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## Original Article

# Remanufacturing using end-of-life vehicles and electrical and electronic equipment polymer recyclates - a paradigm for assessing the value proposition

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## ABSTRACT

Over 400 million tonnes of plastics are produced on earth every year and are the most used materials on the globe. As a result, polymeric material-based waste has been accumulating, and now can be found not only in designated waste tips on land, but also as pollution all over the world. Therefore, it is timely to accelerate global recycling plans to minimize the use of natural resources. At present, end-of-life vehicles (EOLV) and electrical and electronic equipment (EEE) are two major sources of waste. Such waste includes a combination of polymers, which are hard to separate. Because the properties of blends of polymer are not well-understood, the recycling opportunities for such wastes are hindered. Hence, this paper aims to extend the knowledge concerning the re-processing of such recycled polymers, as a first step towards developing a more effective circular manufacturing economy. Three types of recycled polymers were tested under various processing conditions, and the effects of six different process parameters were investigated, including chip size and morphology. The results confirm that properties and processing behaviour of recycled materials can be highly variable, and hence difficult to predict. This highlights the importance of better selection and screening of scrap polymers, to remove undesirable content from the material batch. Knowing the exact constituents of each batch of material to be recycled means that appropriate process settings can be selected, to achieve better material properties in the recycled end product. In addition, energy and materials costs can be reduced by optimizing process parameters such as the set temperatures, pressures, and product dimensions. Tests carried out with different recycled polymer chip sizes indicate that size grading would be worthwhile, as would the reforming of chips to flattened discs or strings. Such additional processing could represent considerable value proposition to the recycled material preparation industry.

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## 1. Introduction

### 1.1. Context

Plastics are one of the popular and commonly used materials in the modern world. The global production of plastics exceeded approx.  $359 \times 10^9$  kg in 2018 (see Fig. 1) and a four-fold increase in production tonnage is forecast by 2050 [1].

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Obviously, such a massive increase in the production capacity of plastics would also lead to the generation of similar tonnage of waste each year; hence the demand for recycling would also increase rapidly. At present, the accumulation of polymeric wastes has become a global issue and both manufactures and consumers have been forced to re-think the current take-make-waste extractive industrial model, and develop strategies for reusing these materials. The International Organization of Motor Vehicle Manufacturers reports that 95 million cars and commercial vehicles were manufactured in 2016 [2], and the Electrical and Electronic Equipment (EEE) industry also manufactures product with a significant polymer content. Undoubtedly, these two industries contribute

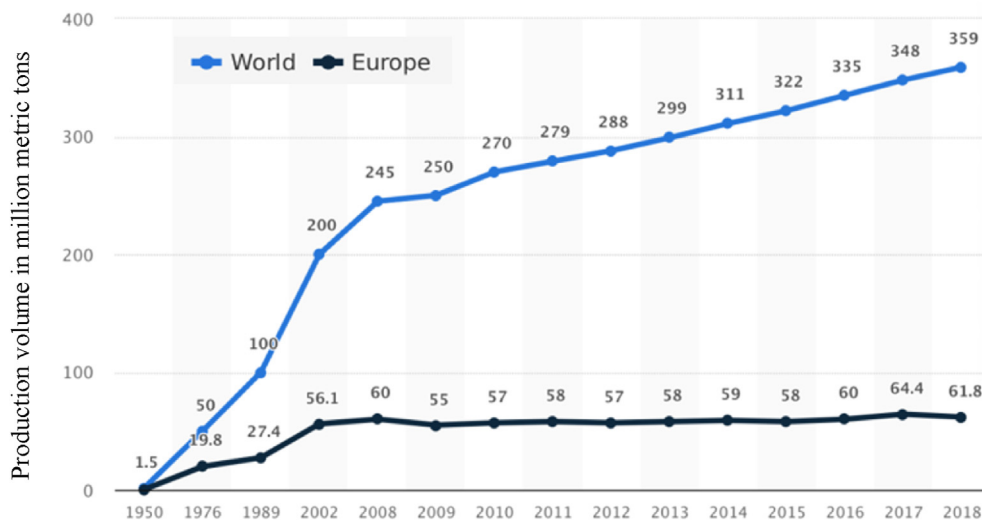


Fig. 1. Production of plastics worldwide from 1950 to 2018 [1].

massively to the global production of polymeric waste, year on year, and research into the feasibility of developing a more circular economy for polymer recycling is timely.

Although the obvious solution to the problem of waste is to recycle, there are significant obstacles to be overcome in the determination and control of the material properties of the plastic materials derived from recycling [3,4]. While it is probably within today's capability to improve the reprocessing, and to measure and control the resulting materials at a suitable level for use in non-critical applications, a new approach would need to be adopted for the control of through-life polymer materials. Such an approach would require a means of marking the first-use material and either controlling its scrap and reuse process, or marking at the material level, so that it can be recognized during re-processing [5]. Some of the necessary capabilities exist, and some are potentially possible but are yet to be developed.

