Evaluation of the quality of post-consumer recyclates obtained from distinct recycling strategies

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ABSTRACT: The focus of the presented research is on the mechanical recycling of post-consumer plastics from Waste Electrical and Electronic Equipment (WEEE) and End-of-Life Vehicles (ELVs). Currently only a limited number of these plastics are mechanically recycled on an industrial scale while the majority is thermally treated for energy recovery. Since there is a significant potential for value recovery and to lower the environmental impact of plastics, the aim of the present research is to develop new mechanical recycling processes based on the disassembly of plastic components and the identification of the plastic by spectroscopic methods. This paper will present this distinct recycling process based on a case study of the recycling of Acrylonitrile Butadiene Styrene copolymer (ABS) from Liquid Crystal Display (LCD) TVs. The quality of the achieved recyclates is evaluated by mechanical testing and results showed high mechanical properties when compared to commercially recycled ABS.

1 INTRODUCTION

1.1 *WEEE and ELV Waste*

Every year between 8 and 9 million tons of waste from End-of-Life Vehicles (ELV) and around 11,6 million tons of Waste Electrical and Electronic Equipment (WEEE) are generated in the European Union (Baldé et al., 2015a; Directive 2000/53/EC, 2000). Due to a growing number of sold cars and electronics and decreasing life spans of electronic products these waste streams are expected to significantly increase in the following years (Baldé et al., 2015b; Kanari et al., 2003). In ELVs around 9 wt% plastic components can be found and the amount of plastics in automotive is increasing due to the low specific weight, the low cost and the durability of plastics (Duval and MacLean, 2007; Energy.gov, 2016; Hopewell et al., 2009). WEEE is a complex waste stream that covers a wide spectrum of different post-consumer products from electric toothbrushes to refrigerators (Cui & Forssberg, 2003). Hence, WEEE is holding a complex mixture of a large number of different materials and components, where plastics constitute around 30 wt% of the waste (Buekens & Yang, 2014; Schlummer et al., 2007). Due to the development of new products and materials, changing legislations and product designs, the waste constantly changes, resulting in new challenges for the recycling industry (Directive 2000/53/EC, 2000; Directive 2012/19/EU, 2012; RoHS recast Directive 2011/65/EU, 2011). At the same time the recycling of plastics has gained increasing interest in the last decades. The high potential for value recovery, stricter legislations and increasing environmental awareness are main drivers for increasing recycling rates in the European Union (Dodbiba et al., 2008; Hischier et al., 2005).

1.2 *Industrial Recycling of WEEE and ELV Plastics*

In commercial mechanical recycling processes plastics are most commonly separated from the other materials after size reduction by shredding. Ferrous and non-ferrous metals are often removed from the plastics by magnetic and eddy current separation (Peeters et al., 2013). For the post-shredder separation of plastics density based separation processes, such as sink-flotation and hydrocyclones are most commonly applied. In a recent study the density distribution of plastics present in Liquid Crystal Display TVs (LCD TVs) has been analysed, which demonstrated that the ABS and HIPS without FRs and plastics with Flame Retardants (FRs) from LCD TVs are characterized by overlapping densities (Peeters, 2015). Consequently, the number of plastics that can be recovered with density based separation techniques from LCD TVs is limited. To separate these plastics with overlapping densities other processes such as mechanical classification, surface wettability or electrostatic separation could be used, since these techniques make use of the differences in the size, hydrophobic or triboelectric properties to separate these materials (Al-Salem et al., 2009; Lungu, 2004; Masoumi et al., 2012). Alternatively, advanced optical separation techniques can be used for plastic separation, since these techniques are able to determine the type of plastic, content of FR additives or colour by spectroscopic analysis (NIR, XRF, visible light). These optical sorting techniques are often applied in an automated on-line system that identifies the shredded pieces and then separates them with ejection nozzles (Masoumi et al., 2012). However, all these separation techniques face the challenge that many plastics possess very similar properties and that the addition of additives such as fillers, fibres or colours strongly influences the properties of plastics. Therefore, a combination of various processes is commonly applied, but in recent studies it has been analysed that the separation of PC/ABS with FRs from LCD TV waste by the combination of commercially applied density based separation processes and advanced optical sorting techniques can only achieve a maximum purity of 79 % (Peeters, 2015).

