Towards a more circular economy for WEEE plastics - Part B: Assessment of the technical feasibility of recycling strategies

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11 ABSTRACT:

12 This two paper series describes a method to develop and evaluate new recycling strategies for
13 WEEE plastics. Part A presents a SWOT analysis that leads to five recycling strategies for the
14 optimal integration of new dismantling based recycling processes for plastic components in an
15 established post-shredder separation infrastructure. In this paper the technical feasibility of the
16 strategies is demonstrated by means of LCD TV back cover housings. The component
17 recycling is shown to produce recycled PC/ABS with phosphorous flame retardants suitable
18 for direct re-application in electronic products. The high quality is characterized by a good
19 mechanical and aesthetical properties as well as a recovered flammability. HIPS with
20 brominated flame retardants was recycled to produce masterbatches. The technical feasibility
21 of this strategy was proven by mechanical and flammability testing. However, the presence of
22 deca-BDE requires this material to be incinerated. A combination of EU legislation research
23 and forecasting shows that the origin of this flame retardant are TV models produced before
24 2008 and restricted concentrations still need to be expected for decades to come. Further, a
25 blending strategy of HIPS/PPE is shown to improve the mechanical properties of post-shredder
26 recycled HIPS. The evaluation of refeeding ABS/PMMA into the post-shredder recycling
27 process of ABS indicates only partial compatibility. Further, complications due to density
28 differences make this strategy more suitable for polymers that are already commercially
29 recycled such as ABS and HIPS. Colour is identified as a key requirements that limits the use
30 of WEEE plastics in high-quality products.

32 **Abbreviations**

- 33 ABS-Acrylonitrile Butadiene Styrene
- 34 Br- Bromine/Brominated
- 35 EoL-End of Life
- 36 FTIR-Fourier Transform InfraRed
- 37 FR- Flame Retardant
- 38 GWFI-Glow Wire Flammability Index
- 39 HIPS-High Impact PolyStyrene
- 40 LCD TV-Liquid Crystal Display TeleVision
- 41 LIBS-Laser Induced Breakdown Spectrometry
- 42 LOI-Limited Oxygen Index
- 43 MB- MasterBatch
- 44 NIR-Near InfraRed
- 45 P-Phosphor/Phosphorous
- 46 PBB-PolyBrominated Biphenyl
- 47 PC-PolyCarbonate
- 48 P(BDE)-PolyBrominated Diphenyl Ether
- 49 PMMA-PolyMehtylMethAcrylate
- 50 PPE-PolyPhenyleneEther
- 51 PWB-Printed Wiring Board
- 52 RoHS-Restriction of Hazardous Substances
- 53 SAN-Styrene AcryloNitrile
- 54 REACH- Registration, Evaluation, Authorisation and Restriction of Chemicals
- 55 SSSP-Sliding Spark SPectroscopy
- 56 SWOT- Strength Weaknesses Opportunities Strengths

57	WEEE-Waste Electrical and Electronic Equipment
58	XRF-X-Ray Fluorescence
59	XRT-X-Ray Transmission
60	

1. Introduction

1.1. Waste Electrical and Electronic Equipment (WEEE) plastics

The amount of Liquid Crystal Display Televisions (LCD TVs) is growing in the coming years with 399 000 tonnes expected to reach End-of-Life (EoL) in Europe in 2020 (Cucchiella et al., 2015). They are characterized by a layered construction with a front cover and back cover housing that are screwed together. These layers are most commonly made of plastic and constitute approximately 15% and 45% of the plastic weight of the product, respectively (Peeters et al., 2013b). The plastics applied in these products are high-tech engineering grades that could make up 43 % of the virgin material price of the entire LCD TV (Vanegas et al., 2017). However, only a limited amount of the value is recovered today. In Table 1 the composition of plastics found in LCD TV back covers are shown based on a LIBS analysis of 2501 back cover housing in Belgium. Acrylonitrile Butadiene Styrene (ABS) is a copolymer that consists of a Styrene Acrylonitrile (SAN) phase made of styrene to provide rigidity and acrylonitrile units that contribute to chemical resistance and the ease of processing of this material and a butadiene phase that largely dominates the impact properties (Utracki, 2001). Blending with Poly(Methyl MethAcrylate) (PMMA) as found in ABS/PMMA adds a good scratch resistance and surface gloss (Nazari Tayebe et al., 2012). High Impact PolyStyrene (HIPS) is composed of polystyrene that is impact modified with butadiene and for flammability, Br FRs are added. The blending with PolyPhenyl Ether (PPE) is relatively expensive, but it improves the flammability resistance and allows the application of P based FRs to the material, as P FRs have shown a better flammability performance if oxygen was present in the polymer chains (Laoutid et al., 2009). Additionally, the PPE content improves the mechanical and dimensional stability at increased temperatures as well as the strength and stiffness (Utracki, 2001). Around 55 wt% of the back covers of LCD TVs contain either Br or P based FRs (Morf et al., 2005; Peeters et al., 2014; Schlummer et al., 2006). While the P based