The purpose of the activities presented in this paper is to demonstrate actions that can be feasibly taken to determine polymer type and volume fraction content in a batch developed from mixed recycled polymer material. Since the major impediment to the use of recycled plastics lies in the uncertainty as to their composition, and therefore material performance, the study reported here focuses on the effects of polymer mixtures in the recycled material. We look at the impact this has on both the material processing and on the material properties of the end recycled material product. Better understanding of the significance of polymer content variation and resulting properties variation will enable an informed approach for future, more systematic, Design for Recycling methods as well as a systematic quality control testing approach for as-recycled materials. The question of cost and the efficiency of the recycling process remains another important issue, as does the control of polymer products that might enter future batches of material for recycling. While high duty components might require virgin polymer to ensure appropriate capability, mixed material might still be suitable for medium duty components that are yet subject to some level of control over composition and process life. Although our present focus is on the mechanical performance of the recycled materials, other interests, such as aesthetic qualities are also important to the recyclability of polymers.

In addition to reporting the experimental investigation, we also consider the appropriateness and applicability of managing recycled polymer materials through standards such as ISO 10303–235

“Engineering properties and materials information” [6,7]. The specific sample material we chose to examine, Axplas® MEP54 resins, are acquired from post shredder treatment of EoLV materials. Furthermore, the main composition includes PP and ABS which are typical content of EEE wastes as well.

## 1.2. A short background review

There are a wide range of recycling techniques for tackling the evolving waste problem, which fall under three main categories: mechanical, thermal, and chemical [8]. Industrially viable methods for materials separation remains a major issue [9–12]. Business related challenges for the polymer recycling industry include: the lack of markets for secondary or recycled materials; the high capital cost for recycling plant machinery; the lack, or inadequacy, of recycling technologies; and tight environmental and industry regulations [13–16]. Much previous research has focused on technology aspects of recycling [17–22].

Many End-of-Life Vehicles (EoLVs) manufactured since 2010 have a high content of polymeric materials with significant potential for recycling, but instead is becoming a growing sector for landfill world-wide. Options for disposal by incineration are limited and Circular Economy requirements are gaining increasing interest [21–34]. Waste from Electric and Electronic Equipment (WEEE) comprises polymers, polymer composites and other vital materials; however, there are many inevitable problems which hinder its recycling, and are discussed in the extensive literature [35–52]. Global directives have been established to tackle these problems, for example, the restriction of hazardous substances (RoHS – 2002/95/EC), and the recycling or disposal of WEEE (WEEE – 2002/96/EC). Phthalate polymers, including butyl benzyl phthalate (BBP), dibutyl phthalate (DBP) and diisobutyl phthalate (DIBP) have also been covered in recent evolutions of these directives [53]. Moreover, the majority of the common issues associated with polymer processing (such as the effects of use of fillers, fluctuations in energy consumption, product quality, melt thermal quality and so forth) that are reported in the literature [70–75] should also be applicable to the processing of recycled polymeric materials as well.

### 1.2.1. Characterisation techniques and re-manufacturing methods used in recycled polymers

The processing required for the extraction of reusable polymer from highly compacted and multi-materials structures, such as in

EoLV and EEE [45], involves the methods of crushing, separating, and shredding [4]. The identification of the polymer constituents can be achieved using X-ray fluorescence (XRF) analysis followed by Fourier Transform Infrared (FTIR) analysis [19]. Gas chromatography is commonly used for the quantitative analysis of polymers and other organic compounds [54]. Acrylonitrile Butadiene Styrene (ABS) is a typical polymer type that can be recycled from EoLV and WEEE waste and is commonly used as a feedstock for 3D printing [76]. Effects of multiple re-processing have also been studied [55,56].

### 1.2.2. Test methods for polymers

The necessary polymer material characterisation tests for mechanical performance include the standard tensile and flexural tests. Additional tests, for example the Charpy impact test, can be carried out to determine properties such as scratch resistance and surface hardness [57]. Dynamic mechanical analysis (DMA) is useful to measure the material viscoelastic property as a function of the temperature gradient.

Tests for informing process and processability include Differential Scanning Calorimetry (DSC), which is commonly used to inspect the non-isothermal crystallisation of the resins. Thermogravimetric Analysis (TGA) is also a useful technique as it can inspect matter residue and content during the thermal process [57]. Some common test temperature parameters are the glass transition temperature ( $T_g$ ), the melting point ( $T_m$ ), and the crystallisation point ( $T_c$ ); however, in industrial settings, parameters such as the Heat Distortion Temperature (HDT) and softening point temperature (Vicat) of the polymer materials are also important. For polymer processability, viscometers provide an off-line measurement of polymer viscosity.

The surface quality of materials or composites can be examined using microscopy. Here, Scanning Electron Microscopy (SEM) is the standard method for morphology analysis. Optical microscopy can also be used to show fibre distribution patterns. X-ray Fluorescence (XRF) and X-ray Diffraction (XRD) methods are also useful to inspect element content and molecular structure in a mixture of materials.

### 1.2.3. Case studies of new uses for recycled polymers

The GENVIND project [58] investigated the recycling of glass fibre reinforced plastics (GFRP) for filler materials. Another construction industry application presented by Gómez et al. [44] uses recycled polymers to replace the aggregate and sand used in conventional concrete. Casey et al. [59] investigated the use of recycled Low-Density Polyethylene (LDPE) and High-Density Polyethylene (HDPE) polymers as bitumen modifiers.