Furthermore, it should be considered that remaining impurities such as metals, fibres, dirt, glass, wood, paper or other plastics might negatively affect the properties of the recycled plastic. A challenge in the recovery of plastics from mixed waste streams is also the high variety of plastics on the market. Plastics are synthetic materials that are designed specifically for defined purposes by the variation of molar mass, copolymer content, the addition of additives or blending with other plastics. The recovery of one dedicated type of plastic will lead to the recovery of various grades of this plastic. Additionally the quality of the polymer chains varies depending on the degree of degradation (Singh & Sharma, 2008). Since the quality of the recycled plastic defines the price on the market, the quality that can be achieved often determines whether a plastic is mechanically recycled or thermally treated (Gent et al., 2009).

To produce a high-quality recycled plastic from strongly mixed WEEE and ELV waste streams a much higher quality can be achieved by narrowing down the waste stream to individual product types or manufacturing sectors. This paper focuses on the mechanical recycling of ABS without FRs from LCD TVs by means of manual disassembly and sorting based on the results of spectroscopic analysis. The purpose of this paper is to describe the distinct mechanical recycling process and to evaluate the quality of the recycled plastics that can be achieved.

2 CASE STUDY: LIQUID CRYSTAL DISPLAY TELEVISIONS

In the European Union 1,7 million tons of e-waste were generated from screens only in 2014 containing around 30 % plastics (Baldé et al., 2015a; Buekens & Yang, 2014; Cryan et al., 2010). Therefore this waste stream is a reliable source to generate relevant amounts of recycled plastics with high quality. The back covers of LCD TVs are relatively large components. Therefore, the amount that can be sorted per spectroscopic measurement equals up to several kilos of plastic. Additionally, LCD TVs are increasingly dismantled to improve the recovery of precious metals that are present in the printed wiring boards (PWB). However, new electronic products show a decreasing content of these precious metals, that used to be the main economic driver for the PWB recycling, which increases the importance of additional value recovery due to the recycling of other materials present in these products (Zhang & Forssberg, 1997). Hazardous elements as mercury present in some LCD TVs require a separate treatment in form of a depollution step during recycling. Therefore, LCD TVs need to be separated from other WEEE which facilitates a dismantling based mechanical recycling strategy.

The composition of LCD TV back covers, as shown in Figure 1, was analysed in prior research, which demonstrated that eight types of plastics often containing brominated (Br) or phosphor based FRs are commonly used for this application. Currently only the non-Flame Retardant (FR) plastics HIPS and ABS are mechanically recycled from waste streams on an industrial scale. Other FR plastics, such as PC, PC/ABS with FR, HIPS/PPE with FR, HIPS with FR and ABS/PMMA are currently mostly thermally recycled, which is a relatively costly process. Therefore, there is a significant potential to increase the value recovery from LCD TVs by targeting all different types of plastics for mechanical recycling.

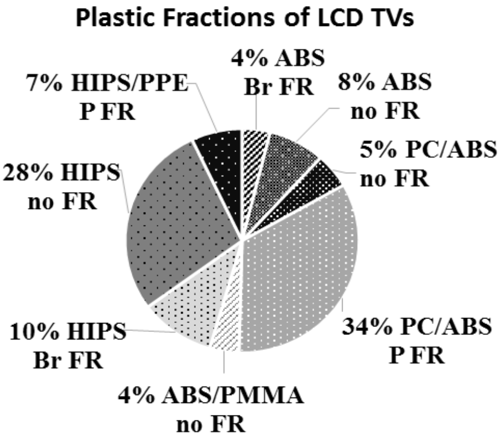

Figure 1. Plastic types found in LCD TVs in Belgium based on the analysis of 2501 back covers in 2015.

3 EXPERIMENTAL

3.1 *Materials*

In total 2501 back covers from LCD TV screens were disassembled in an industrial trial at a recycling company in Belgium. These back covers were then separated based on their plastic type and FR content , where 196 (around 8 %) were ABS without FR that were used for this research. The disassembled post-consumer ABS is compared to a commercial ABS recyclate from WEEE and ELV that was compounded with rubber particles after recycling to improve the impact properties. From all plastics sorted out during the experiment ABS was chosen to be used for initial experiments and to develop the mechanical recycling process, because of a broad knowledge in literature of the behaviour of ABS in a mechanical recycling process. In addition, ABS is mechanically recycled on an industrial scale which already allows comparison of the quality that can be reached.