FRs are not subject to constraints in the recycling process, many Br FRs such as PolyBrominated Biphenyls (PBBs) and PolyBrominated Diphenyl Ethers (PBDEs) are restricted by the European directive on the Restriction of Hazardous Substances (RoHS) and can, therefore, not be reapplied in EEE (European Commission, 2003). Furthermore, some Br FRs such as Octa-BDEs, PBBs and the most recently include deca-BDE are restricted by the European REACH legislation. While it has been shown that plastics containing FRs can be recycled (Delva et al., 2018; Peeters et al., 2014; Tange and Drohmann, 2005), it needs to be safeguarded that restricted substances are removed. Today, X-Ray Fluorescence (XRF) is most commonly used to detect plastics containing Br FRs at concentrations above 2000 ppm (EN 50625) in order to subsequently remove them with ejection nozzles (Hennebert and Filella, 2018). However, only the detection of the element Br is economically feasible in industrial operations without distinction between different Br FR types (Aldrian et al., 2015), which would require relatively slow and expensive analytical techniques. In addition, density treatment can also be used to remove plastic containing flame retardants (Schlummer and Mäurer, 2006).

1.2. Dismantling based recycling of plastic components

Today, mechanical recycling is done by size reduction of WEEE with shredding and subsequent separation of the materials in form of small pieces (Chancerel and Rotter, 2009). A possible alternative to this approach is the dedicated focus on selected End of Life (EoL) products and a treatment by dismantling of large plastic components, such as LCD TV back covers, car bumpers, fridge drawers, laptop back covers or tablet housings. In the case of LCD TVs the plastic back cover housing can be dismantled to access the main Printed Wiring Boards (PWBs), which has been shown to result in a higher recovery of precious metals. However, today the selective disassembly is conducted for the purpose of separating the PWBs while the dismantled plastic housings are processed in a post-shredder based process. From the

perspective of plastic recycling one of the main differences compared to the post-shredder separation process is that the dismantling of back covers allows sorting of large components rather than small flakes. As a result, spectroscopic techniques with contact measurements can be applied, which significantly increases the quality of the spectra used for identification. The following spectrometry techniques can be used to identify the plastic and FR type on component level in a dismantling based recycling scenario:

Fourier Transform InfraRed (FTIR) spectroscopy measures the plastics response to infrared light. The incident light causes characteristic vibrations based on the chemical structure that are visible due to the absorption or transmission of the incident light. In contrast to Near InfraRed (NIR), the mid infrared range is used which allows to identify also black plastics. This technique is commonly used for quality management as it allows a reliable identification of the plastic type. Signoret et al. (Signoret et al., 2019) could show that most relevant WEEE plastics can be discriminated with FTIR. On industrial scale for post-shredder sorting, this techniques is often considered too slow (Becker et al., 2017; Socrates, 2001).

Raman spectroscopy works with the principle of the Raman light scattering effect and delivers spectra similar to FTIR. The technique is seen as complimentary as it is more sensitive to the detection of e.g. C-C, C=C and C≡C bonds where FTIR is more sensitive to the detection of OH bonds (Allen et al., 1999; Socrates, 2001). According to Florestan et al. (Florestan et al., 1994) the technique can also deliver information about TiO₂ and CaCO₃ mineral fillers in plastics if the matrix polymer does not show Raman response in the same region and no fluorescence occurs.

X-Ray Transmission (XRT) analyses the extent to which X-rays are absorbed by a material and determine the atomic density. XRT is capable of differentiating between FR and non-FR

plastics, however, due to overlapping densities no distinction between Br and P based FRs can be made (Peeters et al., 2014).

X-Ray Fluorescence (XRF) on the other hand uses X-rays to excite the atoms of a material in order to create a characteristic fluorescence light response in order to identify the different elements that are present. This technique allows to identify Br and detection limits of 300 ppm have been reported in literature for handheld devices (Aldrian et al., 2015). However, the detection of the specific FR molecule is not possible. The quantification of light elements, such as P is known to be difficult and a testing series for this research in 2015 did not yield results with sufficient accuracy for the detection of P in plastics from LCD TVs.

Sliding Spark Spectroscopy (SSSP) uses a high voltage spark to vaporize a small amount of the plastic surface. The emitted radiation can be measured to differentiate Br and P based FRs. The determination of the plastic type is also possible, but a testing series performed by the authors in 2014 delivered unreliable results for the distinction between ABS and HIPS.

Laser Induced Breakdown Spectroscopy (LIBS) uses a high energetic pulse laser beam to form a plasma and vaporize a small amount of the material surface. Excited atoms emit light and elements present in the surface can be identified (Cremers and Radziemski, 2013). The identification of several plastic types and the detection of Br in plastics has been reported in literature (Anzano et al., 2006; Radivojevic et al., 2004; Stepputat and Noll, 2003). Sorting trials with a LIBS contact measurement in a nitrogen atmosphere showed that this technology is able to detect both plastic type and distinguish between Br and phosphate FRs in LCD TV back cover material.