There are other applications of recycled polymers, such as feedstock for fused filament fabrication (FFF) manufacturing process, which is a 3D printing process [60,61]. A typical recycled polymer, such as the Axplas® MEP54 investigated here, contains a high proportion of PP and ABS thermoplastics. Moreover, if thermosets can be modified by dynamic covalent bonds, it has been proposed that they too have the potential to be multiply recycled as 3D printing feedstocks [62].

## 2. Standards and control of materials properties

### 2.1. Limitations of the simplistic Waste Hierarchy approach

The well-known “Waste Hierarchy” or Lansink’s Ladder [63] (Fig. 2) illustrates a hierarchy of the desirability of outcomes for waste materials. Although it provides a simple message in a simple way, there are some dangers in taking such an over-simplistic approach. For example, in this paper, we consider the recycling

### WASTE HIERARCHY - Step Up & Go Green



Fig. 2. Waste Hierarchy – Step Up & Go Green (Lansink’s ladder) [63] Open Source.

and circular economy of polymers, which implies level C in the hierarchy, and level D when we consider the burning of polymers for energy production. What is not made clear in the hierarchy is the need to store and pass detailed information about the waste product material system: one wants to know, “Is it safe to put in Landfill, Incinerate or burn for Energy; or does it require special disposal methods before it can be rendered safe?” If an answer to that is not forthcoming, then the acts of Recycling or Reuse merely postpone this problem for a later time.

Another over-simplification lies in the notion that any scrap material, from any unknown source, and subject to any through-life usage or environmental contamination, can be rendered recyclable given appropriate treatment. Even where a material is not actually toxic or directly dangerous, it may have adverse effects on other materials that it is mixed with on recycling, leading to tainted material that cannot be made fit for purpose. Mixing unknown recycled feedstock runs the risk of contamination of the whole circular economy for that material type. For example, consider the history of the steel making industry, which has routinely reused scrap iron in its processing since the industry began. By the beginning of the 20th century, the effect of impurities and additives was increasingly understood and better monitored, and as a result, standards were written to specify requirements and limitations on composition and material processing [64]. It is interesting to note that during WWII, when it was more difficult to obtain materials in the quantity required, standards were relaxed, but the need to specify those standards was maintained. At the present time, British Steel has issued an Environmental Product Declaration (EPD) for steel rails and sections [65], indicating an expectation that nearly 100% of that steel will be recycled or reused. Nevertheless, the material being produced during 2020 contained just over a quarter recycled content: all aspects of material sourcing and processing are subject to relevant standards [66].

### 2.2. Standards

What is true for steel can be applied to all materials: the common messages are that it is important to capture data concerning:

- Material composition
- Material processing
- Through-life duty and environment

And to specify all such material according to relevant and sustainable standards. Standards must be relevant: in the first instance there must be standards for all the most common forms of material, and in the longer term this list should expand. Standards must be sustainable: any future planning for reuse or recycling must work

seamlessly with standards that can be developed and applied today.

Work to develop standards, for application to materials recycling, should be built on three principles. The first, and arguably the most obvious, is to recognize the practical and achievable measurement methods, as well as the manufacturing processes and the through-life conditions for that material. This information comprises the data that needs to be stored and addresses the issue of data relevance. The testing activity described in this paper sets out first steps to create a methodology for identifying the key pieces of relevant information to maximize the effectiveness and uptake of recycling of polymers in the near-term. The tests and the processing methods are commonly available at both laboratory and factory scale. The results obtained here are indicative for planning upscaling activity.

The second principle is that of having a data storage architecture that is sufficiently well-designed that it can hold the necessary information. This is essential since without a standard data structure, the data that needs to be stored cannot easily be exchanged between engineering data systems. This is the principle of “data interoperability”, and it is this that ensures that a standard remains sustainable [7,67,68]. The relevant ISO standard in this case is ISO 103030–235, which has been extensively reviewed for application to through-life product management in the composites industry by McMillan et al. [6].

The third principle is that of marking the material, at the point of production, or at some later point in time, so that source or composition can be tracked in the future. It is common to mark valuable engineering components with serial numbers or to assay potentially valuable materials. With increased recognition of the need for through-life data recording and end-of-life planning, engineered parts marked with serial numbers will be the easiest to recycle in the near-term, and their composition will be well-known. Non-serial marked parts coming from highly engineered products such as vehicles and electrical goods are less likely to have predictable properties, and for materials like polymers, where many different types of additives are employed, there is a challenge of sorting and recyclability. Assaying potential recyclates on the basis of individual chips of millimeter size, and in real time, is unlikely to become a feasible technology for many years to come. An alternative might be to mark future polymers with Graphene Quantum Dots (GQDs) [69], or using some other nanotechnology, to implant a serial number or database within the material at nanoscale.

In the case of polymers used for mechanical product applications, the material properties of most interest can be grouped under three headings: mechanical, rheological, and thermal. The mechanical properties are indicative the ability of the polymer product to withstand in-service loading; the rheological properties provide information as to the manufacturing processability of the materials; and the thermal properties provide effective temperature limits on both in-service and processing environments. There are of course other properties of importance, but for the present study, these three classes of property are taken to represent the process of materials characterisation for recyclates.

It should be noted that all materials characterisation tests have associated standards. In terms of mechanical properties, modulus and strength, for both tensile and flexural tests, are the most common. For processability, the shear modulus ( $G$ ) includes both the shear storage modulus that relates to the elastic response of materials and also the shear loss modulus that relates to the viscous response. It is on the basis of these commonly used test methods, that we base the materials re-manufacturing trial activities described next.

