Recycling of Acrylonitrile–Butadiene–Styrene and High-Impact Polystyrene from Waste Computer Equipment

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ABSTRACT: Acrylonitrile–butadiene–styrene (ABS) and high-impact polystyrene (HIPS) are two of the plastics most frequently used as outer casings for computer equipment such as monitors, keyboards, and other similar components. We assessed the effects of the recycling and blending of ABS and HIPS on mechanical properties. We found that the effects of recycling on ABS and HIPS were similar, in that changes in glass-transition temperatures, tensile strengths, and tensile moduli were negligible, but strains to failure and impact strengths were reduced considerably. Blending pro-portions of ABS and HIPS caused no more deterioration in properties than occurred as a result of the recycling process, and the presence of small proportions of one material in the other actually restored significant amounts of ductility, as seen by increases in the strains to failure. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 86: 572–578, 2002

Key words: acrylonitrile–butadiene–styrene (ABS); high-impact polystyrene (HIPS); mechanical properties; plastics; recycling; waste computer equipment

INTRODUCTION

The rapid growth of waste from electrical and electronic equipment (WEEE) can be attributed to the speed of developments in technology, which has resulted in a reduction in product life, to less than 2 years in some cases, for both domestic and commercial products. In 1998, the amount of WEEE in Europe reached an estimated 6 million tons,[1] and it is expected to double over the next decade. This growth is a reflection of what is happening worldwide and concerns over such growth are causing many organizations to develop legislation to control such waste.

Such legislation, which is currently being developed around the world, takes into consideration many issues from the design and manufacture of electrical and electronic equipment through to the end of the lives of these products. When such legislation comes into play, it will mean that recycling rates and other waste management activities will need to be increased to meet imminent recovery and recycling targets and stringent landfill regulations regarding WEEE.

Recycling WEEE is an area that has not yet reached its full potential, and if new legislative targets are to be met, more such waste needs to be collected and recycled. Despite the economic viability of recycling the electrical components of disused computer equipment, the plastic casings are currently landfilled. If these casings were recycled as an alternative to landfilling, this would produce a significant increase in the recycling rates of the electrical and electronic sector and a decrease in the volume of plastics being landfilled. A report by Sofres Conseil and The Netherlands Organisation for Applied Scientific Research on behalf of the Association of Plastics Manufacturers in Europe[2] suggested that the potential use of plastic recyclate in the electrical and electronic sector may increase if more polystyrene or acrylonitrile–butadiene–styrene (ABS) recyclate becomes available. As these are two of the plastics mainly used as computer components, an obvious solution to increase recycling rates in this sector would be to recycle the plastic casings of computer monitors, keyboards, and other similar components. However, there are several issues and barriers concerning the disposal of WEEE that need to be addressed and overcome before recycling and other alternative waste management activities can increase in this area.

One of the main concerns about WEEE is that of the disposal of harmful substances. In the case of disused computer casings, the plastics are generally flame retarded, commonly with brominated flame retardants such as polybrominated biphenyls and polybrominated diphenylethers, which have now been specified as hazardous[3] and are banned from further use. This means that plastic casings containing such flame retardants are not suitable for recycling, although there are also concerns over other methods of disposal for this type of waste. This

concern leads to other issues, which include the difficulty of identification and separation of plastic components into single-source material waste combined with that of eliminating components that contain brominated flame retardants from the recycling stream. Previous work[4] has shown that approximately 40% of plastic casings from computer equipment are correctly and clearly marked. The rest are either unmarked or are marked with the incorrect plastic type; the presence and type of additives are rarely included in any marking scheme.

Other barriers to recycling these plastic components include difficulties with dismantling, removal of impurities such as rubber feet and labels, insufficient knowledge as to how recycling affects these materials, and the viability of mixing the various different plastics that are used to make components for computers. A further issue is the lack of efficient collection or take-back systems for such equipment. Over the past few years in the United Kingdom, the collection of disused computer equipment has begun for the purpose of reuse and recycling. However, in a survey of the United Kingdom's top 100 companies by Technical Asset Management, one of the largest computer and information technology remarketing and recycling companies in the United Kingdom, findings showed that only 5% of these companies sent their computers for recycling.[5] As better collection facilities become available and public awareness increases, it is highly likely that there will be a significant increase in the availability of waste computer equipment for recycling.

