



# Process Window for Direct Recycling of Acrylonitrile-Butadiene-Styrene and High-Impact Polystyrene from Electrical and Electronic Equipment Waste



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## ABSTRACT

The aim of this paper is to assess recycling process window of ABS (Acrylonitrile-Butadiene-Styrene) and HIPS (High impact Polystyrene) from WEEE (waste from electrical and electronic equipment) through a final properties/structure screening study on their blends. Main motivation is to evaluate which amount of one plastic WEEE can be included into the other at least keeping their properties. In this sense, a wider margin of error during sorting could be admitted to obtain recycling materials with similar technological application of recycled ABS and HIPS by themselves. Results are discussed in terms of final blend structure, focusing in the interaction, within blends, of copolymers phases and fillers presents in WEEE. The comparative analysis of mechanical performance and morphology of HIPS/ABS blends indicates that the addition of 50 wt% HIPS to ABS even improves 50% the elongation at break maintaining the strength. On the opposite, HIPS maintains its properties with 20 wt% of ABS added. This study allows enlarging composition process window of recycling plastic WEEE for similar applications. This could be a sustainable way to improve benefit of e-scrap with low costs and easy processability. In consequence, social interest in the recycling of this kind of plastic scrap could be encourage from either ecological or economical points of view.

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## 1. Introduction

Exponentially increasing of technological products consumption in the last two decades generates a similar growth in waste from electrical and electronic equipment, WEEE (European Union, 2003a; Namias, 2013). Junk technology, or e-scrap, is a set of waste which includes computers, cell phones, televisions and appliances in general, considered hazardous. Cell phones are one of major contributors to WEEE because replacement time of the new generation of these devices is less than two years. It means that an equivalent amount of them will be discarded turning into e-scrap. According world statistics, by 2016 it is expected that the users of cellphones will reach 2.08 billion. From overall cells, 49% is reused as phone, 48% is discarded or is kept aside and only a 3% is recycled (Statista, 2016a, 2016b). In terms of the aforementioned reasons, it is expected that the problem continues to grow (Namias, 2013).

E-scrap contains valuable elements which justify the recycling of many of its components. Within this kind of e-scrap, plastics are neither the main residue nor the most contaminant, but they occupy lot of space because of their low density and shapes. Approximately 18 wt% of WEEE stream is constituted by plastic, usually thermoplastics which can be recycled by reprocessing (Baxter et al., 2014; Goodship and Stevels, 2012; Cui and Forssberg, 2003). Jiang et al. (2012) report that WEEE annual worldwide generation is approximately 50 million tons resulting in 10 million tons of plastic e-waste. Particularly in Argentina, 2.5 kg of WEEE per habitant are generated annually giving 0.1 million tons per year (Protomastro, 2009).

In order to reduce the amount of plastic from WEEE in landfills or in other kind of final disposal, USA authorities have adopted different regulations. Particularly, IEEE Standard force manufacturing companies of electronic and electrical housings to use at least 25% post-consumer recycled and certified material (IEEE Standard 1608 Section 4, 2006). In this sense, main manufacturers' countries of electronic devices, like China, Taiwan and Vietnam, import certified plastic WEEE from other countries (Namias, 2013).

Specifically, Argentina exportation of plastic e-scrap involves an income of approximately \$2.5 million per year (CAIP, 2010;

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Ongondo et al., 2011; Protomastro, 2009). Plastics from WEEE usually are chopped, washed and sorted by type involving relatively high costs. In this sense, actions to add value to e-waste are crucial for their business also resulting in an ecological benefit.

Plastics contained in WEEE are very difficult to separate by type using automatic methods because of their composition (Maris et al., 2015). There are some sorting methods which use spectroscopic techniques, but their precision is limited. Also, these devices are expensive and not easy to handle in small recycling industries (Beigbeder et al., 2013). Because of that, in this kind of industries usually, plastic sort by type is made manually including low precision caused by human error. Consequently labor costs increase thus, add value of recycling resins and recycler profit decrease. This procedure is also hazardous and unhealthy for workers (Ceballos et al., 2014). Then, an economical and sustainable alternative to solve this problem is to recycle resins together instead of separate them (Goodship and Stevels, 2012). However, plastics WEEE are complex systems which contain multiple copolymers resulting generally immiscible between them. They also include different fillers to achieve specific properties. Therefore, direct reprocessing of these plastics could result in blends/composites with poor final mechanical properties as resins/fillers could segregate in phases (Utracki, 1989; Datta and Lohse, 1996).

