Recyclability of plastic waste extracted from electrical devices

I Turku* and T Kärki

1 LUT University, Yliopistonkatu 34, 53850 Lappeenranta, Finland

*Corresponding author and e-mail: Irina.Turku@lut.fi

Abstract. The mechanical recyclability of acrylonitrile butadiene styrene, high impact polystyrene, polycarbonate and polypropylene extracted from electric equipment waste was studied. The secondary raw material was tested on a broad range of working characteristics, such as melt flow index, thermal properties as well as tensile properties of injection molded samples were detected. Parameters were compared with parameters of unprocessed counterparts presented in the literature.

1. Introduction

Waste from electrical and electronic equipment (WEEE) grows up very fast that requires optimization of its utilization process. About 17 wt% of total WEEE stream is plastic components. The most abundant plastics in WEEE are acrylonitrile butadiene styrene (ABS), high impact polystyrene (HIPS), polypropylene (PP), polycarbonate (PC) as well as polyvinylchloride, polyybutylene terephthalate and polyamide, which counts up to 70 wt% of total plastic collected [1]. According to statistics, only 25% of e-waste plastics is recycled [2] that is lower than the average rate of post-consumer plastic utilization. Thus, from 27.1 Mio of plastic waste generated in Europe in 2016, about 31.1 % was recycled, 27.3 % landfilled and 41.6 % went to incineration [3]. The main challenges for EEE plastic utilization are the complexity of the different types of plastic used in EEE products, black colored materials, lack technics for separating all plastic types into individual streams and limiting understanding of the effect of the mixing of different plastics on the physical properties of the recycled materials [4]. In addition, the presence of halogen-containing fire retardants and heavy metals negatively influence on the recycling rate.

Carbon-based reinforcing fillers, namely carbon fibers, graphite, carbon nanotubes, carbon black (CB), represent a great interest in the composites manufacturing due to their multifunctional properties including high strength-to-weight ratio, high electrical and thermal conductivity, and fire resistance among others. Many useful properties of CB like variety in structure, low cost and capability of CB particles to form strong attachments to elastomer molecules without using expensive compatibilizer make CB very important and most widely used filler in rubber and plastic industry [5]. Concerning polymer waste recycling it was shown that incorporation of 3-10 % of CB resulted in the improved tensile strength of a recycled PP, ABS and PS up to 12-23 %, depending on the plastic source [6]. The tensile properties of PP and recycled natural rubber glove blend was improved by loading of CB [7]. CB had a positive effect on the tensile and impact strength properties of unprocessed PP and ABS blend [8]. Comparative analysis of compression and injection molded PP/CB nanocomposite showed that injection molded samples have a superior tensile property than compression molded [9].

The aim of this study was to estimate the recyclability of ABS, HIPS, PC and PP extracted from EEE waste. Pieces of plastic were reduced until microsize and then were used for melt flow index
and thermal analysis as well as samples for tensile properties were injection molded. The influence of carbon black filler on the tensile properties was also studied. In addition, elemental analysis of the waste was performed by using energy-dispersive X-ray spectroscopy (EDS).

2. Materials and methods

2.1. Plastic source
There are four types of WEEE plastic, ABS, HIPS, PC and PP were used in this study. The materials were obtained from the post-consumed electronic devices, Figure 1. The coffee machines plastic parts were sources of PP, the refrigerator plastic parts were the source of PC, different plastic parts of computer were ABS and HIPS sources. Plastic pieces were additionally analyzed with FTIR (data not shown). Carbon black was Raven ®1170 grade, mean particle size 21 nm and surface are 107 m²/g (IMCD Finland Oy).

![Figure 1. Sources of plastic waste.](image)

2.2. Samples manufacturing
The plastic fragments were reduced to 2 cm - sized flakes by low-speed granulator Shini SG-1635N equipped with a 5 mm sieve. The samples for testing were prepared in a Boy 30 injection-molding machine. The parameters used for injection molding were the following parameters: $T_1=160 \, ^\circ\text{C}$ and $T_2=170 \, ^\circ\text{C}$ for ABS, PS and PP, respectively; and $T_1,T_2=210 \, ^\circ\text{C}$ for PC; injection pressure 8.0–9.1 MPa and injection time 3 s. In order to improve nanofiller dispersion, two-step injection-molding procedure was applied, e.g. samples were crushed after the first injection molding and molded again.

2.3. Testing
Melt flow index (MFI) was measured by using Dynisco LMI 5000 (Dynisco, Franklin, MA, USA) in accordance with standard EN ISO 1133-1.

The surface morphology of the samples was studied with an SEM, Hitachi SU3500 (Chiyoda, Tokio, Japan). Surfaces were observed directly after processing. Elemental analyses of the blends and single polymers were performed with EDS X-ray spectroscopy (EDS) (Thermo Scientific, Waltham, MA, USA).

The tensile properties test was performed according to EN-310 standard on a Zwick Z020 machine (Ulm, Germany). The cross-head speed was 2 mm/min for modulus testing and 50 mm/min for the other measurements. The gauge length was 25 mm. The test samples conditioned according to the above standard. Tests were carried out with 10 sample replicates.