Most of the discussed spectroscopic techniques are available as table top systems or robust handheld scanners that are suitable for on-site measurements (Aldrian et al., 2015; Sorak et al., 2012). However, dedicated devices and calibrations are necessary to reach relevant

discrimination accuracies. Further, technologies involving lasers or X-rays may require shielding and safety precautions. After sorting of the housings, impurities such as metal inserts or cables can be removed by sink-float separation and compounding with melt filtration will produce plastic granulates (Wagner et al., 2018a). Previous research has shown that the closed loop recycling of the plastic housings is technically possible and can be economically viable (Peeters et al., 2013a; Peeters et al., 2013b; Vanegas et al., 2017). Based on the results of these analysis two Belgian recycling plants also moved from exclusively shredding based process to a hybrid process including a first dismantling step to remove the back cover and the main PWBs and a further shredding based process.

1.3. Waste management strategies for dismantling based recycling

Part A of this two paper series describes a SWOT analysis to define new recycling strategies for WEEE plastics recycling and an economic assessment based on the potential virgin prices of the recycled plastics to make waste management decisions. In part B a technical evaluation of the recycling strategies is conducted to and main practical aspects on the applicability of the strategies are defined. Dismantling based recycling process for LCD TV back covers is taken as a case study and the 5 recycling strategies, defined in part A are shortly summarized as a basis for this paper. The main processes and the integration of the strategies in an existing infrastructure of a commercial post-shredder separation recycling facility are shown in Figure 1. Strategy (1) *Dismantling based recycling of plastics for direct reapplication* processes the single sorted plastic housings to produce granulates that can be used for injection moulding of high quality products. The second strategy (2) *Dismantling based recycling of plastics for masterbatch carriers* allows to recover FR plastics that are present in low volumes and in an additional compounding step adds virgin FRs to create FR rich granulates, so called MBs. They can be later added in injection moulding to FR free granulates to create FR components. In the third strategy (3) *Blending with post-shredder plastic recyclates*, the recycled plastics from the

dismantling process are mixed with post-shredder separated plastics during compounding to improve their quality. In addition, it allows to form streams of mixed plastics in sorting step of the dismantling based recycling process. In the fourth strategy (4) *Refeeding in post-shredder recycling process*, the plastic types which do not result in a significant quality improvement that justifies the recycling by the dismantling process can be reintroduced in the post-shredder separation process, given that the plastic type is already recycled. The fifth strategy (5) *Thermal treatment* aims for plastic types that cannot be recycled by mechanical recycling.

While the strategies show a high potential for additional value recovery and the possibility to recycle plastics that are today incinerated, the technical feasibility and achievable quality of the different strategy needs to be evaluated. In this paper the defined strategies are put into practice and tested by mechanical, flammability and aesthetical characterizations, as well as injection moulding of test component and industrial evaluations. The practical experience achieved by the trials allows to define main quality criteria to identify important practical aspects that need to be considered when applying the strategies.

2. Materials and Methods

The dismantling based recycling process for all plastics included the processing steps dismantling of the plastic housings, spectroscopic sorting, shredding, sink-flotation, fine-shredding, washing, drying, compounding with melt filtration (Figure 1). The dismantling was done by manual disassembly and subsequent sorting was done with a Quantom ® LIBS scanner with a contact measurement with nitrogen atmosphere as developed by Bertin Technologies in Belgium. The identification of the co-polymers has shown to require some further improvement for the HIPS and HIPS/PPE as well as the ABS and ABS/PMMA distinction. HIPS with Br FRs used in this research was generated in a second sorting trial with an XRF hand scanner (Oxford Instruments X-Met 3000 TXR+) to determine a minimum Br content of 2000 ppm and subsequent FTIR spectroscopy to determine the polymer type. This was done due to the limited

availability of the LIBS technology for this research. The sorted back covers are shredded to pieces of approximately 50-60 mm. The density cuts were chosen on the lower and on the higher end of the density distribution data analysed in previous research (Peeters et al., 2014). Sink-float separation was conducted in pure water at 1,00 g/cm³ and water with sodium chloride and calcium chloride for higher density cuts. Afterwards, the plastics were washed with high pressure water to remove salt and dust. Fine-shredding to <5 mm and drying in a DS 503-507 granulate dryer from Piovan Plastics Technologies removed moisture and allowed the pieces to be inserted Werder & Pfleiderer ZSK 25 twin-screw extruder with a melt filtration. The compounding step was used to remove remaining impurities from the plastics in a molten state without the use of additives. The mesh size was 315 µm for all materials, as commonly used in industry. For the evaluation of the quality, the obtained materials were processed with an Engel ES 200/35 HL into multipurpose specimen type B according to ISO 294. The injection moulding and compounding parameters for the different plastics were chosen according to the standards, ISO 2580 for ABS, ISO 15103 for HIPS/PPE, ISO 8257 and ISO 2580 for ABS/PMMA, ISO 2897 HIPS with Br and ISO 7391 and ISO 2580 for PC/ABS. The tensile testing of the all the recycled plastics was done with a Galdabini Quasa 50 at 10 mm/min at room temperature. For the evaluation of the Young's modulus, extensometers were applied and the determination was done between 100 and 300 N, due to unrepresentative results in the range between 0,05 and 0,25 % elongation. Charpy impact strength was tested with a Zwick impact tester (unnotched and edgewise) according to ISO 179 at room temperature. Between 10 and 20 tensile bars were used for every mechanical test. For every analysis the median values are calculated and the 25 % and 75 % quartiles are shown in box plots.