Plastic WEEE stream includes thermoplastics copolymers like ABS: Acrylonitrile-Butadiene-Styrene, HIPS: High Impact Polystyrene, PC: Polycarbonate, among others. ABS and HIPS are two of the most common plastic present in WEEE (Martinho et al., 2012; Maris et al., 2015). ABS is obtained by mixing Styrene-Acrylonitrile copolymer (SAN) with polybutadiene (PB) and HIPS also has PB in its structure (Xu et al., 2005). It is important to note that ABS and HIPS are themselves “mixtures” of several components. Their complex morphologies depend on the copolymers relative composition and phase separation that determine mechanical performance. When they are melt blended, morphology became even more complex either by phase segregation or by bubbles generation during processing (Hoyle and Karsa, 1997; Brandrup et al., 1998; Datta and Lohse, 1996). Particularly, Arnold et al. (2010), working with HIPS/ABS blends from virgin resins, postulates that phase segregation depends on the relative amount of styrene, butadiene and acrylonitrile (in the case of ABS) into each copolymer among of the relative HIPS/ABS concentration. Also mechanical properties mainly impact ones, are highly dependent on the PB phase content and dimensions (Bucknall et al., 1986). Then dispersed PB phase in both ABS and HIPS virgin resins, could act as ‘auto-compatibilizer” in their blends, enhancing phase adhesion and consequently improving mechanical performance (Tarantili et al., 2010). However, this kind of “self-compatibilization” could depend on relative HIPS/ABS proportion in the blend as well as the PB concentration in each one.

The aforementioned claim is not directly applicable to plastic from WEEE as they contain mineral fillers and additives (up to 15 wt%) to tailor desired final properties of ABS and HIPS. Then, they can be used in electrical and electronic devices housings. Typical fillers added to balance stiffness are calcium carbonate, talc and silica. Other additives are carbon black to include black color, titanium dioxide either as antioxidant or to include white color and brominated substances as flame retardants (Vazquez and Barbosa, 2016; Arnold et al., 2009; Maris et al., 2015). In this sense, blends of HIPS/ABS from WEEE could be considered a composite because of fillers/additives presence instead than a typical polymer blend. According Ben Difallah et al. (2012), the addition of 7.5 wt% of graphite produce a strong variation in mechanical behavior of pure ABS. Then, results from pure HIPS/ABS blends cannot be directly extrapolated to WEEE HIPS/ABS ones.

The aim of this paper is to determine the relative content range of HIPS/ABS blends from WEEE where self-compatibilization

occurs in order to assess their recycling process window. Commercial powder of ABS and HIPS from e-scrap were used in a screening study of three different relative blend proportions. The main motivation is to evaluate which content of one plastic WEEE can be included into the other, keeping at least its properties. In this sense, a wider margin of error during sorting could be admitted to obtain recycling materials with similar technological application of recycled ABS and/or HIPS by themselves. Consequently separation step costs could diminish. An accurate comparative analysis of mechanical performance and morphology of HIPS and ABS blends from plastic e-scrap in 80/20, 50/50 and 20/80 HIPS/ABS proportion is proposed in order to determine if self-compatibilization occurs and to assess the aforementioned ranges of relative content.

## 2. Experimental

### 2.1. Materials

Initial materials used were HIPS and ABS from WEEE. They were kindly provided, in powder form, by Ecotécnica del Pilar S.R.L. Each plastic WEEE material sample used in this work were obtained by mixing 10 powder portions of 500 g from different places of a 25 kg commercial bag in order to have a representative sample of each initial plastic e-scrap.

### 2.2. Blending

HIPS/ABS were melt blended in a batch mixer (*Brabender Plastograph W50*) at 180 °C and 30 rpm for 10 min, under nitrogen atmosphere. Initial ABS and HIPS from WEEE were also processed at same conditions in order to perform an accurate comparison with the corresponding blends. Table 1 summarizes all blends prepared including proportions and names.

#### 2.2.1. Characterization

Relative copolymer content as well as total fillers amount of ABS and HIPS from WEEE were assessed by *Thermogravimetric Analysis* using *High Resolution Modulated* mode (*HiRes MTGA*) in a *Discovery TGA* from TA Instruments. Test were performed under nitrogen atmosphere from 25 °C to 700 °C with a HiRes speed rate of 5 °C/min, HiRes sensitivity of 1.00, resolution of 6, Modulated temperature amplitude of 5 °C and period of 200 s. Five runs for each sample was tested in order to obtain a semi-quantitative composition of both plastic WEEE.