3.2 *Methods*

A mechanical recycling process based on the identification of the plastic and FRs with Laser-Induced Breakdown Spectroscopy (LIBS) after manual disassembling of the back cover of LCD TVs was developed. A Quantom® LIBS scanner from Bertin Technologies SAS was applied in an industry trial. During the development of the process the feasibility of implementation at an industrial scale was considered and process steps were chosen based on techniques applied in the recycling industry. The disassembled back covers were shredded in an industrial shredder to 50 - 60 mm pieces for transportation/storage and further treatment. A purification step by density separation was included to remove metal insertions, foams and other plastics found in the back covers as well as to separate plastics containing Br FR. The purification of ABS without FRs has been carried out by density separation steps at 1,00 g/cm³ and 1,10 g/cm³. Density values were chosen based on a density analysis of back cover plastics and correlated with values applied in industry (Peeters, 2015). The purification step at 1,00 g/cm³ was carried out in a container with water at room temperature, where the floating fraction was removed. For the purification step at 1,10 g/cm³ a solution of water and NaCl was prepared and the sinking fraction separated. Afterwards, the size was reduced to pieces <5 mm by shredding to receive plastic flakes suitable for further processing. The shredded pieces contained approximately 5 % moisture and were dried at 60 °C in a granulate dryer for about 10 hours to avoid problems during processing. The developed process for the mechanical recycling of plastics is shown in Figure 2.

Approximately 23 kg of shredded ABS without FRs from back cover housings of LCD TVs have been used. The separation process was repeated five times to evaluate the variation in separation efficiency. The efficiency regarding the separation of FRs has been analysed by X-ray fluorescence (XRF) with an Oxford Instruments X-Met 3000 TXR+. For the evaluation of the quality of the recycled ABS injection moulding was applied to produce multipurpose specimens according to ISO 294. For this purpose an Engel ES 200/35 HL was used. A nozzle temperature of 200 °C, mould temperature of 80 °C, holding pressure of 52,5 bar, injection speed of 39 ccm/min, back pressure of 209 bar were chosen as processing parameters. The holding time as well as the cooling time were 20 s. The evaluation of the mechanical properties is carried out according to ISO 527 using a Galdabini Quasa 50 with extensometers for Young's modulus determination. Experiments are carried out at a testing speed of 10 mm/min with a 50 kN load cell. Young's modulus determination was carried out between 100 and 300 N due to unrepresentative results between 0,05 and 0,25 % elongation. The fracture surfaces were analysed using a Keyence VHX-500F microscope. The Charpy impact strength was tested unnotched and flatwise (ISO 179) with a Zwick impact tester and a 75 kgcm pendulum at room temperature.

4 RESULTS AND DISCUSSION

The results of the purification show that about 3,1 % of impurities were separated by density separation at 1,00 g/cm³ (fraction 1). The main part of this fraction are plastics with a lower density as well as small

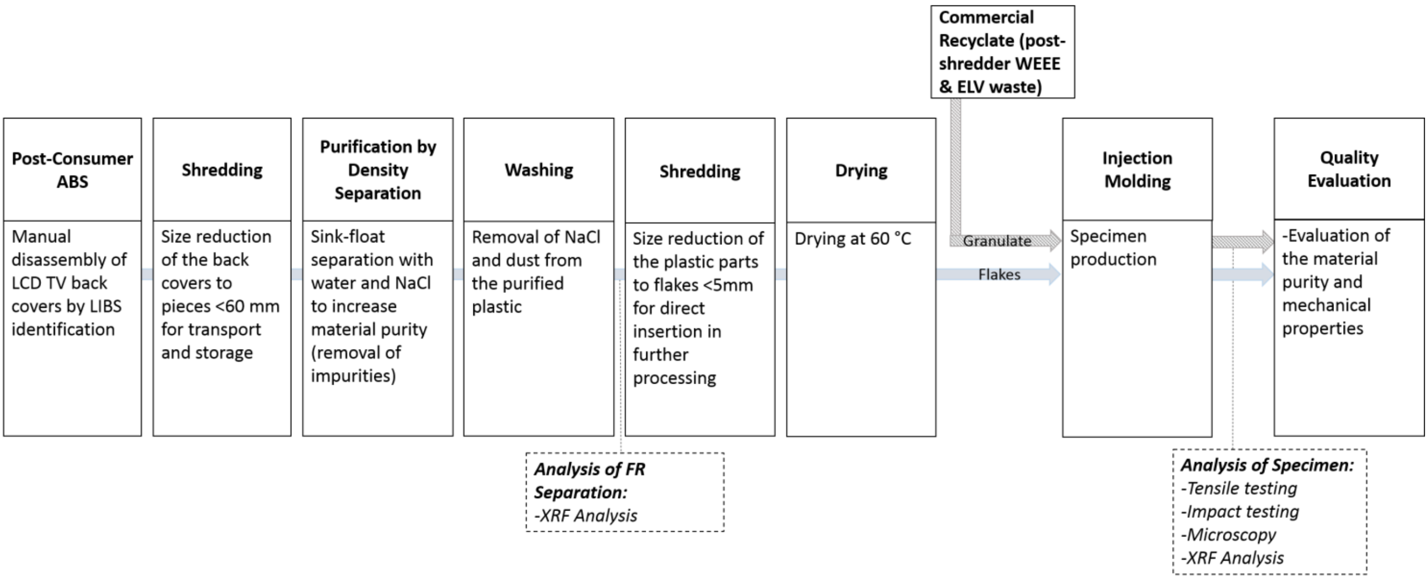

Figure 2. Recycling process for ABS without FR based on product disassembly.