2.1. Dismantling based recycling of plastics for direct reapplication

PC/ABS with P FRs was used as a case study material for this strategy. Compounding with melt filtration was conducted at 220 °C. Injection moulding parameters for the production of

tensile bars were a nozzle temperature of 250 °C, a mould temperature of 80 °C and a holding pressure of 760 bar. Mechanical testing was based on tensile tests and impact tests compared to a commercially available recycled post-shredder separated PC/ABS. The flammability was evaluated based on case study requirements by the standard for test for flammability of plastic materials for parts in devices and appliances (UL94) and glow wire flammability tests (IEC 60695) conducted by industrial partners on 1,7 and 3,2 mm thick specimens. In this research an industry validation of the quality was conducted in four case studies of thin-walled injection moulded products. The internal frame of a payment terminal with the dimensions 25x69x175 mm and a wall thickness of 1,6 mm, a small part of an electrical cabinet with the dimensions 79x51,5x8 mm and a wall thickness of 2 mm, an adapter piece for a socket with the dimensions 19x50x15 mm and an inner part of a socket with the dimensions of 55x55x 3mm. The injection moulding trials were conducted without any product redesign to support the use of recycled plastics.

2.2. Dismantling based recycling of plastics for masterbatch carriers

Recycled HIPS with Br FRs was used for this testing series. These materials were treated in a compounding step with melt filtration at 200 °C. In addition, a second compounding step was used to incorporate additional virgin FRs in order to achieve a highly filled plastic granulate. A virgin MB was produced with the same amount of Br FR added for comparison. In a third compounding step the 8 % of MB were mixed with 92 % of HIPS granulate to produce 2 samples with virgin HIPS mixed with the virgin MB and recycled MB and 2 samples of recycled HIPS with virgin MB and recycled MB. The parameters for the MB production are confidential, but the last compounding step for the mixing was conducted at 200 °C. Injection moulding of tensile bars was done with a nozzle temperature of 210 °C, a mould temperature of 45 °C and a holding pressure of 633 bar. The virgin HIPS used in the tests was a Kumho 425 N without FRs. The Br content and flammability tests, UL94 flammability, Limited

Oxygen Index (LOI) and Glow Wire Flammability Index (GWFI), were conducted by industrial partners. For the chemical analysis of the BR FRs, two sampling trials of 100 and 350 kg of HIPS with Br FR were conducted. Structured sample taking according to the spoon method (Petersen et al., 2004) was used to reduce the samples to 25 kg of pieces of approximately 6 cm and then repeated to achieve a sample size of 200 g with pieces of 5 mm, suitable for chemical analysis. The chemical analysis was based on gas chromatography–electron capture detector, gas chromatography-mass spectrometry and liquid chromatography-mass spectrometry and conducted by two independent testing institutions.

2.3. Blending with post-shredder plastic recyclates

For the blending series, HIPS/PPE recycled by dismantling was mixed with a commercial post-shredder HIPS recyclate. Compounding with melt filtration was conducted at 200 °C for the HIPS/PPE. The mixing with HIPS was conducted in form of granulates, by hand to make compositions of 0, 10, 50, 70, 100 wt% HIPS/PPE. Afterwards, the mixed granulates were introduced and further mixed in injection moulding with a back pressure of 209 bar. Further, injection moulding parameters were a nozzle temperature of 220 °C, a mould temperature of 60 °C and a holding pressure of 791 bar. Since both commercial recyclate resulted in a different maximum pressures in the injection moulding process compared to the plastics from dismantling based recycling, the holding pressure needed to be adapted to 712 bar. The holding pressures applied for the mixtures HIPS/PPE with commercial HIPS were adapted accordingly.

2.4. Refeeding into the post-shredder recycling process

The direct testing by refeeding into an industrial post-shredder separation process was not possible for this case study. The effects of mixing ABS/PMMA recycled by the dismantling based process and commercial post-shredder ABS recyclate were tested at lab scale by blending as described for HIPS/PPE and HIPS. The blending was done at 0, 10, 50 and 100 wt%. of

dismantled ABS/PMMA. Compounding with melt filtration was conducted at 200 °C for the ABS/PMMA. Injection moulding parameters were a nozzle temperature of 250 °C, a mould temperature of 60 °C and a holding pressure between 696 and 854 bar, depending on the mixture. Final evaluation is based on discussion with industrial recyclers.

