Upgrading of recycled ABS from end of life vehicles and waste of electrical and electronic equipment

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ABSTRACT: Recycled acrylonitrile butadiene styrene (rABS) is a common material, originated from End of Life Vehicles (ELV) and Waste of Electrical and Electronic Equipment (WEEE). During the life time of the product, ABS is subjected to degradation in the form of chain scission and crosslinking with a low impact strength as a consequence. To use this stream in closed- or open-loop recycling the impact value of rABS should be recovered. This is attempted by adding a compatibilizer/impact modifier styrene-ethylene/butylene-styrene grafted with maleic anhydride (SEBS-g-MA) and/or a recycled polypropylene (rPP). At first the influence of processing on the degradation mechanisms was investigated, with as conclusion that the degradation takes place mainly during the life time of the product. Secondly the impact strength and flexural modulus were measured from the compatibilized rABS, with different amounts of compatibilizer and from a blend rABS/rPP with or without SEBS-g-MA. Test samples were injection moulded and tested. The SEBS-g-MA has a positive effect on the impact strength, but only starting from an amount of 10wt%. This effect is the same for the rABS/rPP blend. Of course the addition of an impact modifier will have a negative influence on the stiffness and the flexural modulus. This in combination with the economical aspect should be kept in mind to select this upgraded material for a new application.

1 INTRODUCTION

Design from recycling (DFR) is a new concept in the polymer industry. Instead of creating products for efficient recycling, the waste stream (post-industrial or post-consumer) will be the base for the design of a new product. (Ragaert, 2016) The material is the key of the concept. In vehicles and electronic devices a common used material is acrylonitrile butadiene styrene (ABS). After use these products are collected and the different components are separated. During the life time of the product and during processing, the ABS degrades due to chain scission (Fig.1b) and crosslinking (Fig.1a). (Arostegui et al., 2006; Peydro et al., 2013; Scaffaro et al. 2012) Due to degradation, volatile components are exposed, especially styrene gas. (Arnold et al., 2009) This can lead to hole formation. Degradation during the reprocessing depends on the number of cycles and processing temperature. Peydro et al. investigated the effect of reprocessing and concluded that degradation has the biggest effect in the first cycle and decreases in the next cycles. (Peydro et al., 2013) They also conclude that the processing temperature should be kept as low as possible.

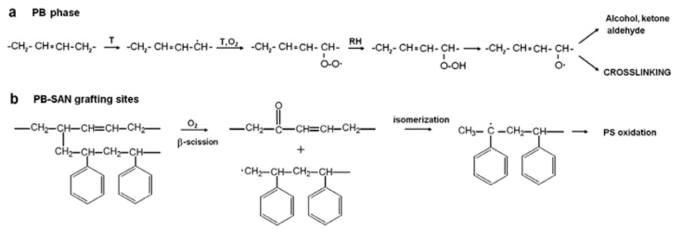


Figure1. Degradation mechanisms ABS (Scaffaro et al., 2012)

The combination of chain scission and crosslinking has a catastrophic effect on the impact strength, making it challenging to produce functional products. The use of additives and/or blending with other polymers can solve this problem. In this research the effect of SEBS-g-MA and the blend with rPP were investigated. In literature an increase of 75% in impact strength of the virgin ABS is described, when 5wt% SEBS-g-MA is added. (Peydro et al., 2013) In the first place SEBS-g-MA was added as impact modifier and secondly as compatibilizer. Due to the flexibility of the rubber particles higher loads can be applied and the material becomes tougher. (Greco, 1998) The ligament length, distance between the rubber particles, is an important factor. If more of the compatibilizer is added, the ligament length decreases. This value should be smaller than a critical length to enhance toughness. (Bacci et al., 2013) Yang investigated the properties of a compatibilized blend of virgin ABS and virgin polypropylene (PP). (Yang, 2007) The notched Izod impact value increased from 64.1 J/m to 250.9 J/m, a remarkable increase of 390%. Based on this study, the decision to blend the rABS with an in house rPP was made. The big difference in polarity between the two components makes a good interaction impossible. A compatibilizer will be needed to enhance the miscibility of the two components, and to obtain better properties. Also here SEBS-g-MA was used as impact modifier/compatibilizer. The compatibilization effect will be mainly due to dipole-dipole (with rABS) and Van der Waals (with rPP) interactions. The materials were first compounded, after which test samples were injection moulded. Flexural and impact tests were performed. Also the effect of degradation was investigated by a thermogravimetric analysis combined with gas chromatography (TGA-GC).