### 3. Materials and experimental methods

#### 3.1. Materials used in the experiments

For this study, three different recycled polymers were used: Axplas® MEP54, and two unknown samples designated S1 and S2. Axplas® MEP54 is described by the supplier, Axion Polymers (UK), as “Mixed Engineering Plastic” and is created from a materials fraction with a density of at least  $1.12 \times 10^3 \text{ kg m}^{-3}$ . Axplas® MEP54 polymers are recovered from advanced post shredder treatment of EoLV materials and processed through a sophisticated cleaning and separating process. The main composition of Axplas® MEP54 contains rich PP with regular particle size, ABS and rubber content is also existed. The detailed composition of Axplas® MEP54 is presented in Table 1. Plastic wastes from EEE have similar compositions. It is used as a control sample in this study. The samples S1 and S2 were also supplied by Axion Polymers from their production lines, but information as to their composition was not disclosed. This is to ensure an unbiased investigation into their nature and behaviour. No further information as to their composition has been disclosed at the time of writing.

#### 3.2. Separation and identification

Samples of the three raw materials, Axplas® MEP54, S1 and S2, were prepared using laboratory density separation techniques. A beaker, partially filled with water, was placed on the magnetic stirrer plate and Magnesium Sulphate was added to create a solution. The concentration of Magnesium Sulphate was adjusted until the density of the solution reached  $1.3 \times 10^3 \text{ kg m}^{-3}$ . The density was measured by a hydrometer. The raw material chips were then added to the solution, to separate them by density. The PVC and other impurities formed the sink fractions and the target polymers remained in the float fraction. A Fourier-transform infrared spectroscopy (FTIR) was also performed to figure out the compositions and proportions of MEP54, S1 and S2 (before and after density separation).

#### 3.3. Compression moulding and machining

The nature of recycled polymer chips before processing is shown in Fig. 3(a). A small scale LOSCA compression moulder was used to prepare test samples of  $80 \times 80 \text{ mm}$  as shown in Fig. 3(b) and (c). The moulder was heated to the set temperature used for each test condition (which was between 170 and 210 °C) before materials were placed in the cavity. The press was closed slowly, and a low pressure was applied for a few minutes before it was re-opened to release any volatile gases and closed again. After the sample was consolidated, the mould was allowed to cool to a temperature below 100 °C. Finally, the mould was opened, and the sample was removed.

**Table 1**  
The detailed materials composition of Axplas® MEP54.

Material	Percentage range
PP	26%
ABS	16%
PS	2%
PCABS	23%
HIPS	10%
Rubber	19%
Other	>4%

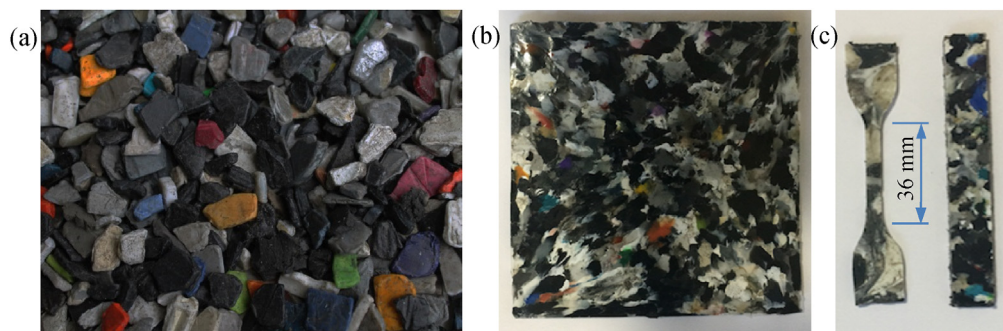


Fig. 3. Photographs of (a) recycled polymer chips before processing, (b) a compression moulded 80 × 80 mm sample, and (c) tensile and bending test specimens.

The effect of moulding process parameters on the MEP54 derived material was investigated. Six control parameters were varied in the moulding process: moulding time, moulding temperature, moulding pressure, chip size, sample thickness, and the non-plastic content, such as rubber. Test samples were prepared from each mould batch. In each set of experiments, one parameter was varied while the other five parameters were fixed.

### 3.4. Testing techniques

For each test, five specimens were tested, and the mean values were reported. All the measurements were performed at  $23 \pm 2$  °C and at least 24 h after the sample was moulded. First, the 80 × 80 mm moulded samples were cut into the rectangular flexural test specimens of 80 × 10 mm. Then a lathe machine was used to prepare the tensile test dog-bone specimens.

#### 3.4.1. Tensile tests

The measurements were made following the ISO 527–1:2012 standard. Tensile specimens were tested using an Instron 3365 (USA) universal testing machine with a crosshead speed of 1 mm per minute. Each specimen was 72 mm long, 12 mm wide and between 0.8 and 3 mm thick with a 36 mm gauge length as shown in Fig. 3. A stress–strain plot was obtained from each test, from which both the Young's modulus and tensile strength were obtained.

#### 3.4.2. Flexural tests

Bending specimens were tested using an Instron 4301 (USA) universal testing machine with a crosshead speed of 1 mm per minute, according to ISO 178:2010 + A1:2013. Each specimen was 80 mm long, 10 mm wide and between 0.8 and 3 mm thick. A stress–strain plot was obtained from each test, from which both the flexural modulus and flexural strength were obtained.

