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Influence of impurities on the performances of HIPS recycled from Waste Electric and Electronic Equipment (WEEE)

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A B S T R A C T

In order to produce a high quality recycled material from real deposits of electric and electronic equipment, the rate of impurities in different blended grades of reclaimed materials has to be reduced. Setting up industrial recycling procedures requires to deal with the main types of polymers presents in WEEE (Waste Electric and Electronic Equipment), particularly High Impact Polystyrene (HIPS) as well as other styrenic polymers such as Acrylonitrile-Butadiene-Styrene (ABS), Polystyrene (PS) but also polyolefin which are present into WEEE deposit as Polypropylene (PP). The production of a substantial quantity of recycled materials implies to improve and master the compatibility of different HIPS grades. The influence of polymeric impurities has to be studied since automatic sorting techniques are not able to remove completely these fractions. Investigation of the influence of minor ABS, PS and PP polymer fractions as impurities has been done on microstructure and mechanical properties of HIPS using environmental scanning electron microscopy (ESEM) in order to determine the maximum tolerated rate for each of them into HIPS after sorting and recycling operations.

Keywords:

HIPS
Recycling
Waste
Impurities
Compatibility

1. Introduction

Plastics waste represents a great source of pollution in the case of landfilling or incineration for some materials. If the expected properties of recycled plastics would be really taken into account, a tremendous wastage could be avoided by considering such materials as garbage. Although many studies on polymer recycling from DEEE have been carried out over the past few years (Mantaux and et al., 2004; Balart et al., 2004; Larsson and Bertilsson, 1995; Wäger and Hischier, 2015; Wang and Xu, 2014), only a small increase in the use of recycled polymers has been observed. One explanation for that could be related to requirements for a recycled matter, regardless of the pollutants contained within it.

Recycled plastics could meet stringent specifications (Leroy et al., 2006) if separation techniques could be improved. However, sorting, especially by Near InfraRed Spectroscopy (NIR) for different polymer grades or additivated polymers remains difficult or not yet possible. Hence, it would be easier to improve the blending process and the re-additivation of recycled polymers (Murakami, 2001; Wang et al., 2015; Pospisil et al.,

2005). As a matter of fact, there is a lack of studies about the mechanical performance of recycled polymers with minor fractions of other polymers. Mastering the influence of impurities could be of prime interest for enhancing the quality and the stability of the recycled polymer structural properties. Thus, the impact of impurities on the microstructure and on the properties of the matrix/impurity interface has to be studied. This study focused on one styrenic polymer: HIPS (High Impact polystyrene). This commodity polymer is used when both aspect and impact strength specifications are required and is largely used for electric housings and fridge fittings. Consequently, HIPS is widely present in WEEE (Waste of Electric and Electronic Equipment) and ELV (End of Life Vehicles). HIPS is a multiphase polymer composed of a rigid polystyrene matrix containing butadiene rubber nodules (from 0.5 to 10 µm in diameter) (Alfarraj and Bruce Nauman, 2004; Vilaplana et al., 2006). Many commercial grades of HIPS can be found with different viscosity and butadiene rate. It should be noted that although HIPS represents 19% of the overall plastic WEEE content, only 1% of HIPS is recycled today in Europe (Dillon and Aqua, 2000; Xu et al., 2002). The recycling rate of HIPS could increase provided that (i) the effect of impurities on this material would be completely mastered (Gent et al., 2015), (ii) no added compatibilizer could