In this study, we investigated ABS and high-impact polystyrene (HIPS) in an attempt to overcome some of the barriers that currently hinder the progress of recycling activities of computer equipment. A study was carried out on a random selection of disused computer equipment to determine the proportions of different types of plastics that were used to make such products and also to identify the presence of additives, in particular brominated flame retardants. A recycling process was simulated with the aim of studying the effects of recycling on mechanical properties of pure ABS and HIPS as well as the effects of recycling on blends of ABS and HIPS. Previous work has shown that multiple recycling passes had negligible effects on the tensile properties of HIPS and ABS, although more apparent effects were observed in elongational and impact properties.[6–9] It has also been suggested that most thermoplastics may be reprocessed with small amounts of other polymers without substantial decreases in mechanical properties,[10] although the impact properties may only be improved by the addition of a suitable modifier.[7,9]

EXPERIMENTAL

Materials

The materials for this study were supplied by Precious Metals Industry Ltd. (PMI; Briton Ferry, Port Talbot, United Kingdom), one of the largest computer recyclers in Europe, which recycles or reuses almost 20,000 tons of redundant computer and telecom equipment each year. At present, the copper and precious metal content of the waste electrical equipment are recovered successfully, but plastic components are generally landfilled. The following materials were supplied for use in this study:

- Circular samples (100) approximately 60 mm in diameter, taken from the plastic casings of a range of redundant computer equipment. We used these samples to identify the types of plastics used as casings for computer equipment and the proportions of each type and also to determine the presence of flame retardants and other additives.
- Unrecycled ABS in the form of computer monitor casings, from which specimens for mechanical testing were machined from various locations.
- Unrecycled HIPS in the form of disused keyboard casings, from which specimens for mechanical testing were machined.
- Unrecycled ABS and HIPS from plastic casings of redundant computer equipment, chopped into pieces approximately 50–60 × 20–40 mm, ready for reprocessing as part of a recycling process.

Identification process

A PerkinElmer System 2000 Fourier transform infrared (FTIR) spectrometer, which used the attenuated total reflection method, was used to identify 100 samples taken from various plastic casings of waste computer equipment. Known samples of pure ABS, HIPS, polycarbonate (PC), acrylic, poly(methyl methacrylate) (PMMA), poly(vinyl chloride) (PVC), and polypropylene (PP) were also tested in the FTIR spectrometer to create reference spectra. The spectrum for poly(phenylene oxide) (PPO) was obtained from a database of spectra by RAPRA Technology.

The spectrographs produced from each test were averaged from 30 scans per sample. We identified each sample by comparing the main peaks from the spectrum of the unknown sample with those from the reference spectra. However, some unknown samples of copolymers and polymer blends exhibited peaks that were characteristic of more than one.

Elemental analysis was also carried out on each of the 100 samples with a scanning electron microscope with an Oxford Instruments energy dispersive X-ray (EDX) spectroscopy microanalyzer. We did this so that

the presence of flame retardants and other additives could be detected, particularly with the view to determine the proportion of plastic casings containing brominated flame retardants.

Samples of material were cut from each plastic disc that had previously been used for FTIR analysis and attached to aluminum stubs with a silver-based adhesive. About 10 samples were attached to each disc and subsequently coated with a layer of gold prior to elemental analysis. Samples were positioned in a Jeol JSM-35c scanning electron microscope and exposed to an electron beam accelerated to 1–50 keV. The X-ray photons emitted due to the interaction of the electron beam with the surface of each sample were detected with an energy dispersive detector, and the elemental composition was analyzed with the EDX software linked to the detector.

The resulting analyses from these techniques were compared with results from two other commercial methods used for rapid identification of polymers, which were undertaken on behalf of PMI Ltd.