#### 3.4.3. Rheology tests

The rheological properties were determined by using a flat plate rheometer. The test samples were solid discs of 25 mm in diameter and roughly 1 mm in thickness. Frequency sweep tests were conducted in the frequency range of 0.1–100 rad s<sup>-1</sup>, with 1% strain amplitude. The aim was to explore the relationship between the elastic modulus and the loss modulus for increasing frequency. Temperature ramp tests were also conducted over a temperature range of 50–210 °C, with a heating rate of 5 °C per minute, which simulated the heating rate in the compression moulder. This was undertaken to explore the possibility of a relationship between the elastic and loss modulus with temperature.

#### 3.4.4. Differential Scanning Calorimetry (DSC)

The melting and crystallisation behaviours of samples were measured under a liquid nitrogen atmosphere using a DSC (model QS100) with 3–10 mg samples in an aluminium pan. Using this technique, it was anticipated that the  $T_g$  of each of the constituent polymers of the recycled material would be individually identifiable. The first step was to increase the temperature from 25 °C to 220 °C. Subsequently, in the second step this process was reversed by reducing the temperature back from 220 °C to 25 °C. Finally, in the third step, the temperature was increased to 220 °C again. The aim of the first heating and cooling was to erase the thermal history. The heating rate was 5 °C per minute, to simulate the heating rate in the compression moulder.

#### 3.4.5. Scanning Electron Microscope (SEM)

Morphology analyses were performed by SEM in a LEO EVO 40 XVP electron microscope. Samples were mounted on bronze stubs and then coated with a gold layer (3 nm), using an argon plasma metallizer (sputter coater PELCO 91000), to make them electrically conductive during imaging. Secondary electron images of the materials were obtained by using an accelerating voltage from 5 to 20 kV. The magnification level range was between 500 and 20,000×.

## 4. Results

### 4.1. Identification of composition

Fourier-transform infrared spectroscopy (FTIR) was performed to determine the compositions and proportions of the constituents of the material sample MEP54, before and after density separation, and also of the material samples S1 and S2. The results showed that the main constituents of these “recyclates”, which were derived from EoLVs and EEE, were Acrylonitrile Butadiene Styrene (ABS), High Impact Polystyrene (HIPS), Polypropylene (PP), Polycarbonates (PC)/ABS and Polyamide (PA) also known as Nylon. Additionally, small amounts of Polyvinyl Chloride (PVC), Polyethylenimine (PEI) and Polyethylene (PE) were also seen, meaning that only a portion of the recyclates could be used directly in moulding. Components like PVC and PA are unsuitable to be moulded by compression together with other constituents as they will have different behaviour and hence should be removed.

### 4.2. Tensile properties of MEP54 derived samples

The homogenized tensile strength and Young's modulus were obtained by tensile testing of the MEP54 derived samples and values compared for the six control parameters defined in Section 4.3. All the related sub-figures of modulus and strength were plotted in the same scale for the ease of comparison. As shown in Fig. 4(a), both the tensile strength and Young's modulus are

