 800 ppm H2O, 'WET' = 2500 ppm H2O in impact additive powder; IA = new 'impact additive'

KEYWORDS: APET, clarity, aging, additive