Mechanical recycling process

The pieces of ABS and HIPS from previously broken down casings were granulated separately into smaller particles approximately 4 mm square. These granules were used for simulated reprocessing either as single materials or as ABS/HIPS blends. The blends were weighed according to the required proportions and hand mixed. This was followed by thermal processing in a torque rheometer. This was an intensive batch mixer, similar to a Brabender, with two counterrotating mixing blades, and it was used to mimic the typical processing that might be expected in a commercial injection-molding machine. The torque rheometer was charged with about 30 g of granulated material at a cavity temperature of 190°C. We subsequently allowed 10 min for the charge to reach the cavity temperature, before processing at 30 rpm for 5 min. This part of the process was used on pure ABS and HIPS and blends of the two materials, which enabled the thermal mixing of the ABS and HIPS blends into a homogeneous material.

Following removal from the torque rheometer cavity, the solid material was allowed to cool, regranulated, and then injection molded into mechanical test specimens with a Ray Ran injection molding press for test samples. The different blends of ABS and HIPS produced are shown in Table I.

Mechanical testing

We determined the glass-transition temperature (T_g) of each material with a Mk III rheometric dynamic mechanical thermal analyzer to observe and compare any changes due to recycling or blending and also to

TABLE I
Combinations of ABS and HIPS Blends

ABS (wt %)	HIPS (wt %)
100 (unrecycled)	0
100	0
90	10
50	50
10	90
0	100
0	100 (unrecycled)

ensure that the blends were homogeneous. Samples were taken from the narrow gauge-length section of the tensile test pieces made by injection molding (or machining for the unrecycled materials). We used a combined head to perform dual cantilever tests at a frequency of 1 Hz as the temperature was increased from 25 to 150°C.

Tensile tests were performed with dog-bone samples of 36 mm gauge length and 6 mm width that had been injection molded or machined from casings. We performed tests with a Hounsfield electromechanical tensile tester at a cross-head rate of 5 mm/min to measure the tensile strength, strain to failure, and tensile modulus. We did this to observe the effects of recycling and blending on the strength, ductility, and stiffness of the materials.

Impact properties were also measured on parallel-sided $80 \times 12 \times 3.5$ mm samples. We measured the impact energy absorbed with a Ray Ran universal pendulum impact system, with an unnotched Izod geometry. Unnotched samples were used so that the results were representative of the material as a whole and not of the small region immediately behind the notch. Prior to testing, we calibrated the impact system by conducting a test without a sample. Following calibration, the sample was loaded into the impact tester, and the pendulum hammer, weighing 0.4528 kg, was set at a velocity of 3.6 m/sec, delivering 2.93 J onto the sample on impact.

RESULTS AND DISCUSSION

Identification analysis

The proportions of plastics identified from the 100 unknown samples are shown in Table II. The weight proportion was calculated with information on the relative number of components provided by PMI. From this table, it can be seen that ABS and HIPS were the two most widely used plastics for casings of this computer equipment, comprising more than 65 wt % of the casings. Moderate proportions of PPO and blends such as ABS/PVC and PC/ABS made up more than 30 wt %, whereas only about 3 wt % of casings were made with plastics such as PVC, PC, PMMA acrylic, or PP

TABLE II
Proportions of the Range of Plastics Used as Casings
for Computer Equipment

Material	Number of samples	Weight proportion (%)
ABS	46	45
ABS/PVC	8	7
Acrylic	3	<1
HIPS	16	21
PC	1	1
PC/ABS	8	15
PMMA	2	<1
PP	1	<1
PPO	14	9
PVC	1	1

Results from the X-ray microanalysis showed that 68 of the 100 samples tested contained elements that could be identified as flame retardants, a proportion of almost 60 wt %, and approximately half of these (about 30 wt %) contained bromine (Br). Other elements found included antimony (Sb), tin (Sn), chlorine (Cl), and aluminum (Al). Combinations of chlorine, bromine, and antimony appeared to be the most common flame retardants used in most plastics including ABS, acrylic, HIPS, and PC/ABS blends, whereas the other plastics contained just small amounts of other elements such as Sn or Al. Almost 90% of samples contained titanium (Ti), which is known to be from the whitening pigment titanium dioxide (TiO_2). Only 8 samples appeared to contain no additional elements that could be classified as pigments or flame retardants; however each of these samples was either black, clear, red, or dark brown in color.