2.5. Thermal treatment

This treatment option is based on a literature review and discussions with industrial partners (Tange and Drohmann, 2005; Weber and Kuch, 2003, Nnorom and Osibanjo, 2008).

3. Result and discussion

3.1. Dismantling based recycling of plastics for direct reapplication (1)

The dismantling based recycling strategy for direct reapplication strategy was applied to the PC/ABS with P FRs of the LCD TV back covers. The quality of the material was assessed by industrial partners in injection moulding trials of an inner frame of the payment terminal, a part of the electrical cabinet, an adapter piece and the internal component of a light switch (Figure 2). The trials resulted in a very positive feedback for all four case studies. The processing behaviour of the recycled plastic allowed a reapplication without any redesign of the mould or adaptations to the process. Comparable or reduced cycle times were achieved compared to currently applied virgin plastics. For the inner electrical housing the concern of criticality for silver streaks of the recycled PC/ABS was expressed, which can occur due to moisture or gas formation by degradation (Harper, 2006; Rosato and Rosato, 2012). However, the material performed better than a commercial plastic recyclate and it is expected that stabilization against degradation and improved processing can avoid this problem. Overall, the injection moulding trials resulted in a confirmation of a very quality of the recycled plastic by the industrial partners.

The mechanical performance of the recycled PC/ABS is characterized by a tensile modulus of 2103 MPa, a tensile strength of 56 MPa, strain at break of 16 % and an unnotched impact strength of 123 kJ/m². The values are shown in Table 2 along with the performance of a commercial post-shredder PC/ABS grade for reference. The dismantled PC/ABS is not modified in a compounding step and the values therefore reflect a baseline quality that can still be improved. Impact modifications result in a higher impact strength and a reduction in the Young's modulus, which is expected to be the case for the post-shredder recycled PC/ABS (Liu and Bertilsson, 1999). Further, P FRs present in the dismantling based recycled PC/ABS can negatively influence the mechanical properties (Zhang et al., 2012). The results can be confirmed by tests performed by industrial partners and is in line with previous testing of recycled ABS, which showed an improved mechanical performance compared to post-shredder separated plastics (Wagner et al., 2018a). Thus, a very good mechanical performance of the dismantling based recycling of plastics for direct reapplication can be concluded.

The flammability testing of the recycled PC/ABS with P FR by GWFI tests of specimen with a thickness of 1,7 mm resulted in a pass when tested at 750 °C, while at 850 °C the material failed the test. UL 94 testing resulted in V0 grading with a specimen thickness of 3,2 mm and V2 with a thickness of 1,7 mm. The flammability performance depends both on the material as well as on the component design. Therefore, flammability tests were conducted on the case study components and could fulfil the requirements set by industrial partners. Hence, it could be shown that FR properties of the P FRs could be recovered sufficiently well to reapply the recycled material without the addition of virgin FRs. Further improvement of the flammability by virgin FRs is considered an option.

The close cooperation with manufacturers indicates that the aesthetical requirements are very important, especially for potential visible applications. The colour and gloss of the PC/ABS

were evaluated on thin-walled sample plates for visual inspections. The Lab colour values were $L^*31,22$ $a^*-0,18$ $b^*-1,97$ and fit approximately with RAL 9004. The gloss values were 101 at an angle of 60° and 100 at an angle of 85° which is characterized as high gloss. Colouring trials showed that grey shades of $L^*44,74$ $a^*-0,56$ $b^*-2,90$ can be produced by the addition of 9 wt% of white TiO_2 MB (Wagner et al., 2018b). However, the additional colouration will reduce the flammability and mechanical properties and sacrifice the economic performance of the recycling strategy. Thus, the colour requirements that are mostly defined based on virgin plastics by the final component manufacturer will limit the applicability of the plastic.

It can be concluded that when a dismantling based recycling for direct reapplication is applied for PC/ABS with P FRs a high quality recyclate can be produced which is characterized by good processing, mechanical, flammability and aesthetical properties. In discussion with component manufacturers, the processing behaviour, colour as well as material price and availability have been identified as key criteria for the application of recycled PC/ABS.

3.2. Dismantling based recycling of plastics for masterbatch carriers (2)

The dismantling based recycling of plastics for MB carriers was tested with HIPS with Br FRs. Previous to compounding these plastics showed a concentration of 8,2 wt% of Br. The density treatment used for purification resulted in a relatively low yield of 72 wt%. Changes in the density cuts could increase the yield of the process, however, possibly influencing the Br concentration as a higher density with increasing FR content has been reported (Peeters et al., 2014).