2 MATERIALS AND METHODS

2.1 *Materials*

The rABS was kindly donated by the company Galloo Plastics, located in Halluin (France). They are specialized in recycling of household waste, ELV and WEEE. They separate the rABS, based on differences in density. After which a melt filtration and regranulation of the material is performed. These granules can be used in new applications, but the low impact strength makes it difficult to produce high quality products. The rPP originates with the shell of suitcases, produced by Samsonite, located in Oudenaarde (Belgium). The used SEBS-g-MA is the FG1901 G from Kraton (USA).

In Table 1 the flexural modulus and the impact strength of the pure components are given. The values for the rABS and for the rPP are measured within the research group Center for Polymer & Materials Technologies (CPMT).

Table 1. Component properties

Material	Producer	Type	Impact strength	Flexural modulus
			kJ/m²	MPa
rABS	Galloo Plastics	GP-ABS-457	4,6 ± 0,3	2138 ± 6
rPP	Samsonite	rCurve	3,6 ± 0,4	1414 ± 11

2.2 *Methods*

All blends of rABS and rPP, with or without SEBS-g-MA, were compounded with the co-rotating twin-screw extruder (Coperion ZSK 18, 40 L/D), using two different feeders, a main and side feeder. Compounding was performed at a temperature of 230°C. The blends are named ‘rABS : rPP : xwt%SEBS’. Wherein x is the weight percentage of the compatibilizer. For clarity SEBS-g-MA will be shortened to SEBS. The specific compounds can be found in Table 2.

Table 2. Specific compounds

Name	rABS	rPP	SEBS
	%	%	%
rABS	100	0	0
rABS:5wt%SEBS	95	0	5
rABS:10wt%SEBS	90	0	10
rABS:20wt%SEBS	80	0	20
rABS:rPP	50	50	0
rABS:rPP:10wt%SEBS	45	45	5

SEBS was added via the side feeder for the rABS and manual mixed with the rABS in the blend with rPP. The rABS was also compounded using a vacuum (500mbar) at the end of the extruder, to remove possible degradation gasses. The compounds were dried for minimal 12 hours. For all compounds test bars were injection moulded on a BOY 22S injection machine at the same temperature of compounding and were left at room temperature for minimum two days prior to testing. These test bars include rectangular bars for flexural and impact testing.

In the initial phase of the research, density measurements were performed on the extruded rABS with and without the use of a vacuum. The density was gravimetrically determined by the immersion method according to ISO 1183. This was executed in propanol by means of a density kit and a Precisa balance. Next to the density measurement also a TGA-GC was performed to analyze degradation products. The sample was heated to 750°C with an isothermal of 20 minutes at 230°C. This measurement was done by the Insitute of Polymers (Sofia, Bulgaria). The different compounds were tested on their mechanical properties, especially impact and flexural properties. First the rABS with and without SEBS-g-MA were compared, where after the blend of rABS and rPP (with or without SEB). According to the Charpy method of ISO 179-1/1eA the rectangular test bars (100x10x4mm) were notched 2 mm and tested for impact strength. The impact bars (without notch) were also used to perform the flexural testing. The bending tests were executed according the norm ISO 178, using an Instron 4464 testing machine. The reported values for the mechanical properties are the average of at least ten measurements. All data was processed with SPSS Statistics 22 and screened for outliers. The outliers were deleted in order to work within the 95% confidence interval.

3 RESULTS AND DISCUSSION

3.1 *Degradation of ABS*

The effect of processing was assessed by a density experiment between extruded samples with and without vacuum. There was no significant difference in density between the sample without and with vacuum, 1.054±0.033 g/cm³ and 1.052±0.008 g/cm³ respectively. Also the degradation gasses were monitored in a TGA-GC experiment. A small loss of mass (0.83%) could be noticed at the temperature of 230°C, but too small to perform an analysis of the evaporating gasses. It can be concluded that the effect of degradation during processing is very small and occurs mainly during the lifetime of the product. impact strength is influenced most due to the crosslinking phenomenon. The use of a vacuum to prepare the compounds was not necessary.

3.2 *Properties of the rABS with and without SEBS*

Due to the low impact strength of the rABS, SEBS was added as impact modifier/compatibilizer. In Table 3 the impact strength and flexural modulus for the rABS with different amounts of SEBS can be found.