Comparisons were made between the results from FTIR and X-ray analysis and rapid plastics identification methods. For the most part, with regard to the identification of the unknown samples, the FTIR analysis and the two commercial analyses were in agreement, although there were occasional differences, and the IR spectroscopy method was found to be 78% accurate, whereas the method based on sliding-spark-discharge spectroscopy was 71% accurate. With regard to the elemental analysis, however, the elements detected by these two identification methods did not appear to be as accurate as X-ray analysis. The IR method did not detect correctly the presence of any additional elements. The sliding-spark method was capable of detecting bromine and sometimes chlorine or phosphorous in some samples and was approximately 70% accurate in this, although no other elements were detected.

Although FTIR and X-ray microanalysis are more accurate identification methods, they would not be suitable as part of a commercial recycling process due to the slow speed of identification. However, although the more rapid identification methods can identify unknown plastics at speeds sufficient for a recycling

process, their accuracy in identifying different plastic components and detecting those that contain brominated flame retardants will need to increase. This will only happen as technological improvements on such methods continues.

Mechanical testing

The thermomechanical history of a polymer may affect its T_g, and any changes to the T_g may be an indication of changes to other properties. Because recycling involves thermal reprocessing, the T_g of a polymer could decrease according to the severity of the process. Figure 1, compares T_g values of each material tested, obtained from dynamic mechanical thermal analysis (DMTA).

From these results, it appears that recycling may have had a slight effect on the T_g of ABS and HIPS. The T_g of ABS was lowered from 102 to 96°C, and the T_g of HIPS also decreased from 104 to 102°C after recycling. Such decreases of T_g could be explained by the onset of chain degradation, which commonly occurs during melt processing. However, a previous study[6] suggested that changes to the T_g of ABS over a number of recycling passes are negligible as the results showed only slight reductions of one or two degrees between each recycling pass. However, to be able to conclude the same from this study, additional recycling passes would have had to be undertaken.

Blending may also affect the T_g of a polymer in one of two ways. If a polymer blend has not been sufficiently mixed into a homogeneous material, DMTA test results will show that the material has more than one T_g corresponding with the T_g of each constituent material. A polymer blend that has been sufficiently homogenized will instead exhibit a T_g determined by the blend composition in relation to the T_g of each individual polymer. The DMTA results for each of the blends of ABS/HIPS all displayed a single T_g peak,
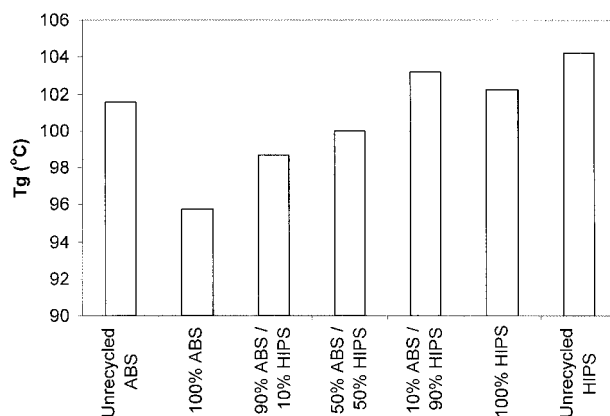
Figure 1 T_g values of unrecycled, recycled, and blends of ABS and HIPS.
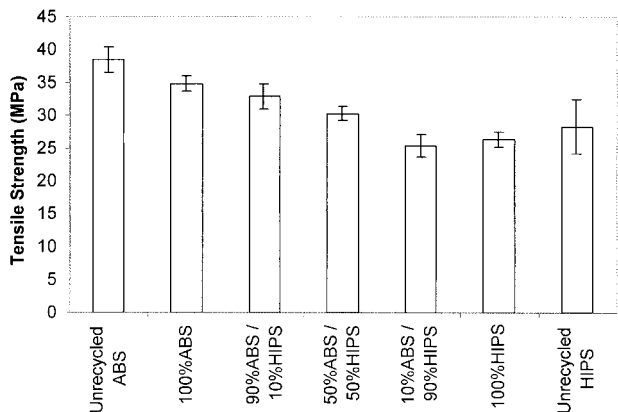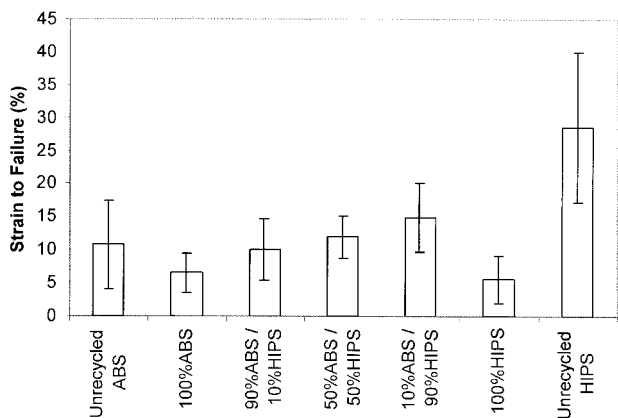
Figure 2 Tensile strengths of unrecycled, recycled, and blends of ABS and HIPS.