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modify the behavior of the material and (iii) the processing parameters could remain close to virgin HIPS (Boldizar and Möller, 2003; Luzuriaga et al., 2006). Because of the difficulties in sorting HIPS fractions with different additives after grinding, recycling this polymer requires the different grades to be blended. In addition, the quality of styrenic thermoplastic resins can be affected by a small content of impurities (Tall et al., 1998) or by the extrusion process (Tang and Chaffey, 1989; Jamil and Shubber, 1988; Liu and Bertilsson, 1999). This can be explained especially by the degradation of the butadiene phase which induces an embrittlement of the polymer. Tarantili et al. (2010) showed that the degradation of butadiene phase generates a significant decrease in the *trans*-1-4 functional group and the apparition of hydroxyl and carboxyl groups. In addition, other studies have shown that impact strength and elongation at break of ABS and HIPS were affected by chain scission phenomena during recycling or ageing (Tarantili et al., 2010; Vilaplana et al., 2007, 2010; Vilaplana and Ribes-Greus, 2007; Luzuriaga et al., 2006). Although some authors (Tjong and Jiang, 1999) showed that ABS and HIPS could be miscible in specific proportions, generally a small amount of ABS lowers the impact strength of HIPS. So, in order to study the mechanical behavior of recycled HIPS regarding different sources and the influence of impurities, the first part of this work is dedicated to the study of the mixing of different grades of HIPS recycled from real deposits. In the second part, the sensitivity of HIPS to different polymer-impurities such as ABS, Polystyrene (PS) and Polypropylene (PP) is investigated through impact testing and original in-situ tensile measurements using an Environmental Scanning Electron Microscope (ESEM). Formulations of recycled materials have been simulated by incorporating only a single polymer impurity (PP, ABS or PS) in each mixture.

2. Waste materials and sorting techniques

In this study, manually sorted (from PS, HIPS and SB (styrene butadiene) marking) batches of plastics waste came from Waste of Electric and Electronic Equipment (WEEE), particularly computer housings dismantled in the Bordeaux region – France (7 batches noted “HIPSWE3EMV1” to HIPSWE3EMV3 and “HIPS-W3ETP1 to HIPS-W3ETP4”). On the average, the mean age of these products is around ten years old. The manual sorting by marking was carried out using a Phazir® NIR hand-detector test. The 7 batches represented 25 kg of matter, considered as pure recycled HIPS. Nevertheless, the HIPS may contain different rates of polybutadiene (PB).

Another batch of impure HIPS was obtained by grinding refrigerator fittings called “HIPSfridgeAC”. This batch was sorted automatically using a NIR facility (Pellenc ST®) based on IR spectra recognition and air ejection of the separated pellets regarding their IR spectrum. This batch also contained a lot of polymer impurities and foreign bodies (foam, glue, labels, and pieces of rubber) and was considered as a low quality for recycled HIPS.

After automatic sorting by IR spectroscopy, it is supposed that polymer impurities result from:

- (a) Trajectory errors during the ejection of undesirable chips; some of the most common polymer found in WEEE deposits are then likely to be found as impurities, so Polypropylene (PP) was which is used in small household equipment, was selected.
- (b) Errors due to close NIR spectra. Consequently, this led to the selection of two polymers as impurities:
 - (i) PS because it may lower the impact strength of HIPS.
 - (ii) ABS which may not be compatible with HIPS.

These impurities were manually added to pure HIPS (obtained by decontamination of previous batches) at rates corresponding to realistic sorting errors defined below.

3. Recycling and mixing process

Each batch of recycled matter was decontaminated manually (stickers, glue, metallic inserts) and was cleaned by immersion during 4 h in a mixture of water and 4 vol% of 100% biodegradable detergent. 1 kg batches of matter were then constituted after the grinding process. The flakes of matter were extruded, pure or mixed with a determined rate of impurity (PP, PS or HIPS). The impurity rate was chosen within the range of capacity of an industrial sorting facility. One run of sorting induces an impurity rate of around 5 wt% whether 2 runs of sorting allow less than 3 wt% impurity. As a consequence, impurities in the range 1–8 wt% were added. Before drying, the flakes were mixed slowly by hand with the flakes of matrix in order to lower electrostatic phenomena.

Before extrusion, the flakes were dried for 40 min: 20 min under air, then under vacuum at 85 °C. Extrusion was performed in a single-screw extruder SCAMEX VM 30/26 L/D. The screw speed was 60 rpm; the extrusion temperature in the die was 225 °C. Before injection molding, the pellets were dried (once more) for 40 min similarly. The samples for mechanical testing were injected using a DK 50/200 NGH injection molding machine at a 210 °C injection temperature and 50 °C mold temperature.