foams, films and labels. The subsequent separation step at 1,10 g/cm³ removes 8,2 % (fraction 3). Metals and plastic pieces containing metal insertions or screws represent 0,9 % of this fraction. Parts of PWBs, rubber particles and pieces of glass are only found in relatively small quantities of around 0,1 %. Main part of this fraction are plastics (7,1 %). The purified ABS fraction (1,00 – 1,10 g/cm³) represents 88,7 % and is used for further processing and testing. Table 1 shows the results of the purification process including standard deviations.

Table 1. Purification Process by density separation of ABS without FR.

Purification Process Output	wt%	Std. Dev.
1 Fraction (<1,00 g/cm³)	3,1	0,68
1.1 Foams	0,2	0,10
1.2 Films, Labels	0,0	0,01
1.3 Plastics	2,9	0,63
2 Fraction (1,00 – 1,10 g/cm³)	88,7	0,64
3 Fraction (>1,10 g/cm³)	8,2	0,51
3.1 Metals, Plastics with Metals	0,9	0,27
3.2 PWB parts	0,1	0,03
3.3 Labels, Rubber, Glass	0,1	0,06
3.4 Plastics	7,1	0,39

A detailed XRF analysis shows that approximately 73 % of the plastic pieces in the separated fraction 3, with a density higher than 1,10 g/cm³, contain between 33 000 and 73 000 ppm of bromine. In the purified fraction 2 around 90 % of the plastic pieces contain less than 1000 ppm bromine, which is defined by the RoHS as the threshold for FR free plastics from WEEE waste (RoHS recast Directive 2011/65/EU, 2011). In only 10 % of the plastics a bromine content of 1000 ppm or higher can be measured. After injection moulding of fraction 2 a bromine content of approximately 235 ppm is measured, which is significantly lower than the RoHS threshold. The purification process can be shown to be an important process step to remove various impurities that are present in the disassembled back covers such as metal inserts or attached foams or labels, which cannot be removed economically by manual disassembly. Additionally, the industrial environment and possible poor signals due to dusty surfaces in the LIBS identification may have resulted in a contamination with plastics during the manual recycling process that could be removed during the purification process.

The comparison of the tensile tests of the purified fraction 2 of disassembled ABS without FRs and commercial ABS recyclate is shown in Figure 3. In this figure the measured values are displayed as bars with standard deviations. A Young’s modulus of 1897 N/mm² can be measured for the disassembled ABS grade and 1851 N/mm² for the commercial grade. The tensile strength results in a similar tendency with 39,4 N/mm² for the disassembled ABS and 33,6 N/mm² for the commercial ABS grade. Differences in the tensile values can be caused by the added rubber particles in the commercial recyclate that might lower the tensile values. Additionally a different mix of ABS types in the commercial ABS recyclate as well as contaminations might affect the tensile properties. The strain at break of the disassembled ABS is with around 7 % higher than the strain at break of the commercial recyclate, but due to a very high standard deviation of 4,5 % no reliable value could be determined. The strong variations in the strain at break values are most likely due to remaining impurities found in the fracture surfaces of the specimen after tensile testing. The microscopy of 30 fracture surfaces showed the presence of labels, metals and plastic pieces of 500 to 2000 µm. Visible yielding regions could determine the impurities as crack initiators. The fracture surface of a tensile bar after tensile testing with an impurity is show in Figure 4.

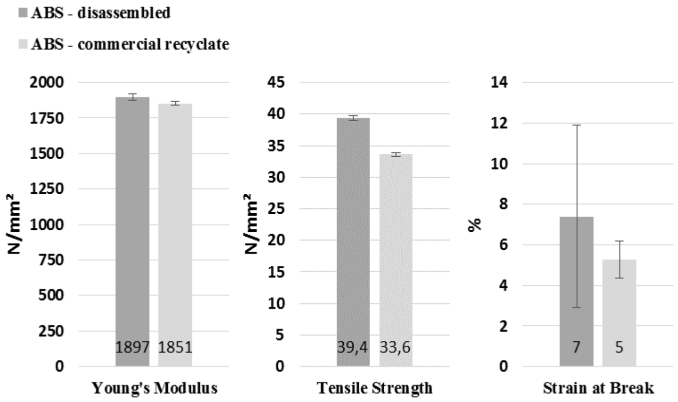

Figure 3. Tensile properties of disassembled ABS from LCD TV back covers and a commercial ABS recyclate.