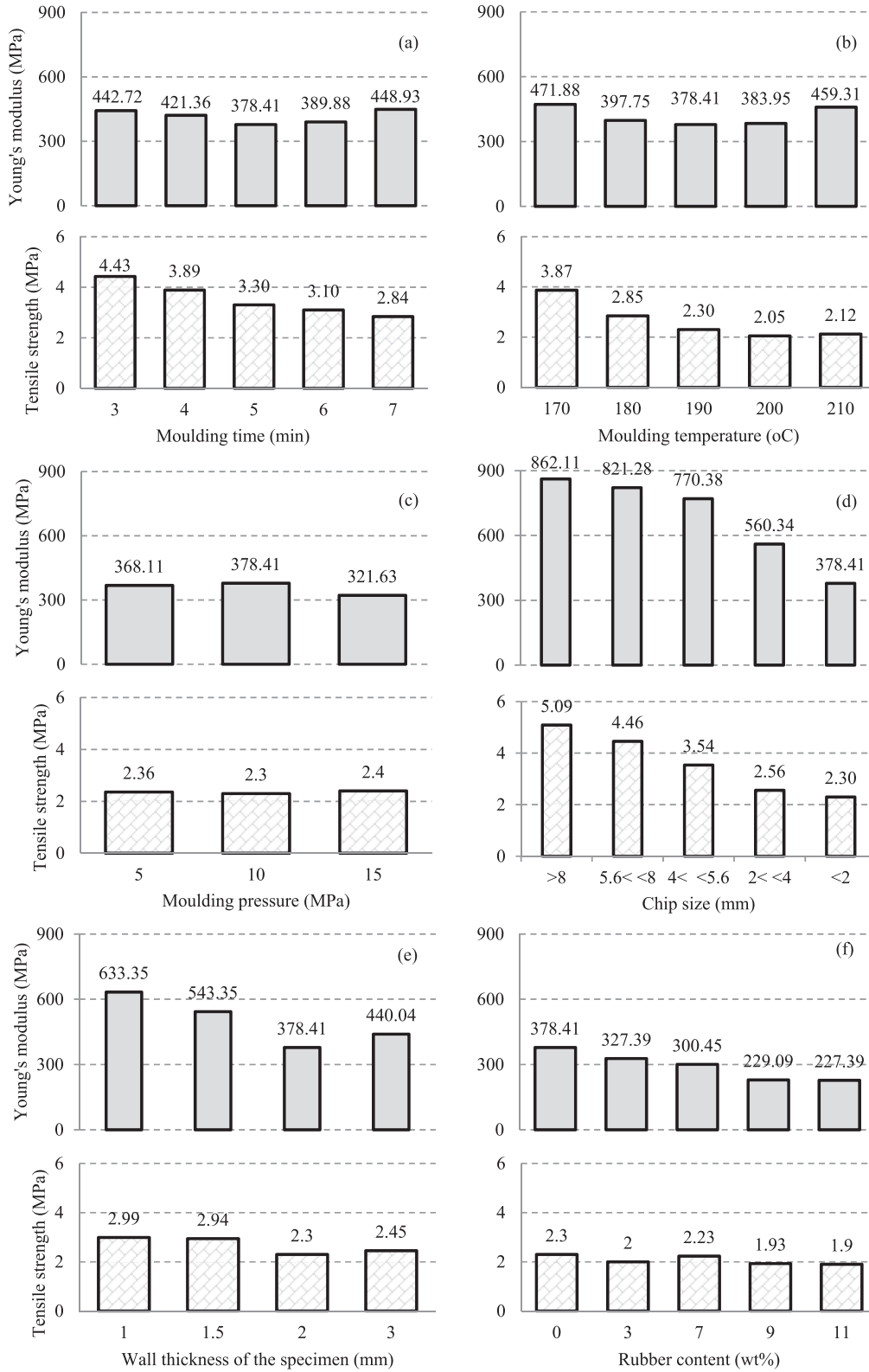


Fig. 4. Variation of Young's modulus and tensile strength of MEP54 with: (a) Moulding time, (b) Moulding temperature, (c) Moulding pressure, (d) Chip size, (e) Wall thickness of the specimen, (f) Rubber content by wt%.