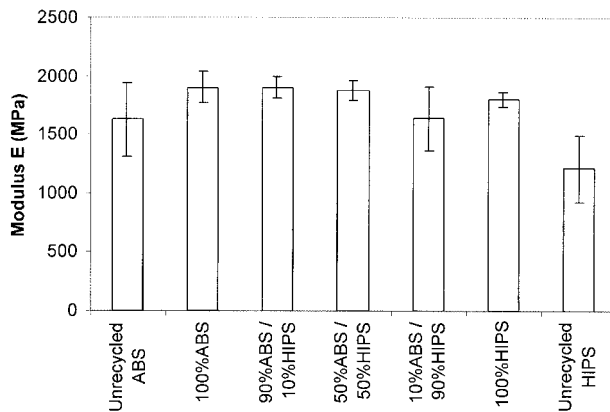**Figure 4** Tensile moduli of unrecycled, recycled, and blends of ABS and HIPS.

indicating that each blend had mixed sufficiently into a homogeneous structure.

The histogram in Figure 1 shows that the T_g of ABS increased with an increasing proportion of HIPS, as would be expected according to the law of mixtures. However, when 10% ABS was blended in HIPS, the T_g did not decrease to a temperature between that of ABS and HIPS but slightly increased to a T_g similar to that of unrecycled HIPS. The exact reasons for such small changes are unclear, but given the complex two-phase structure of both ABS and HIPS, small changes in the relative amounts of rubbery and rigid phases could be the cause. However, one important feature of these DMTA data is that the recycling and blending had no serious detrimental effects on the T_g values.

The average tensile strength, strain to failure, and elastic modulus of each material tested are shown in Figures 2, 3, and 4, respectively. Each average result was taken from approximately 10 individual tests, with the exception of unrecycled ABS and unrecycled HIPS, which were averaged from a total of 36 and 32 individual tensile tests, respectively. Standard devia-

tions were also calculated to take into account the range of scatter from these results.

The effects of recycling on the tensile strength of ABS appeared to be negligible as the tensile strength decreased only by a few megapascals (Fig. 2), which corresponds with observations by Kang and Kim,[6] who also concluded that recycling ABS had negligible effects on sample elongation and the modulus. However, as a different recycling process was used for this study, these results may differ somewhat compared with results from the previous work. Results from this study show that recycling actually decreased the average strain to failure of ABS from 11 to 6% (Fig. 3). This decrease implies that the recycling process brought about mechanical degradation of the polymer. The decrease in the elongational properties of ABS coincided with a slight increase in material stiffness, which could be seen by an increase in the tensile modulus of approximately 300 MPa (Fig. 4).

Similar conclusions could be drawn as to the effects of recycling on HIPS. The tensile strength was affected by just a small amount, which is consistent with previous studies.[7] Strain to failure of HIPS decreased dramatically from 29 to 6% after recycling (Fig. 3). The elastic modulus increased by almost 600 MPa, a greater increment than was seen with ABS (Fig. 4). These results indicate a significant reduction in the ductility of the recycled material with an increase in stiffness, which may have been caused by molecular weight changes and/or by the effects of crosslinking, due to the onset of degradation during processing. However, further investigations will need to be undertaken to verify this.