Blends morphology was analyzed from *Scanning Electron Microscopy (SEM)* images obtained in a *LEO EVO 40 XVP* electron microscope operated at 10 kV. Samples were cryo-fractured by liquid nitrogen immersion, mounted on bronze stubs and then, coated with a gold layer (~30 Å), using an argon plasma metallizer (sputter coater PELCO 91000).

#### 2.2.2. Mechanical properties

Three point flexural tests were performed at room temperature in a *Universal Testing Machine Instron 3369*. Flexural specimens were cut from plates prepared by compression molding at

**Table 1**  
Name and composition of all blends prepared.

Names	HIPS/ABS (wt%/wt%)
ABS	0/100
H20/A80	20/80
H50/A50	50/50
H80/A20	80/20
HIPS	100/0

**Table 2**

Relative mass content of volatiles, acrylonitrile, styrene, butadiene and fillers in ABS and HIPS from WEEE determined by HiRes MTGA (error  $\leq 5\%$ ).

	Volatiles (wt%)	AN (wt%)	St (wt%)	PB (wt%)	Fillers (wt%)
ABS	2.0	32.5	28.1	30.6	8.8
HIPS	2.1	6.0	60.6	28.8	4.6

180 °C. Test conditions and specimen dimensions were set according to ASTM D790-03 standard for plastic. Eight specimens for each sample were measured. Modulus, ultimate strength, toughness and elongation at break were assessed from stress–strain curves.

### 3. Results and discussion

The accurate characterization performed on initial WEEE materials evidences the presence of typical additives: calcium carbonate, silica, carbon black, talc and titanium dioxide. ABS contains 8.8 wt% of fillers while HIPS, 4.6 wt%. This data was obtained by a combination of X-ray Diffraction, X-ray Fluorescence, Infrared Spectroscopy and Thermogravimetric Analysis, as it was presented in a previous work on the same system (Vazquez and Barbosa, 2016). In the same paper, those techniques also allowed to determine that plastic WEEE used satisfy European regulations regarding bromine content (European Union, 2003b). In this sense, according to WEEE initial materials characterization, it is possible to claim that blends performance will be conditioned by plastic e-scrap fillers and their interactions with the rubbery phase (butadiene) present in both, ABS and HIPS.

In order to complete plastic WEEE characterization a semi-quantitative analysis of their composition was performed using HiRes MTGA with an error less than 5% from the average of 5 runs. Results are shown in Table 2. The first important information to extract from this table is that HIPS WEEE contain a few amount of ABS, because of the detected presence of 6 wt% of acrylonitrile (AN). This fact evidences that actual plastic WEEE is not completely sorted, as expected. The presence of HIPS into ABS is not possible to assess with this technique (and others ones) because of the similarity of their components (Cafiero et al., 2015). Regarding HIPS composition, the amount of styrene (St) duplicate the PB one. While an equal amount of AN, St and PB was found in ABS. Both samples contain around 2 wt% of volatiles and fillers content in ABS duplicate those in HIPS. It is important to note that PB amount in both plastic WEEE is similar meanwhile HIPS St content duplicate the ABS one.

Blend compatibilization effectiveness is directly related with mechanical properties which depends on phase adhesion. Young Modulus (E) is a low strain property that only depends on the internal structure of the species and relative concentration of the components in the case of blends. However, changes in high strain

**Table 3**

Flexural mechanical properties for plastic WEEE initial materials, compatibilizer copolymers and all blends prepared, according ASTM D790-03.