Thermal analysis was performed by mean a thermogravimetric analysis (TGA) with a linear temperature increase (Netzsch DSC TGA 204 F1 Phoenix®). The sample, approx. 10 mg of the specimen was heated from 29 °C to 900 °C at a rate of 10 °C/min under a helium atmosphere of 40 mL/min at a constant flow rate.
Table 1. Elemental composition of ABS, PS, PC and PP, wt%; standard deviation is given in parenthesis

| Element | C    | O    | Mg  | Al  | Si  | Ti  | Ca  | S    | Cl   | P    | N    | K    | Na   | Cu   | Mo  |
|---------|------|------|-----|-----|-----|-----|-----|------|------|------|------|------|------|------|-----|
| ABS     | 97.1 | <0.1 | 0.3 | 0.04| 1.1 | -   | 0.66| <0.03| 0.1  | 1(4) | <0.02| <0.01| -    | 0.05 |
|         | (0.1)| (0.2)| (0.2)| (0.2)| (0.05)| (0.03)| (0.03) |       |
| HIPS    | 99   | -    | 0.2 | 0.04| 0.6 | 0.2 | -   | -    | <0.01| -    | -    | -    | <0.01| -    |
|         | (0.2)| (0.2)| (0.01)| (0.05)| (0.08) |       |       |       |
| PC      | 83   | 16   | 0.4 | 0.04| -   | <0.01| 0.4 | -    | <0.01| -    | <0.01| <0.01| -    | -    |
|         | (2)  | (2)  | (0.04) | (0.04) |       |       |       |       |
| PP      | 99   | -    | 0.06| 0.25| 0.17| 1   | 0.1 | <0.01| -    | -    | -    | -    | -    | 0.01 |
|         | (0.3)| (0.01)| (0.2) | (0.03) | (0.1) | (0.03) |       |       |

3. Results and discussion

3.1. SEM-EDS analysis

Results of SEM-EDS analysis of secondary plastics are displayed in Table 1. As it can be seen recycled plastic contains different chemical elements which related to the additives used for a properties modification. Elements amount changes from traces to several percents. Distribution of detected elements is not uniform that is seen in the large standard deviation values, often exceeding the average amount of specified element. The main conclusion that can be made on the basis of the result, plastic waste does not contain toxic elements, such as heavy metals, Cd, Pb, and halogens, Br, Cl, except ABS which have traces of Cl. Halogen-containing compounds which can present in plastic can be attributed to fire retardants. Absence of Br among detected chemical elements can be explained by the fact that only very insignificant amount of plastic waste, 2.5% of total WEEE plastics, contain Br based fire retardants [10].

![SEM micrograph of 3 wt% CB containing plastics fracture surfaces.](image)

There are a broad range of other elements, such as Mg, Ti, Al, Na, K, Ca, Mo, Cu, S, P and O, originated from additives that are usually applied in plastic production or contaminants. Metal-containing fire-retardant Mg(OH)₂ and Al(OH)₃ are sources of Mg and Al, respectively. Phosphorus-containing flame retardants are widely used in the plastic industry [11]. The small amount of silicon, Si, which is a component of sand, can have originated from soil impurities. However, SEM analysis showed that PP waste contains glass fibers (GF), Figure 1, that can explain enhanced detected amount of silicon there. Nitrogen was detected in ABS due to it is a component of acrylonitrile. Oxygen is a component of PC that explain its 16 wt% detected amount. Oxygen was detected in trace amount in
ABS as well. Oxygen is a component of many chemicals including fire retardants, TiO$_2$ and other compounds often used in plastic manufacturing.

### 3.2. MFI analysis

Table 2 presents the measured MFI parameters for the recycled polymers and parameters found in the literature for virgin ones. For the comparative analysis of our samples and virgin ones, the database [12] was used where some range of MFI values is present. As it can be seen measured MFIs are fitted in the values range of the virgin counterparts, however, due to the absence of information/knowledge about recycled material applied grade it is difficult to make conclusions about morphological changes during plastic usage and recycling. In fact, MFI is sensitive to environmental impact and thermomechanical stress, e.g., during lifecycle and reprocessing. It was reported that the MFI of low-density polyethylene decreased from 2.31 g/10 min to 0.02 g/10 min after 100 extrusions [13].

### 3.3. Heat resistance and thermal stability

Experimental results of the thermal degradation of the polymers, mass losses and the corresponding differential thermogravimetry (DTG) curves, are shown in Figure 2. Pyrolysis of all studied polymers occurs in a one-step decomposition under the applied conditions. Polycarbonate is known as plastic which has good thermal stability and left a large fraction of char after burning [14,15]. As it is seen degradation started at about 430 °C with maximum weight loss at 483 °C and no mass loss happened after 650 °C. After the finish of pyrolysis about 20 wt% of char was left, that is significantly more compared to other tested plastics; 4.5, 2 and 3 wt% for ABS, PS, and PP, respectively. Other studied polymers started to degrade significantly early with peaks of mass loss at approx. 420, 430 and 454 °C for ABS, PS, and PP, respectively. Usually, plastics burn incompletely if some amount of inorganic additives, e.g. fire retardants, contaminant and/or reinforcing fillers are present in recycling materials.