Because the T_g of a polymer blend is usually between those of the constituent polymers according to the proportion of the blend, it is possible that blending may also affect other properties in the same way. Indeed, this appeared to be the case for tensile strength (Fig. 2) because that of ABS was reduced (although only slightly) by the addition of 10% HIPS

Figure 3 Strains to failure of unrecycled, recycled, and blends of ABS and HIPS.

and decreased even further with the 50/50 ABS/HIPS blend, which gave a strength value between the two unrecycled materials. A possible exception to this trend, however, was the 10/90 ABS/HIPS blend, which actually decreased as opposed to an increase that could be expected if considering the law of mixtures approach. This could be due to problems of miscibility between ABS in HIPS, although an increase of ABS up to 50% gave a value comparable with the tensile strength of unrecycled HIPS (Fig. 2). This indicates that a large enough increase in the ABS content may restore the small amount of tensile strength lost on recycling. However, even in this worst case, it must be remembered that the tensile strength of the 10% ABS/90% HIPS blend at 25 MPa was only slightly below that of unrecycled HIPS at 28 MPa, and such changes in tensile strength that occurred due to recycling and blending were not significant.

The effects of blending on the elongational strains of recycled ABS and HIPS were favorable. The presence of 10% HIPS in ABS increased the strain to failure to approximately 10%, which was almost that of unrecycled ABS, and so the blending seemed to restore most of the ductility lost due to recycling. These results are similar to those obtained in previous investigations.[10] For a proportion of 50% HIPS in ABS, the strain to failure again increased to 12%, although for such a considerable increase in the HIPS content, this blend did not show that much more ductility. The blend of 10% ABS in HIPS increased the strain to failure more than three times that of recycled HIPS, which was a considerable improvement when one considers that the effects of recycling had so drastically reduced the strain to failure of HIPS to just a fifth of unrecycled HIPS. However, with a further increase of ABS up to 50% in HIPS, the strain to failure decreased. Therefore, it appears that the presence of a small amount of ABS in HIPS and HIPS in ABS significantly improved elongational properties that were otherwise lost due to the recycling process.

Blending ABS and HIPS did not appear to have any other significant effects on the tensile modulus other than those already apparent as an effect of recycling (Fig. 4) apart from the results for the 10% ABS/90% HIPS blend, which showed a slight decrease. When the effects of recycling and blending on the tensile modulus with effects on the strain to failure are compared, it appears that recycling dramatically reduced the strain to failure of HIPS while increasing the tensile modulus. This indicates that the material had become stiffer and more brittle. The addition of 10% ABS into the HIPS matrix significantly increased the strain to failure but did not restore full ductility, although it was sufficient to decrease the overall stiffness slightly but not enough to return to that of unrecycled HIPS.

The impact strengths of both ABS and HIPS decreased significantly after recycling as shown in Fig-
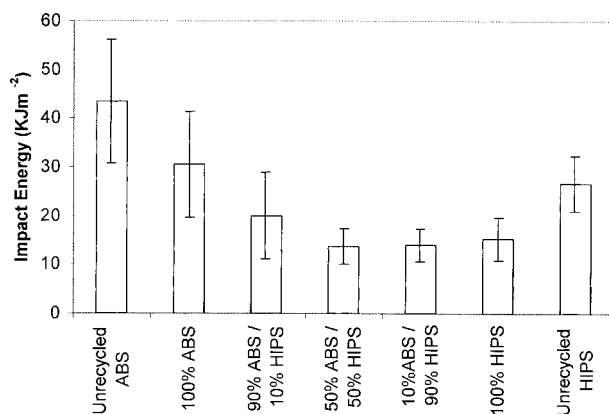
Figure 5 Izod impact strengths of unrecycled, recycled, and blends of ABS and HIPS.