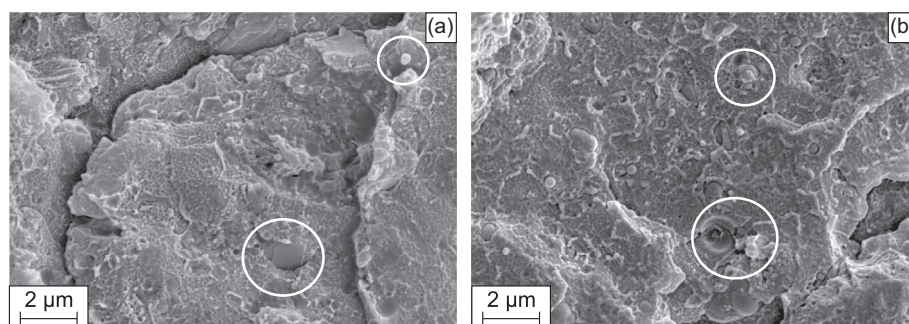
Sample	E (MPa)	$\sigma_u$ (MPa)	$\epsilon_b$ (%)	Toughness (J/m <sup>3</sup> )
HIPS (H)	2068 $\pm$ 208	35.6 $\pm$ 1.6	6.96 $\pm$ 0.26	2.25 $\pm$ 0.16
ABS (A)	2339 $\pm$ 29	43.4 $\pm$ 1.7	2.44 $\pm$ 0.16	0.63 $\pm$ 0.07
H20/A80	2385 $\pm$ 113	43.7 $\pm$ 2.7	2.69 $\pm$ 0.14	0.67 $\pm$ 0.13
H50/A50	2148 $\pm$ 56	43.6 $\pm$ 2.3	4.03 $\pm$ 0.68	1.40 $\pm$ 0.18
H80/A20	1946 $\pm$ 112	37.4 $\pm$ 2.0	5.90 $\pm$ 0.33	1.81 $\pm$ 0.15

properties like ultimate strength ( $\sigma_u$ ) and elongation at break ( $\epsilon_b$ ) are the best evidence of phase adhesion effectiveness and consequently compatibilization between phases. Then, adhesion between phases, the strongest evidence of blend compatibility, is better assessed by analyzing the variation of  $\sigma_u$  and  $\epsilon_b$  of blends respect to initial materials.

Table 3 shows flexural mechanical properties (E,  $\sigma_u$ ,  $\epsilon_b$  and Toughness) for WEEE initial materials and all blends prepared. HIPS has lower E and  $\sigma_u$ , and higher  $\epsilon_b$  and toughness than ABS. Please note that the stiffer behavior in ABS is mainly due to the higher content of filler. ABS is more rigid (E: 13% higher) and strength ( $\sigma_u$ : 22% higher) than HIPS and consequently less ductile ( $\epsilon_b$ : 25% lower). This behavior is corroborated through HIPS and ABS fractography. Cryo-fracture surface SEM micrographs of ABS and HIPS are shown in Fig. 1. Fillers (circles in Fig. 1) from different natures like calcium carbonate, silica, carbon black, talc and titanium dioxide are evident because of their typical particle shape. Also, it is possible to see that ABS (Fig. 1a) has a more brittle behavior evidenced by sharper fracture edges than HIPS (Fig. 1b) according with its low toughness value in Table 3. On the other hand, rubber phase domains are well distributed in both polymers surfaces but, domains in HIPS seems to be bigger than ABS ones. This kind of morphology agrees with major error in zero and low strain properties like E and  $\sigma_u$ , respectively. In fact, E standard deviation is one magnitude order higher in HIPS.

Before discussing HIPS/ABS blends compatibilization behavior, it is important to note that fillers present in each material, 4.6 wt% in HIPS and 8.8 wt% in ABS, make each plastic WEEE a composite with, HIPS or ABS as matrix. Then, from now on it will be named “blend” which actually is a mixture of composites.

In order to determine if self-compatibilization occurs in plastic WEEE blends under study and to assess the range of concentrations, initially mechanical behavior is analyzed as a function of blend relative concentration, and then results are confirmed by an accurate morphological study. Flexural mechanical behavior of WEEE initial materials and all blends prepared are presented Fig. 2; meanwhile mechanical properties values are listed in Table 3. Stress-strain curve for blend with 80 wt% of ABS (H20/A80) match with ABS one. E,  $\sigma_u$ ,  $\epsilon_b$  and toughness did not suffer changes, their variations fall within error values. It seems that addition of HIPS into ABS