Several SEM images (Fig. 1) at a magnification of 5000× were taken on cryo-fractured cross-sections of samples, prior to the tensile tests, in order to evaluate the influence of the different impurities as function as both the nature and the rate of impurity. Impurities appear in white and the matrix in black.

The nature of the impurity strongly influences the final topographic morphology of the material. Observations highlight that the nodules of PS within the HIPS matrix present a similar morphology from 1 to 8 wt% of PS. It is also possible to consider that a few numbers of micrometric PS nodules reveal a possible partial miscibility of PS into HIPS. Moreover, no PS nodules pullout is noticeable which indicates a good interfacial adhesion with the HIPS matrix.

4. Mechanical characterization

4.1. Charpy impact test (unnotched samples)

Upon ageing and recycling of a polymer, the impact strength is one of the most damaged mechanical property (Xu et al., 2002; Gent et al., 2015). To investigate loss of HIPS impact resistance due to impurities and recycling, Charpy impact tests according to ISO 179 standard were performed on unnotched samples. Samples were temperature stabilized for 16 h at 23 °C before testing.

4.2. Uniaxial tensile tests with environmental scanning electron microscopy (ESEM)

New tailor made method based on ESEM in-situ tensile tests were conducted to view the damage sequence of the studied samples.

The nature of the impurity influences the final topographic morphology of the material significantly and thus corroborates both the values of the impact strength of unnotched impact tests as well as the analysis of the fracture aspect.

Tensile tests were carried out by ESEM Quanta FEG 200 on rectangular shaped samples (30 × 10 × 4 mm) with a 10 mm gauge length and repeated three times. Two notches (depth 0.5 mm ± 0.02 mm) were performed symmetrically on each side

Fig. 1. ESEM observations on the influence of the nature and rate of artificial impurities embedded within a recycled HiPS matrix sorted prior to in-situ tensile test (magnification: 5000 $\times$).

of samples to initiate the fracture in the observation field (Fig. 2). A unique signature was created on each sample by abrasion with SiC grinding paper P1000 in order to perform digital image correlation (DIC). The experimental conditions were focused on the analysis of topographic images (secondary electrons) using a low vacuum of 0.75 torr, an acquisition voltage of 15 kV and a 14.2 mm working distance. Each image presents a scale factor of 2.2 $\mu\text{m}/\text{pixel}$. The tensile test support is a Deben Microtest apparatus designed by Gatan (Fig. 3).

The device is equipped with a 5 kN load cell and the test speed is set at 0.2 mm/min. One image is recorded every ten seconds and force and displacement are recorded at a frequency of 2 Hz.

Fig. 2. Samples geometry (in mm) for tensile test in scanning electron microscope.

Recorded images were then used to perform DIC. Lagrangian strain (E_{xx} , E_{xy} , E_{yy}) were calculated at the tip of the notch until the crack initiation (Fig. 4). Thus, it is possible to follow the kinetics of the deformation and damage mechanisms.

Fig. 3. In situ ESEM Deben Microtest tensile test support.

Fig. 4. ESEM image of the notch before loading and after the crack initiation.

5. Results and discussion

5.1. Impact tests

The impact strength of different batches of recycled HIPS is shown in Fig. 5.

It can be noted that impact strength (unnotched Charpy) of pure recycled HIPS ranges from 40 kJ/m² to 60 kJ/m² which is close to the impact strength of virgin HIPS (67–75 kJ/m² depending on the butadiene nodule size) (Budtov et al., 1978). This first result, combined with large amount of HIPS in the deposit of WEEE, confirms that the HIPS should be recycled. The dispersion of the impact strength of the recycled HIPS can be explained by the blending of different grades, different fillers and additives present in such materials. HIPS from ground refrigerators have the lowest impact strength. This can be explained by the remaining impurities and foreign bodies glued to HIPS flakes despite the cleaning operation. This observation shows that the dismantling and sorting stages prior to recycling of plastics determines the quality of the recycled material.