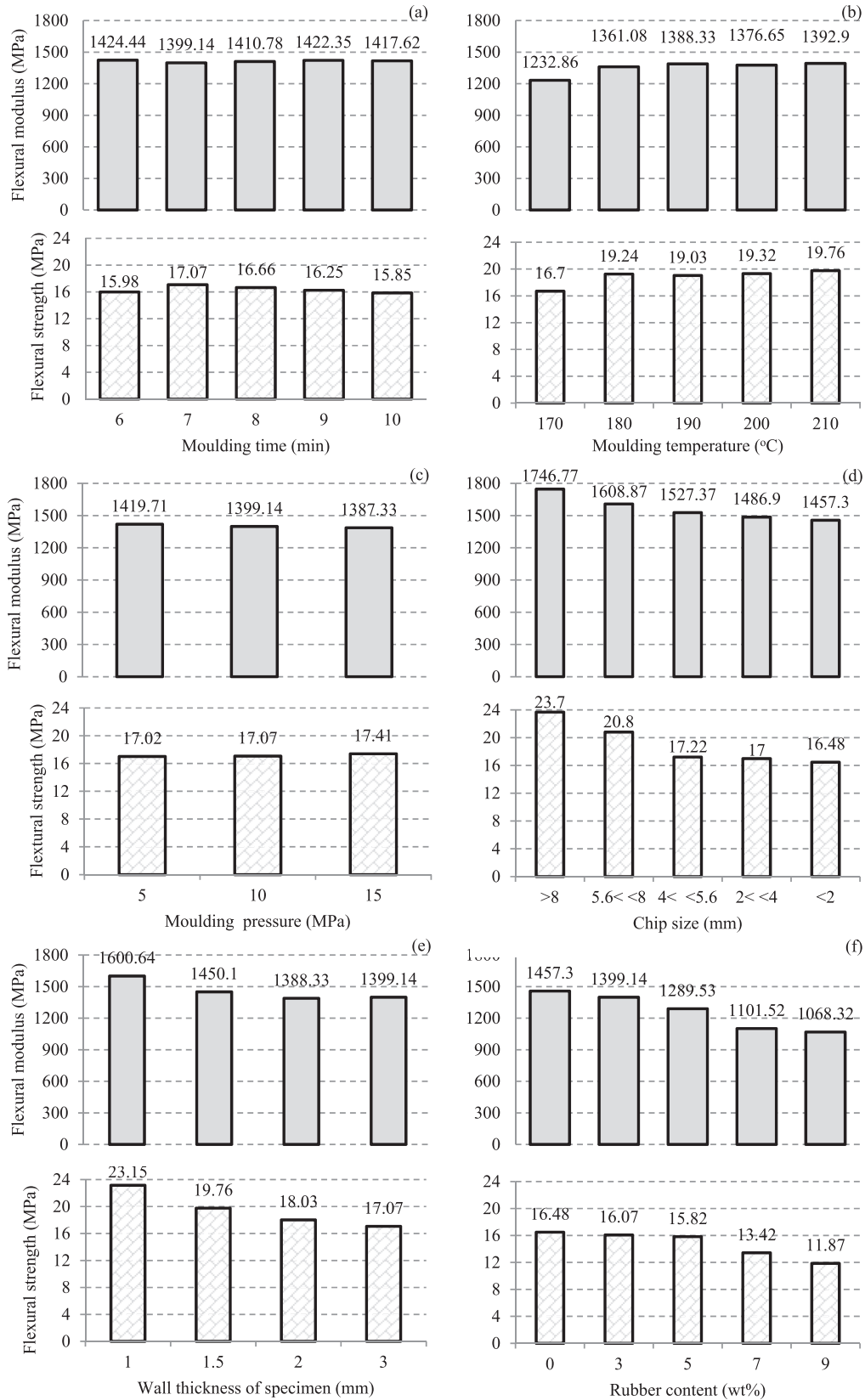


Fig. 5. Variation of flexural modulus and flexural strength of MEP54 with: (a) Moulding time, (b) Moulding temperature, (c) Moulding pressure, (d) Chip size, (e) Wall thickness of the specimen, (f) Rubber content by wt%.

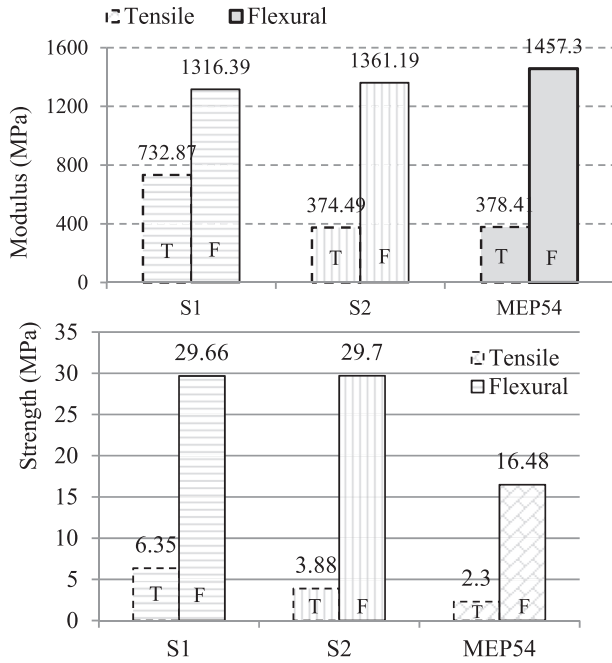


Fig. 6. Modulus and strength of different materials (S1, S2 and MEP54) – Chip size < 2 mm.

clearly influenced by the moulding time: the tensile strength decreases with increasing moulding time, while the Young's modulus decreases and then recovers. A similar pattern can be seen with the moulding temperature, Fig. 4(b). These results suggest that the best operating temperature for compression

moulding of MEP54 derived material would be at 170 °C, which would also be favourable in terms of energy savings as less heat is needed. The moulding pressure has only a small influence on tensile properties, Fig. 4(c), and based on this information, it can be concluded that a moulding pressure of 5 MPa would be sufficient. The size of the chips also has clear effect on the strength, see Fig. 4(d), where decreasing chip size causes in strength reductions. The Young's modulus and tensile strength were also affected by the wall thickness of specimens, as shown in Fig. 4(e). Rubber is not a common choice for a moulding additive, but rubber content was considered in this study as there was rubber present in the test material samples. As is evident from Fig. 4(f), the rubber content has a significant impact on the modulus of the sample and reduces with increasing rubber content, particularly when the content is higher than 7% (the common rubber content of MEP54 is 5–7%).

### 4.3. Flexural properties of MEP54 derived samples

As can be seen in Fig. 5(a), neither the flexural modulus nor strength was significantly influenced by the moulding time. For Fig. (5) also all the related sub-figures of modulus and strength were plotted in the same scale for the ease of comparison. The property variation was less than 1.5% and 5%, respectively, for the six tested conditions. Fig. 5(b) shows that both the flexural modulus and flexural strength increase with increasing moulding temperature, with the sharpest increase seen in the temperature range from 170 to 180 °C. Moulding pressure had little influence, Fig. 5(c): the flexural modulus showed a decrease of 2.2%, while the strength increased by 2.2%, as moulding pressure changed from 5 to 15 MPa. This again confirms that a moulding pressure of 5 MPa is adequate. Fig. 5(d) shows that both flexural modulus and strength are significantly greater for samples comprised of