**Fig. 1.** SEM micrographs (20,000x) of cryo-fracture surface of: (a) ABS and (b) HIPS.

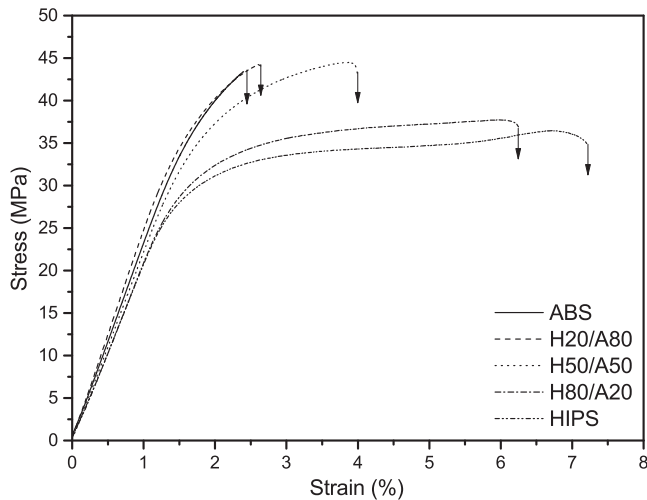


Fig. 2. Flexural stress-strain curves of ABS, H20/A80, H50/A50, H80/A20 and HIPS.

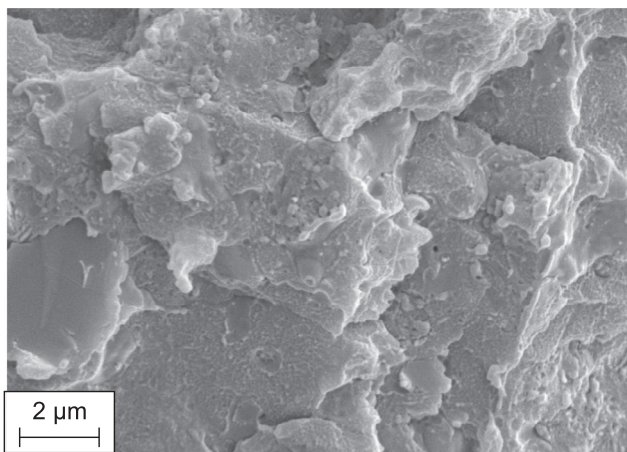


Fig. 3. SEM micrographs (20,000x) of cryo-fracture surface of H20/A80 physical blend.

allows to conserve the strength because, besides a more rubbery material was added, global mechanical behavior is still govern by fillers/matrix interaction. This is consistent with morphological aspects which can be observed from cryo-fracture SEM micrograph of H20/A80 blend in Fig. 3. The overall phases/fillers relative dispersion and distribution, as well as sharpen fracture edges showed in Fig. 3, are very similar to the ABS one (Fig. 1a). These results are evidence that it is possible to use a blend of ABS with 20 wt% of HIPS for same applications of initial ABS.

Blend with 50 wt% of HIPS mechanical behavior (Fig. 2) indicates a possible partial location of the rubbery phase into the interphase filler/matrix. Firstly, same  $\sigma_u$  as the ABS was achieved with higher elongation at break. Also, from comparative analysis of H50/A50 and ABS stress-strain curves it is clear that fillers still govern the overall behavior at low strain because  $E$  value did not change. However, at higher deformations in, the rubbery phase resist which results in lower stress value for H50/A50 blend compared to the ABS at the same strain. This claim is corroborated by the comparison between cryo-fracture SEM micrographs of H50/A50 (Fig. 4) with the ABS one (Fig. 1a). It is possible to note that H50/A50 has bigger phase domains with fillers inside them, agreeing discussion made from mechanical analysis. From the point of view of H50/A50 final application, results indicates that this material can be used as a replacement of the ABS with better ductility.

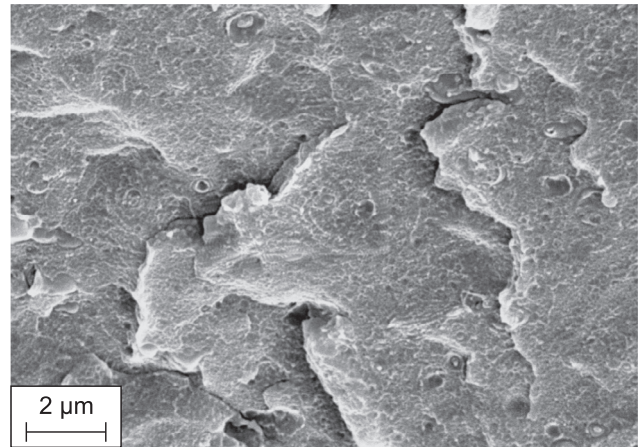


Fig. 4. SEM micrographs (20,000x) of cryo-fracture surface of H50/A50 physical blend.