In a first testing series 100/0, 25/75, 50/50 and 0/100 mixes of virgin HIPS with recycled HIPS were compounded and injection moulded to show the effect the recycled and Br FR containing plastic has on the mechanical properties of a virgin HIPS. The tensile strength increased with the recyclate content from 19,6 to 21,4 MPa and the strain at Break decreased from 42 to 27 %

according to the composition. The Young's modulus were measured between 2098 and 2056 MPa for the virgin plastic and the 25/75 and 50/50 mixture. The 100 % recycled plastic showed a lower Young's modulus of 1847 MPa and was tested before compounding with melt filtration. A significant decrease in impact strength was observed with 112 kJ/m² for the virgin plastic and 46-36 kJ/m² for the mixtures where no relevant difference between the 25/75 and 50/50 mixtures and the 100 % recyclate could be observed. This could be due to the presence of the FRs, which were 8,2 % for pure recycled plastic, approximately 6 % for the 25/75 mixture and 4 % for the 50/50 mixture (Radloff et al., 1996). A lower impact strength of the recycled plastic itself might also contribute to this decrease.

The influence of using the recycled plastics as a carrier materials compared to a virgin carrier is investigated by mechanical properties and flammability testing of samples produced in injection moulding. Mechanical tests of virgin HIPS with virgin and recycled MBs resulted in comparable Young's modulus, tensile strength and strain at Break (Table 3). The impact strength of 94 kJ/m² for the recycled MB and 74 kJ/m² for virgin MB can also be seen as comparable due to a high deviation in results. However, there is a significant difference between the impact strength of the pure virgin HIPS of 112 kJ/m² and the impact strength of the recycled material. A decrease in impact strength is expected due to the Br content (Cui et al., 2007). For the recycled HIPS with virgin and recycled MBs also no relevant differences in the properties were observed. The GWFI and LOI flammability tests also resulted in comparable results for the virgin and recycled MB. The results of the mechanical tests show that the use of recycled HIPS with Br FRs can be used for MB carrier materials without influence on the mechanical properties compared to virgin HIPS. While significant advantages of the pre-existing Br content in the recycled HIPS could not be properly analysed in these tests, the production of MB carriers based on recycled plastics allow a higher recyclate content of the final plastic. In addition, results show that the adopted strategy allows to target a Br rich

waste stream and efficiently separates it from the not Br containing stream. One limitation for this plastic stream is the colour that can be achieved by using this MB, as it could only be produced in black. This limits the final applicability of the recycled MB as colour is seen as one of the mayor hurdles in the use of recycled plastics. Most commonly standardisation bodies and equipment manufacturers do not take this into account at the moment of design of a new product.

To comply with RoHS and REACH requirements the substances present in the recycled HIPS with Br FRs were analysed. In total 4 analysis showed the presence of 0,4, 0,47 and 0,5 wt% of deca-BDE in the samples. One analysis resulted in no detectable amount (<20 ppm) of deca-BDE. No other restricted substances were measured. Further, a minimum mix of four different types of Br FRs was found in a total of 5,5 wt % (thereof 0,4 wt% deca-BDE). Previous research based on discussions with industry experts had resulted in the conclusion that deca-BDE is not expected in LCD TVs (Peeters et al., 2015). However, a literature review of relevant regulations and directives was conducted to investigate if the presence of deca-BDE is likely to be found or was the result of single contamination. Deca-BDE was excepted from the restriction for the application in electronic products until the first of July 2008, when a threshold of 0,1wt% became active. In March 2019 the restriction will be expanded, as REACH will restrict deca-BDE as a substance or in any article with a threshold of 0,1 wt%. In 2018 a lower threshold level of 0,001 wt% was proposed by the European Parliament, which resulted in the beginning of 2019 in a final threshold of 0,001 wt% for deca-BDE if present in form of a substance and 0,05 wt% for all BDEs cumulatively, if present in mixtures and articles (Council of the EU, 2019). The measured deca-BDE content in the samples is therefore 8-10 times higher than the allowed threshold. Thus, the strategy of using this material as a carrier for MBs is not possible and the material needs to be incinerated for decontamination in a thermal treatment with sufficiently high temperatures (Weber and Kuch, 2003). Peeters et al.

(Peeters et al., 2015) developed a forecasting model based on sales and lifetime distribution data to predict material compositions in future waste streams. In Figure 3 a modified version of their graph displays the forecasted composition of LCD TVs to come to EoL in function of the production year. The concentration of 0-0,5 % deca-BDE as measured in the HIPS with Br FR of this stream in 2016 is expected to be caused by TV models produced before 2008, which make up around 42 % of the waste stream. Assuming the relative concentration of deca-BDE in the mix of all Br FRs does not significantly change, the levels will in the worst case still be more than 5 times above the required threshold by 2025. The presence of this restricted FR is still expected to be above the allowed limits for more than 20 years for HIPS with Br FRs from LCD TV back covers. To afterwards reinvestigate the applicability of this recycling strategy, a comprehensive risk analysis along with systematic quality testing and effects of FR mixes will be necessary to better understand the properties and stability of the recycled FRs.