Figure 4. Fracture surface with impurity.

The Charpy impact strengths of unnotched ABS samples are shown in Figure 5. The disassembled ABS is characterised with an impact strength of 61 kJ/m² which is approximately 4 times higher than the impact strength of the commercial recyclate of 17 kJ/m². When comparing the results it has to be considered that the impact strength of the commercial recyclate was improved by the addition of rubber particles during a compounding process. In addition, it is expected that a compounding step applied to the commercial recyclate negatively influences existing butadiene phase. However, in prior research it was shown that ageing of ABS due to several reprocessing steps affected the impact strength significantly while Young's modulus and tensile strength showed no significant changes after the first reprocessing (Scaffaro et al., 2012). In ABS the butadiene phase is mainly responsible for the impact properties of the material and ageing of the plastic usually affects this phase stronger than the SAN phase (Scaffaro et al., 2012; Tiganis et al., 2002). Another possible reason for the lower quality is that a larger mix of ABS grades with different butadiene contents is present in the commercial recyclate. Therefore, the quality could possibly also be improved by limiting the input of the density based separation process to only LCD TVs. Nonetheless, the significant difference in impact values suggests that a very high quality of the ABS can be achieved by the disassembly of LCD TV back covers.

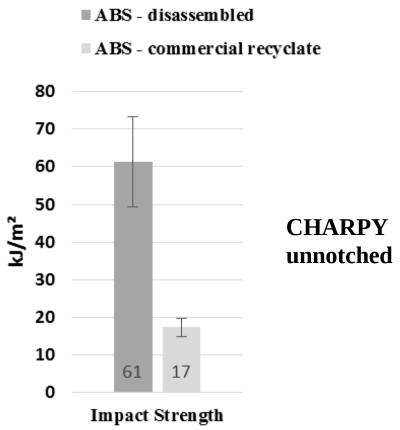

Figure 5. Charpy impact strength (unnotched) of disassembled ABS from LCD TV back covers and a commercial ABS recyclate.

The mechanical tests of the disassembled ABS showed systematically higher values when compared to a commercially recycled ABS grade from various EEE and automotive products. Considering the relatively small amount of ABS present in LCD TVs (approx. 8 %) a very high purity and quality of the recycled plastic could be achieved with the proposed recycling method. The good mechanical properties could allow the use of these recycled plastics in high-end products. Additionally, production or recycling flaws of commercial recyclates could be compensated by the addition of plastics recycled by the proposed method.

5 CONCLUSIONS

In this paper the mechanical recycling based on spectroscopic identification and disassembly of plastic back covers from LCD TVs to achieve high quality plastic recyclates was investigated. The results from mechanical testing suggest that this distinct recycling strategy can lead to a high quality recycled plastic compared to post-shredder separation. The identification of plastic types and FRs by LIBS and the disassembling of the back covers of the LCD TVs leads to a high purity of the recycled plastic. This high purity can be achieved because of the developed mechanical recycling and purification process as well as because of a limited number of ABS grades that are applied for this specific application. The high quality of the ABS recycled after disassembly is demonstrated by high mechanical properties, especially by the high impact strength compared to the commercial recyclate. However, significant impurities were found in the fracture surfaces of the ABS recycled after disassembly. For this reason as well as to further improve the plastic quality and stability future research will investigate the implementation of compounding and melt filtration of plastics separated by manual disassembly and spectrometric analysis.

6 ABBREVIATIONS

ABS – Acrylonitrile Butadiene Styrene
Br – Bromine
ELV – End-of-Life Vehicles
FR – Flame Retardant
HIPS - High Impact PolyStyrene
LCD TV – Liquid Crystal Display TeleVision
LIBS – Laser-Induced Breakdown Spectroscopy
NIR – Near InfraRed
P – Phosphorous
PC – PolyCarbonate
PMMA – Poly(Methyl MethAcrylate)
PPE – PolyPhenyl Ether
SAN-Styrene AcryloNitrile
WEEE – Waste Electrical and Electronic Equipment
XRF – X-Ray Fluorescence

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