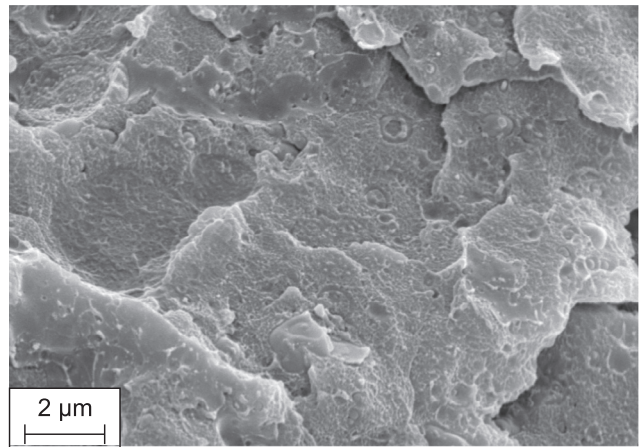
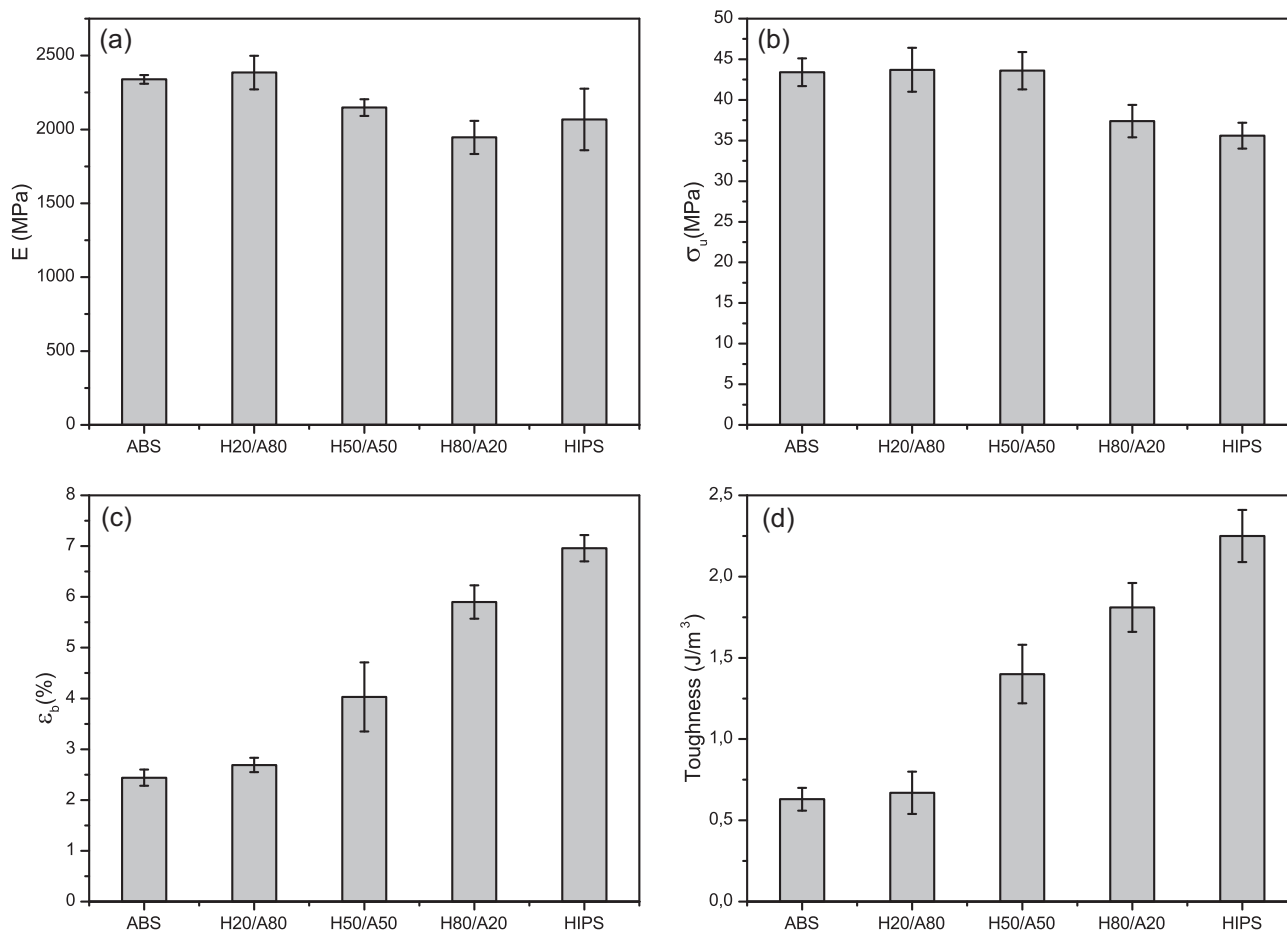


Fig. 5. SEM micrographs (20,000x) of cryo-fracture surface of H80/A20 physical blend.