3.3. Blending with post-shredder plastic recyclates (3)

HIPS/PPE is one of the smaller fractions of the LCD TV back cover housing plastics making it difficult to create a new market for this plastic for direct reapplication. The blending of HIPS/PPE with a commercially recycled HIPS from post-shredder separation shows that the Young's modulus, the tensile strength and impact strength have a linear correlation with regards to the HIPS/PPE content (Figure 4). As these mechanical properties are significantly higher compared to these of the HIPS post-shredder recyclate, the HIPS/PPE could be used to improve the mechanical properties of the commercial recyclate. At the same time, the strain at break does not seem to be influenced if blended with amounts smaller than 10 wt% and shows a decrease when mixed with 50 wt% or more. The mostly linear correlation of properties and compositions of HIPS/PPE and HIPS indicates an additive behaviour and therefore a full compatibility of the two polymers (Ragaert et al., 2017). The compatibility of PPE in HIPS is also confirmed by literature (Utracki, 2001), where PPE and PS are compatible and no

interaction with the rubber phase in the PS is expected to influence the impact properties. In addition, the PPE content in HIPS increases the heat deflection temperature and therefore improves the thermal stability of the mechanical properties (Utracki, 2001).

There are two options to apply this strategy: HIPS/PPE can be processed by the dismantling based recycling process and mixed with post-shredder recycled HIPS by weight before compounding. As an alternative, HIPS/PPE and HIPS back covers can be sorted together after the dismantling process to be later mixed with post-shredder HIPS. Both options might require adaptations of the compounding parameters and will allow to buffer quality flaws or improve the properties of the post-shredder HIPS. The latter option reduces processing complexity and increases the volumes, however, the ratio needs to be measured to allow a controlled quality improvement. Otherwise, blending of <10% HIPS/PPE without influences on quality is not considered to justify the additional operational costs. Depending on the PPE content all mixing strategies will result in a higher density of the blend. As a consequence, if the plastic comes to the End-of-Life for a second time the blend is likely to be separated together with brominated and other heavy plastics and subsequently incinerated or landfilled.

3.4. Refeeding into the post-shredder recycling process (4)

ABS/PMMA makes up only 4 % of the LCD TV back cover housing plastics. In contrast to HIPS/PPE the blending of post-shredder recycled ABS and ABS/PMMA from dismantling based recycling did not show a linear behaviour of the mechanical properties according to its blend composition, where the strain at break as well as the Charpy unnotched impact strength did not show a relevant increase when mixed with up to 50 % of ABS/PMMA (Figure 5). Kim et al. (Kim et al., 1993) however observed a strong drop in impact strength of PMMA blends with ABS with 28 wt% SAN phase while the blending with ABS with a 17 wt% SAN phase resulted in a smooth variation in properties according to the blend composition. The impact strength is strongly influenced by the SAN-PMMA miscibility as it affects the size and

distribution of the rubber particles (Kim et al., 1993). Dedicated processing with dismantling based recycling of the ABS/PMMA is therefore not a viable strategy. To avoid incineration and increase recycling rates, the dilution of ABS/PMMA in low concentrations in post-shredder recycled ABS is possible without significantly influencing the mechanical properties. The dismantled back covers can be sent directly to the post-shredder process with other input material. The feasibility of this strategy, however, depends on the applied post-shredder separation techniques. The separation with a densimetric process is commonly done around 1,10 g/cm³, which would separate the ABS/PMMA with a density of 1,12-1,19 g/cm³ together with the heavier plastics that are incinerated or landfilled (Peeters et al., 2014). For the HIPS and ABS without FRs commercial post-shredder separation processes are available and they can be re-fed into this process. However, a potentially higher quality that can be achieved by the dismantling approach will be lost and not strategically used (Wagner et al., 2018a).

3.5. Thermal treatment (5)

In this research the thermal treatment is seen as an external option for material that cannot be treated in mechanical recycling (Tange and Drohmann, 2005; Weber and Kuch, 2003). The collaboration with the recycling facility showed that today, HIPS/PPE, ABS/PMMA, ABS with Br, HIPS with Br and also the PC/ABS with P FRs go to the thermal waste treatment option. Due to the optimized application of the innovative recycling process also the PC/ABS with P FRs, the HIPS/PPE and the ABS/PMMA could be recycled only remaining Br plastics to be thermally treated. A quantitative environmental and economic comparison to further evaluate the most suitable of these End-of-Life treatment options would be a relevant possibility for future research. Also novel chemical recycling approaches, such as the Creasolv® process that allows to dissolve polymers and remove Br FRs, are considered promising options for the future (Nnorom and Osibanjo, 2008; Schlummer et al., 2016a, 2016b).