The impact strengths of HIPS contaminated with 0, 1, 4, 8 and 100 wt% of virgin PS, ABS and PP at unnotched Charpy test are shown in Fig. 6.

It can be observed that, unlike what could be expected, the PS impurities do not lower the impact strength of recycled HIPS. PS, despite its low impact strength, is quite compatible with the HIPS

through the dilution of the nodules of butadiene into both HiPS and PS matrices. This has no measurable effect on impact strength up to 8 wt% rate of impurities. Furthermore, the presence of ABS in HIPS tends to reduce the impact resistance compared to virgin HIPS. Also, the standard deviation is also reduced.

Although the impact strength of the ABS (80 kJ/m²) (Budtov et al., 1978) is close to that of the HIPS and despite the chemical similarity of these two materials, ABS brings down the recycled HIPS impact strength.

Lastly, as PP has a lower impact resistance than HIPS, it can be observed that PP significantly drops the impact strength of the recycled HIPS, showing the incompatibility of PP and HIPS, even at low rates of PP.

5.2. Fracture surface observation

The SEM observation, according to the protocol previously defined in paragraph 3, of the fracture surface of HIPS +8 wt% PS provides an explanation of the low influence of PS as a contaminant (Fig. 7).

The PS impurity takes the form of very small nodules (clearer parts on ESEM image) with a diameter less than 1 µm in HIPS. The rate of these nodules can be estimated at around 5 wt%. The small size of PS nodules proves that the undissolved fraction of PS is compatible with HIPS. This means that PS is partially miscible with HIPS. In addition, the low number of cavities confirms the

Fig. 5. Impact strength of the different batches of recycled HIPS.

Fig. 6. Impact strength of PS, ABS and PP-contaminated recycled HIPS (unnotched Charpy test).

Fig. 7. Fracture surface of a specimen of recycled HIPS containing 8 wt% PS.

good adhesion between the nodules of PS and the HIPS matrix. These three points explain why the PS impurity does not alter HIPS impact strength.

SEM Observations of the fracture surface of the HIPS containing 8 wt% ABS can provide an explanation to the incompatibility between recycled HIPS and ABS (Fig. 8).

The ABS contaminating the HIPS takes the form of 1–3 μm diameter nodules. The rate of these nodules was established at 8 wt%. The ABS impurity appears entirely as nodules, which shows

a lack of miscibility between ABS and HIPS. In addition, some craters resulting from the pullout of ABS nodules are observed, which reveals a poor adhesion between ABS and HIPS. This illustrates that ABS and HIPS are not compatible and explains how even a low rate of ABS can lower the impact strength of recycled HIPS.

SEM observations of the fracture surface of the HIPS containing 8 wt% PP show the complete incompatibility between HIPS and PP (Fig. 9).

PP contaminating HIPS takes the form of large size nodules from 3 to 5 μm diameter. The rate of these nodules may be established at approximately 8 wt%. This highlights the immiscibility between PP and HIPS. In addition, it is possible to observe many craters resulting from the pullout of PP nodules, which reveals the poor interface between PP and HIPS. Hence, PP behaves as a filler without cohesion with the matrix which lowers impact strength.

5.3. ESEM in-situ tensile tests

In-situ ESEM tensile tests focus that damage phenomena behind the notch are very heterogeneous whatever the impurities. Two phases of the test can be considered, namely an initiation phase of the crack initiation followed by a propagation phase.

5.3.1. Crack initiation

Fig. 10 displays results obtained from in situ tensile tests and before the crack propagation. The evolution of load as a function

Fig. 8. Fracture surface of recycled HIPS sample containing 8 wt% ABS.

Fig. 9. Fracture surface of recycled HIPS sample containing 8 wt% PP.

Fig. 10. Load/Lagrangian strain curves for PP (a), ABS (b) and PS (c) impurities at 1 and 8 wt%.

of longitudinal (E_{yy}) and transverse (E_{xx}) strains at the tip of the notch is calculated subsequently testing, for ABS, PS and PP impurities incorporated at 1 and 8 wt%. Shear strain which was found very small, is not represented on this graph.