Blend containing 80 wt% of HIPS (H80/A20), even remaining into the experimental error, present a small negative deviation of the rule of mixtures in  $E$  (Table 3). This slight reduction in  $E$  is not an expected result because ABS has higher filler content than HIPS. Based on changes in blend morphology respect to initial materials, it is possible that during blending fillers and rubber suffered a relative redistribution, then rubber locates around fillers. In comparison to ABS, H80/A20  $\epsilon_b$  and toughness have been increased in approximately 200% while  $E$  and  $\sigma_u$  have decreased in a 15%, this also can be attributed to filler encapsulation. This behavior can be appreciated in stress-strain curve, where the overall mechanical performance is now govern by rubber phase since for the same strain the stress for H80/A20 is much lower than ABS value and ductility has been notably increased in comparison to the ABS. This blend results in a material more similar to the initial HIPS. Regarding blend morphology, in Fig. 5 is possible to note that phase domains size of this blend are smaller than H20/A80 and H50/A50 ones, and also well dispersed and distributed. The lack of sharpen fracture edges evidence the rubbery behavior of H80/A20 blend. These morphological characteristics agreed the mechanical analysis.

In order to determine if HIPS/ABS blends from WEEE can be used in same applications of the initial ABS WEEE and HIPS WEEE, and then enlarge recycling opportunities, mechanical properties variation are compared. Fig. 6 shows  $E$ ,  $\sigma_u$ ,  $\epsilon_b$  and toughness as a function of relative blend concentration. It is possible to observe



**Fig. 6.** Summarized mechanical properties analyzed of initial WEEE materials and all blends prepared: (a) Elastic Modulus (E); (b) Ultimate Strength ( $\sigma_u$ ); (c) Elongation at break ( $\epsilon_b$ ); and (d) Toughness.

that E (Fig. 6a) and  $\sigma_u$  (Fig. 6b) of ABS did not change even with the addition of 50 wt% of HIPS as it was explained above. Also, it is important to note that  $\epsilon_b$  (Fig. 6c) and indirectly toughness (Fig. 6d) are the most sensitive properties at high strain deformations, which corroborate what was mentioned as the beginning of the discussion.

From the overall view is possible to determine that the ABS can support the addition of HIPS up to 50 wt% without suffer mechanical deterioration and also with considerable improvements in the elongation. However, the addition of 80 wt% of HIPS results in a deterioration of ABS properties. In the opposite, HIPS can tolerate 20 wt% of ABS, with a small decreasing in the elongation, higher amounts of ABS deteriorate HIPS mechanical properties notably. This claim agrees Brennan et al. (2002) results for the case of blends with major content of HIPS from e-scrap, which shows that direct blending with ABS improves mechanical performance of it. It is important to note that these results give an idea of the plastic e-waste recycling process window. In the case of ABS it has been increased, due to the improvement of the elongation with the addition of HIPS up to 50 wt% and on the other hand, HIPS process window also was improved with the addition of ABS up to 20 wt%.

#### 4. Conclusions

The system under study contains either fillers or complex copolymer morphology with rubbery domains resulting in a complex composite. Those aspects contribute oppositely to mechanical behavior of final materials allowing to determine the concentration range where blends are similar to ABS and HIPS from WEEE.

ABS can support the addition of higher amounts, up to 50 wt%, of a second material (HIPS) keeping mechanical properties and also improving them, elongation and toughness in 200% approximately. On the other hand, HIPS tolerate the addition of ABS up to 20 wt% increasing the application field of it but not as much as the ABS.

The present work allows to say that all blends present a lack of “selfcompatibilization” behavior, given a possible increase of the process window for recycling of ABS and HIPS from WEEE. Direct blending of HIPS and ABS from e-scrap in the studied proportions seems to be a good method to obtain recycled materials with similar or better mechanical properties than initial plastic e-waste. This way could be a sustainable route to improve plastic WEEE benefit with low costs and easy processability. Consequently, social recycling interest could be encouraged by both ecological and economical points of view.

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