4. Conclusions

Five diversified strategies that aim for an optimized integration of a dismantling based recycling approach in an existing post-shredder recycling facility were tested. The (1) *dismantling based recycling of plastics for direct reapplication* was tested with PC/ABS with P FRs, which demonstrated that flammability, good mechanical and aesthetical properties can be recovered without the addition of virgin additives. Thus, allowing a reapplication in thin-walled injection moulded products directly replacing virgin plastics. The (2) *Dismantling based recycling of plastics for masterbatch carriers* was tested with HIPS with Br FRs. Mechanical tests showed that with MB contents of 8% no significant differences between virgin or recycled MBs could be observed. However, between 0 and 0,5 wt% of deca-BDE requires a (5) *thermal treatment* of this waste stream. The (3) *blending with post-shredder plastic recyclates* of dismantling based recycled HIPS/PPE has been shown to improve the Young's modulus, tensile strength and impact strength of post-shredder HIPS with full compatibility of the two polymers. The (4) *refeeding into the post-shredder recycling process* strategy was tested for ABS/PMMA and ABS which resulted in only partial compatibility. While this strategy is the preferred option for plastics that are already recycled by post-shredder separation, such as ABS or HIPS, density differences of new plastics might not allow the application of this strategy. The communication with industry showed that the processing behaviour, e.g cycle time, strongly influences the economic performance and is considered one of the most important criteria for material selection. Most producers seek a "drop in" solution, where a virgin plastic is directly replaced by a recycled plastic without any changes to product or process design. However, the main hurdle in all mechanical recycling strategies was the colour of the plastic recyclate. Plastics in WEEE are mostly black or grey. Changes in colour require virgin additives which influences the economics of the plastics recycling and can also influence the mechanical or flammability properties. Strict material requirements, mostly set for virgin

plastics with full colour freedom, should be reconsidered to support the use of recycled plastics. It should be taken into account that all strategies have to deal with relatively small volumes of plastics for recycling. Hence, a main practical hurdle for their applications is that most available processes of the post-shredder separation are designed for large, continuous throughputs. In this context the processing of small volumes is economically, logistically and organisationally challenging. An integration of more flexible and batch-wise processing in the state of the art recycling operations could open new possibilities in plastics recycling.

Forecasting results showed the presence of deca-BDE caused by TVs produced before 2008 is still to be expected in this waste stream for decades to come, long after chemical restrictions were put in place. This shows the importance to include these considerations in legislative processes as well as the need for reliable data about substances present in waste products. Future research should focus on strategies and systems to efficiently remove restricted substances during recycling, as this is one of the mayor obstacles on the way to a circular economy for WEEE plastics.

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 660

Figure 1. Visualization of the process steps of the dismantling based recycling process with integration options in a post-shredder recycling process.

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666 Figure 2. Injection moulded internal component of a light switch produced with recycled

667 PC/ABS with P FR.

668

Figure 3. Forecasted composition of LCD TVs produced before 2008.

Figure 4. Mechanical properties of dismantling based recycled HIPS/PPE, post-shredder recycled HIPS and their blends.

Figure 5. Mechanical properties of dismantling based recycled ABS/PMMA, post shredder recycled ABS and their blends.

680 Table 1. Composition of LCD TV back cover housings (Wagner et al., 2018a).

Plastic	FR	Composition [%]
ABS	-	8
HIPS	-	28
ABS/PMMA	-	4
PC/ABS	P	39
HIPS/PPE	P	7
HIPS	Br	10
ABS	Br	4

681

682

Table 2. Tests of recycled PC/ABS with P FRs compared to commercial post-shredder recycled PC/ABS.

	PC/ABS	PC/ABS
Properties	dismantling based	post-shredder
	recycling	recycling
Tensile Modulus – 10 mm/min	2103 MPa	1830 MPa
Tensile Strength – 10 mm/min	56 MPa	48 MPa
Strain at Break – 10 mm/min	16 %	18 %
Impact Strength - unnotched	123 kJ/m ²	134 kJ/m ²
GWFI @750 °C	Pass (1,7 mm)	No FR grade
GWFI @850 °C	Fail (1,7 mm)	No FR grade
UL 94 Flammability Test	V0 (3,2 mm)	No FR grade
UL 94 Flammability Test	V2 (1,7 mm)	No FR grade

687 Table 3. Mechanical and flammability tests of four mixes of 92% virgin (V) and recycled
688 (R) HIPS and 8 % MBs.

Properties	V-HIPS	V-HIPS	R-HIPS	R-HIPS
	V-MB	R-MB	V-MB	R-MB
Br	2,79 %	2,4 %	10,2 %	9,9 %
Sb2O3	0,82 %	1,6 %	3,7 %	4 %
Tensile Modulus – 10 mm/min	2126 MPa	2186 MPa	2130 MPa	2122 MPa
Tensile Strength – 10 mm/min	19 MPa	20 MPa	21 MPa	21 MPa
Strain at Break – 10 mm/min	43 %	42 %	18 %	22 %
Impact Strength - unnotched	75 kJ/m ²	94 kJ/m ²	28 kJ/m ²	32 kJ/m ²
GWFI @850 °C	Pass (3mm)	Pass (3mm)	Pass (3mm)	Pass (3mm)
GWFI @850 °C	Fail (1,7mm)	Fail (1,7mm)	Pass (1,7mm)	Pass (1,7mm)
LOI	21 %	22 %	26 %	23 %

689