In all cases, the transverse strain is negligible. As expected, this result indicates that the solicitation at the tip of the notch is biaxial (also known as pure shear solicitation). All these tests report a local strain before crack initiation between 0.47 and 0.60.

The higher is the rate of impurities, the higher is the ultimate load and the lower is the ultimate strain. To a certain extent, this phenomenon depends also on the nature of impurities: The impurity rate sensitivity on the ultimate strain is negligible for ABS, significant for PS and very important for PP. The latter presents a 29% decrease when the impurity rate increases from 1 to 8 wt%. A softening behavior is observed for PS and ABS when the elastic limit is crossed, while PP presents a quasi-perfect plastic behavior.

Fig. 11 displays the evolution of load as a function of Lagrangian longitudinal strain for HIPS contaminated with 8 wt% of ABS, PS and PP respectively. The load and strain reached before crack initiation is reduced by PP impurities compared with ABS and PS, revealing a decrease of HIPS mechanical properties with additional PP impurities. HIPS contaminated with PS shows a stronger softening than the one observed for HIPS contaminated with ABS, as well as a lower strain before crack initiation.

These results of tensile tests are coherent with impact strength ones (Fig. 6) for PS and PP impurities. Moreover, independently of the nature of the impurities, the two types of mechanical testing are hardly comparable, taking into account the difference between their solicitation rates. For ESEM in-situ tensile test, the impurity rate does not seem to modify the mechanical response probably due to the plastic dissipation of HIPS matrix that negates the impurity effect, contrary to the Charpy test where the impact strength lowers quasi-linearly with the rate of impurities for ABS and PP.

5.3.2. Crack propagation

For a steady rate of 8 wt%, the ESEM images (Fig. 12) show that after fracture initiation, the crack does not grow along a direction normal to the solicitation in the HIPS systems containing PP and ABS. Damage is initiated from the coarse nodules impurities (PP and ABS) causing an interfacial de-cohesion and fracture between impurity and matrix. Yet, at 0.2 mm/min, and at this rate of impurities (8 wt%), small-sized nodules of impurities cause a more stable growing of the crack in a plane normal to the solicitation as regards PS nodules within HIPS. Thus, it could be thought that fracture could occur from growth and coalescence of the cavities which become visible within all the studied organic impurities.

The analysis of the photographs carried on in situ ESEM tensile tests reveals that the most significant cracks are observed for the PP impurities, which is wholly immiscible with the HIPS. More noticeable cracks are observed with ABS impurities than with PS ones because ABS nodules are bigger. We can likely suppose that the partial miscibility between ABS and HIPS can generate a more homogeneous shear (energy provided by extrusion) than for the PP

Fig. 11. Comparison of load/Lagrangian strain curves for each type of impurity at 8 wt%.

Fig. 12. ESEM cryofractures observations on the propagation of cracks about recycled HIPS systems depending on the nature of impurities embedded at steady rate of 8 wt% and 0.2 mm/min during an in-situ ESEM tensile test (magnification: 1000 $\times$).

impurity. PS impurities do not modify crack propagation, since PS and HIPS are nearly miscible.

The duration of testing for failure analyses are perfectly in accordance with the in-situ ESEM tensile test images, and regardless the tensile speed, PP impurities strongly contribute to the premature failure of the material. The presence of ABS, which is partially miscible with the HIPS, generates medium-sized nodules.

5.4. Discussion

The impact on the mechanical behavior of the blends based on recycled HIPS depends on the nature of the polymeric impurity.

It is obvious that only partially miscible particles, with HIPS, can contribute to a decrease in mechanical properties, even for the rates below 4 wt%. Three kinds of behaviors, depending on the miscibility between impurity and matrix, have been highlighted. At first, a small amount of PS in the recycled HIPS matrix does not deeply alter the impact strength. Micrographs have clearly shown that only few and small PS nodules appear in the HIPS continuous phase for the different loadings. The impact strength remains

steady, which shows that the miscibility between PS and HIPS is quite good. Thereby, this kind of impurity is characterized by a good interfacial cohesion for PS/HIPS.