#### Table 2. Parameters of extruded filaments for recycled samples and their virgin counterparts found in the literature [12].

| Material/parameter | $T_{	ext{ex}}$(°C)/mass | Density (g/cm$^3$) | MFI (g/10min) | MFI, virgin (g/10min) |
|--------------------|--------------------------|-------------------|--------------|---------------------|
| ABS                | 220/10kg                 | 1.06(0.06)        | 35.4(4)      | 2.1-36              |
| HIPS               | 200/5kg                  | 1.005(0.19)       | 4.9(1)       | 2.3-8.2             |
| PC                 | 300/1.2kg                | 0.996(0.16)       | 24.8(5)      | 1.5-24              |
| PP                 | 230/2.15kg               | 0.796(0.135)      | 15(5)        | 0.3-27              |

### 3.4. Mechanical properties. Influence of CB on the tensile properties

The tensile properties of the recycled plastics and their virgin counterparts reported in the literature are listed in Table 3. According to the results, the tensile properties of the recycled materials are fitted into the range of the virgin counterparts and some values even exceeded the maximal value. Thus, the tensile strength and modulus of PP were slightly higher than the highest values reported for virgin PP. This might be attributed to the presence of a reinforcing filler(s). As it was detected by SEM analysis, glass fibers were used in some PP based EEE, Figure 2.

CB is known as a reinforcing additive, which improves mechanical properties and durability of polymers. However, the properties of modified material will depend on the CB dispersion level. Incorporation of CB, 1-5 wt%, had no significant effect on the mechanical properties of the plastics, Table 3. The tensile properties started to increase only for PC when 5 wt% was loaded. In all other cases, the tensile strength was unchanged or decreased slightly with CB loading. The tensile modulus, however, increased in comparison to unfilled polymer almost for all cases. The increase in the modulus was due to the stiffness of CB is higher than polymer. Lohar and Jogi [8] studied the influence of CB on mechanical properties of PP/ABS blend and showed that the tensile properties of the blend increased to a maximum at 2.5 wt% of CB loading and then decreased with increased CB percentage. This properties droplet was due to CB particles aggregation. Uniform dispersion of CB resulted in the increased the tensile properties of the PP/CB nanocomposite with increasing CB.
content (1-30 %) [9]. Composites morphology inspection with SEM, Figure 2, showed that CB was aggregated that might negatively influence the performance of the material. The CB stay aggregated despite the double extrusion procedure applied for the processing of the samples.

![Figure 2: SEM images of composites with different CB contents.](image)

**Figure 2.** SEM images of composites with different CB contents. The aggregation of CB particles negatively influences the mechanical performance of the material.

**Table 3.** Tensile properties of virgin [12] and recycled plastic filled with different amount of carbon black; standard deviation is given in parenthesis.

| Plastic | Virgin Reference | 1 wt% CB | 3 wt% CB | 5 wt% CB | MOR, MPa | MOE, GPa | MOR, MPa | MOE, GPa | MOR, MPa | MOE, GPa | MOR, MPa | MOE, GPa |
|---------|------------------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|----------|
| ABS     | 29-57            | 1.5-3.1  | 41(0.9)  | 2.3(0.2) | 41(2)   | 2.3(0.1) | 40(2)   | 2.4(0.04) | 39(0.8)  | 2.5(0.1) |
| HIPS    | 16-30            | 1.6-2.4  | 33.7(0.8)| 2.2(0.04)| 33(2)   | 2.17(0.1)| 33.5(1)| 2.2(0.03) | 32.6(0.9)| 2.2(0.03)|
| PC      | 54-67            | 1.9-2.7  | 57.6(0.3)| 2.2(0.04)| 51(2)   | 2.17(0.04)| 56(2)   | 2.4(0.07) | 58.5(0.7)| 2.3(0.03)|
| PP      | 16-31            | 1-1.7    | 32.7(1.7)| 1.8(0.1) | 32(2)   | 1.9(0.04)| 32(0.6)| 1.9(0.06) | 32(1)    | 2(0.06)  |

4. Conclusions
On the basis of the provided analysis, it can be concluded that characteristics of recycled material are comparable with related characteristics of their unprocessed counterparts. Elemental analysis showed the presence of additives, however, toxic chemicals were not detected. Thermal analysis showed the presence of some inorganic material. Melt flow indexes of recycled materials were comparable with virgin counterpart values found in the literature. In order to improve the mechanical properties of recycled material, carbon black (CB) filler was added. Results showed that CB has no significant effect on mechanical performance. Tensile samples fracture micrographs showed that CB particles were not uniformly dispersed in the polymer, mostly they stay aggregated.

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