ure 5. For ABS, the impact strength decreased from 44 to 31 kJm^{-2}, and for HIPS, the impact strength was reduced to 15 kJm^{-2} from an initial strength of 27 kJm^{-2}. This reduction was due to the effects of thermal mixing in the torque rheometer, which probably caused polymer chain degradation, decreasing the ability of both polymers to withstand sudden impact loads. Previous investigations[6,7,11] have also recognized the substantial deterioration in impact properties as a result of recycling. It has also been concluded that the decrease in impact properties of HIPS due to recycling may not be solely due to the effects of chain degradation but may also be brought about by the presence of the pigment TiO_2. Despite being a very fine particle, TiO_2 actually forms an inhomogeneous blend[11] within the HIPS matrix. The poor interfacial adhesion between TiO_2 and the HIPS matrix can have detrimental effects on the impact properties of HIPS. These studies have also suggested that the presence of TiO_2 may affect the tensile strength. However, the loss of such properties in recycled HIPS may be remedied through the addition of an appropriate modifier. It is also likely that the presence of TiO_2 may affect impact properties of ABS, but additional testing would have to be undertaken to verify this point.

Blending was detrimental to impact properties of ABS, although further effects on HIPS appeared insignificant. The presence of 10% HIPS in ABS decreased the impact strength to 20 kJm^{-2}, less than half that of unrecycled ABS, and an increase in the HIPS content decreased impact strength even further. The impact strength of recycled HIPS was slightly decreased by the presence of 10% ABS; however, due to the range of scatter of these results, this decrease was small enough to be considered negligible. A further increase in the ABS content up to 50% had no further significant effects on impact properties past those already brought about by recycling.

Therefore, it appears that on the whole, recycling ABS and HIPS was detrimental to impact properties,

although blending did not have any more effects than those of recycling.

CONCLUSIONS

From the samples that were taken from disused plastic casings of computer equipment that were identified by the FTIR analysis, we concluded that ABS was the most widely used plastic (45 wt %) and that HIPS was the second most common plastic (21 wt %). We also found that PPO (9 wt %), PC/ABS (15 wt %), and ABS/PVC (7 wt %) blends were used in moderate proportions, whereas only a small amount of casings were made of acrylic, PVC, PC, PP, and PMMA.

Almost 90% of these samples contained the white pigment TiO_2, and about 68% (approx. 60wt%) contained flame retardants. Of the samples that contained flame retardants, more than half contained Br and would, therefore, need alternative ways of disposal to comply with upcoming legislation, such as the WEEE directive.

From comparisons of these results with the two rapid identification methods, we found that the IR method was 78% accurate and the sliding-spark method was 71% accurate in identifying unknown plastics from waste computer casings. The sliding-spark method was able to identify the presence of Br and sometimes Cl and P and, although this method was about 70% correct, we found that some samples containing brominated flame retardants went undetected. Therefore, it appears that there is a need for technological improvement in the field of rapid plastics identification methods for the following two reasons:

1. To better identify the types of waste plastic collected, so that these different types may be separated without difficulty and recycled separately to control the composition and quality of the recycled material.
2. To better detect the presence of flame retardants, especially those that are to be banned due to legislation, so that they do not re-enter the production cycle.

Recycling had no substantial effects on T_g, tensile strengths, or tensile moduli of ABS and HIPS, although ductility was reduced significantly, particularly in HIPS. This was an effect of material degradation that occurred due to the extent of thermomechani-

cal processing. Recycling also decreased the impact strength of ABS and HIPS, which was probably due to both polymer degradation and the presence of the pigment TiO_2 in each polymer, causing some inhomogeneity.

Blending proportions of ABS and HIPS also had no significant effects on T_g, tensile strengths, and tensile moduli. The presence of 10% ABS in HIPS and vice versa had a favorable effect on the strains to failure, providing some recovery of the significant amounts of ductility that are generally lost in the individual polymers due to degradation during recycling. However, just as recycling had adverse effects on the impact properties of ABS and HIPS, blending HIPS into ABS only served to decrease the impact strength even further, whereas the presence of ABS in HIPS did not affect impact properties further than those due to recycling.

Overall, it appears that blending small proportions of ABS in HIPS and HIPS in ABS will generally improve mechanical properties that are lost due to degradation during recycling, with the exception of impact properties. However, the loss of impact strength may be improved by the addition of a suitable impact modifier.

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