Conversely, it has been shown that the ABS impurity was only partially miscible in HIPS, in agreement with literature (Arnold et al., 2010; Brennan et al., 2002), leading to a decrease of impact strength. ABS could have strengthened the HIPS if the miscibility of two polymers would have been complete. This proves that, despite their chemical similarity, the different butadiene phases of ABS and HIPS are not compatible. This incompatibility clearly contributes to the modification of mechanical behavior.

Lastly, the third highlighted behavior is the addition of a completely immiscible impurity (PP): the fully immiscible system HIPS/PP generates coarse PP nodules within the HIPS phase. As these nodules have no cohesion with the HIPS matrix, the latter behaves as a filler without interfacial cohesion with the matrix and strongly decreases the impact strength of the HIPS matrix.

Impact tests and tensile tests present opposite trends, showing the strong influence of loading speed on the behavior of those composite materials.

Table 1

Values of maximal impurities tolerance (ABS, PS and PP) to assure the quality of the recycled product.

<i>Values of maximal impurities tolerance within HIPS for:</i>	
ABS	<4 wt% without compatibilizing agents
PP	<1 wt%
PS	>8 wt%

It is very likely that the respective influences of miscibility and relaxation processes could be antagonistic. Indeed, the low loading speed of ESEM in situ tensile tests allow ABS impurities (partially miscible within HIPS) to have a creep relaxation response. Thus, the local relaxation phenomenon could prevail on physico-chemical interactions leading to miscibility. Nevertheless, PP impurities, showing a lack of cohesion with the matrix, compared to PS, logically lead to a decrease of the properties for both the mechanical tests.

On the contrary, we show that for high loading speed, the local relaxation does not take place at any time, only interfacial interactions predominate.

Tensile tests performed in an ESEM allow observing and studying local deformation phenomena. A drawback of this technique could be the representativeness of the observed zone in front of the whole section. Actually, this zone is 1.8 mm wide, while the cross section is 9 mm wide. Another difficulty concerns the symmetry of the test. At this magnitude, a small shift when positioning the sample can have an important impact on the result. The major problem, from the authors' opinion, is the low speed of this test which allows HIPS to soften around impurities, levelling off some other differences between materials.

The whole values of maximal impurities tolerance (ABS, PS and PP) in order to ensure the quality of the recycled product are summarized in Table 1.

By hence, the importance of an upstream quality sorting is mandatory if all these impurities are in mixture in the presence of HIPS. Indeed, a high rate of PP (totally immiscible in HIPS) or of ABS (partially miscible in HIPS), and dependent on the rate of acrylonitrile rate, could lead to premature fractures which does not lead to an optimal quality of recycled materials.

6. Conclusions

Industrial-scale recycling of post-consumer materials requires automatic sorting. These high-speed techniques entail the presence of impurities in the recycled polymer. It is therefore necessary to study their influence and to set the maximal allowed rate of each impurity. It has been shown that it was not necessary to sort the PS from HIPS with a deep accuracy since the HIPS impact strength is not decreased by the presence of PS impurity up to 8 wt%.

It was also found that ABS was only partially miscible in HIPS and that its presence as impurity could lead to a significant decrease in the impact strength of the recycled HIPS. Yet, their chemical similarity complicates the sorting of these materials at industrial throughputs. Nevertheless, the chemical similarity could be an asset for the possible compatibilization of these two polymers. In consequence, studies should move towards the research of compatibilizing agents allowing HIPS to tolerate larger amounts of ABS. Without compatibilizing agent, the maximal tolerance of ABS within HIPS can be up to 4 wt%.

It can also be noticed that PP impurity in the HIPS has to be avoided, since this kind of impurity behaves as a filler and strongly decreases on the impact strength of the matrix. However, since HIPS and PP have quite different NIR spectra and densities, a sorting better than 1 wt% of PP could be possible regarding the sorting scheme.

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