Table 3. Mechanical properties of rABS:SEBS compounds

Material	Impact strength	Flexural modulus
	kJ/m²	MPa
rABS	4.1 ± 0.5	2227 ± 61
rABS:5wt%SEBS	4.1 ± 0.1	1947 ± 3
rABS:10wt%SEBS	4.5 ± 0.3	1787 ± 23
rABS:20wt%SEBS	6.9 ± 0.3	1373 ± 23

The addition of SEBS will only have a noteworthy effect starting from 10wt%. The impact strength of the compound with 10 and 20wt% SEBS is significant different from the rABS and the compatibilized rABS with 5wt% SEBS. At first the introduction of 5wt% is too small to have an effect on the impact strength. When more SEBS is added the function as impact modifier will be fulfilled. More rubber particles are present and the ligament length will decrease, with a toughening effect. Starting from 20wt% a big increase can be seen. Due to the addition of the impact modifier, the material becomes more tough and will lose stiffness. The use of SEBS can be considered, depending on the application (loss in stiffness) and economical interest.

3.3 *Properties of rABS:rPP blend with and without SEBS*

The comparison was made between the unmodified and the compatibilized blend. The results can be found in Table 4.

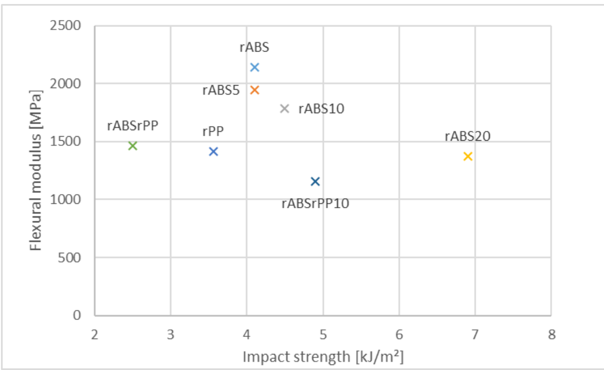
Table 4. Mechanical properties of rABS:rPP blend with and without SEBS

Material	Impact strength	Flexural modulus
	kJ/m²	MPa
rABS	4.1 ± 0.5	2227 ± 61
rPP	3.6 ± 0.4	1414 ± 117
rABS:rPP	2.5 ± 0.3	1463 ± 35
rABS:rPP:10wt%SEBS	4.9 ± 0.3	1153 ± 60

The results of the unmodified blend show a decrease in impact strength, the values are even lower than the separate components. The flexural modulus shows an increase, compared to the value of rPP. But this increase is minimal. The immiscibility of the two components is the cause for the low mechanical properties. Compatibilization is needed to restore the mechanical properties. A blend compatibilized with 10wt% SEBS was compounded. The impact strength increased significantly, compared to the unmodified blend and the separate components. The flexural modulus decreases, due to the same reasons as in the previous paragraph. The compatibilizer has a positive effect on the toughness of the blend, with an increase of almost 100%. But the effect of the SEBS as compatibilizer is less than expected. Compared to the pure components, the increase in impact strength is not that high and the loss in flexural modulus should be kept in mind, especially in comparison with rABS.

3.4 *Flexural modulus vs. impact strength*

In Graph 1 the relation between impact strength and flexural modulus for every compound can be found. The standard deviation is left out for clarity. The flexural modulus is lower for every combination in comparison with the rABS. The impact strength is improved significantly by adding 20wt% SEBS or an impact modified blend with rPP. Also the possibility to improve the toughness of the rPP by a compatibilized blend with rABS, with an acceptable decrease (18%) in flexural modulus, could be a worthy option.



Graph1. Flexural modulus vs. impact strength

4 CONCLUSION

The biggest influence on degradation is the first processing and the life time of the product. Chain scission and crosslinking can occur, resulting in low impact strength. During reprocessing a slight degradation is possible, but the effect is minimal.
It can be concluded that the impact strength of rABS can be improved by addition of impact modifiers and/or other polymers. The addition of SEBS to the rABS is only effective when at least 10wt% is added. For 20wt% the impact strength is increased with almost 70%, due to the bigger amount of rubber particles and a decrease in ligament length. The additive serves mainly as an impact modifier and less as compatibilizer. The blend with rPP shows inferior properties. With the addition of SEBS the impact strength is recovered, even with a small increase. The SEBS has a compatibilizing effect, Van der Waals and dipole-dipole interactions are possible. The counterpart is a decrease of the flexural modulus. Economic interest and application are the two key factors in considering the use of this upgraded material.

5 ACKNOWLEDGEMENTS

Thanks are extended to Galloo Plastics in Halluin (France) and Samsonite in Oudenaarde (Belgium) for the donation of the recycled polymers and to Kraton (USA) for the SEBS. Also the TGA-GC measurement in this research was conducted by the Institute of Polymers in Sofia (Bulgaria).

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