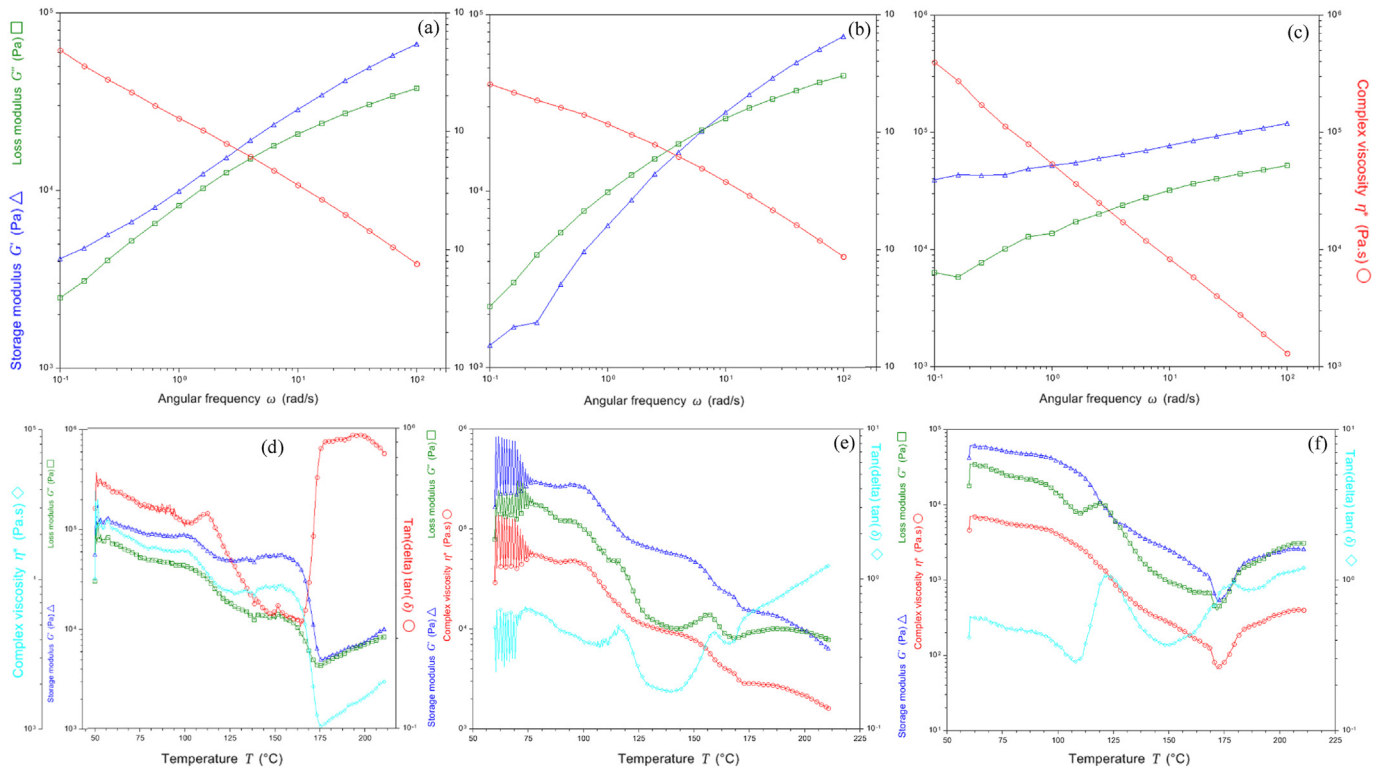


Fig. 7. Top Row: storage, loss modulus and complex viscosity with angular frequency for the three example polymers: (a) MEP54, (b) S1, (c) S2. Bottom Row: storage, loss modulus, tangent ( $\delta$ ), and complex viscosity with temperature for the three example polymers: (d) MEP54, (e) S1, (f) S2.

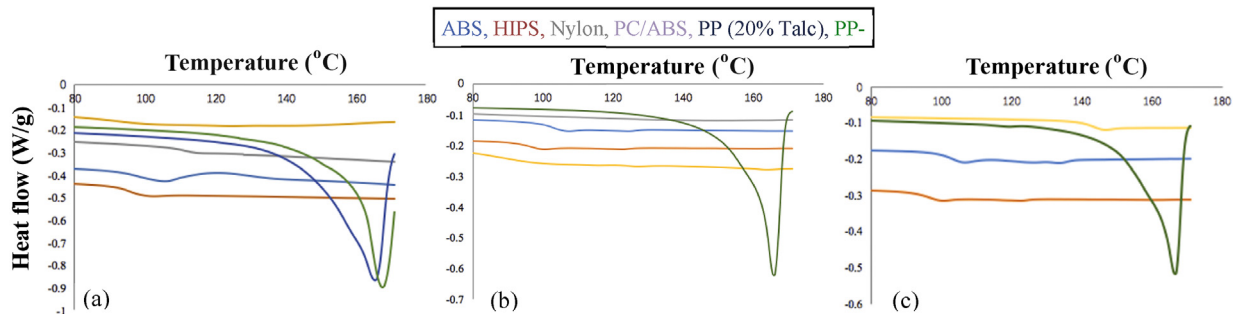


Fig. 8. DSC curves: (a) - MEP54, (b) - S1 and (c) - S2.

Table 2  
T<sub>g</sub> of the constituents of 3 materials from DSC test.

Sample No	ABS	HIPS	Nylon	Blended PC/ABS		PP (with 20% Talc)	PP-C
	T <sub>g</sub> (°C)	T <sub>g</sub> (°C)	T <sub>g</sub> (°C)	T <sub>g</sub> (°C)		T <sub>g</sub> (°C)	T <sub>g</sub> (°C)
1	97.97	97.12	111.21	89.23	/	165.69	168.18
2	103.05	96.10	110.98	84.68	/	166.34	165.67
3	103.45	96.89	/	/	143.50	166.78	165.98
4	102.98	97.19	111.67	87.67	/	165.86	167.98
5	103.20	97.00	/	87.82	145.98	166.12	166.90
<b>Average T<sub>g</sub> (°C)</b>	102.13	96.86	111.28	87.35	144.74	165.69	166.94
<b>(SD)</b>	(±2.33)	(±0.44)	(±0.35)	(±1.91)	(±1.75)	(±0.43)	(±1.14)

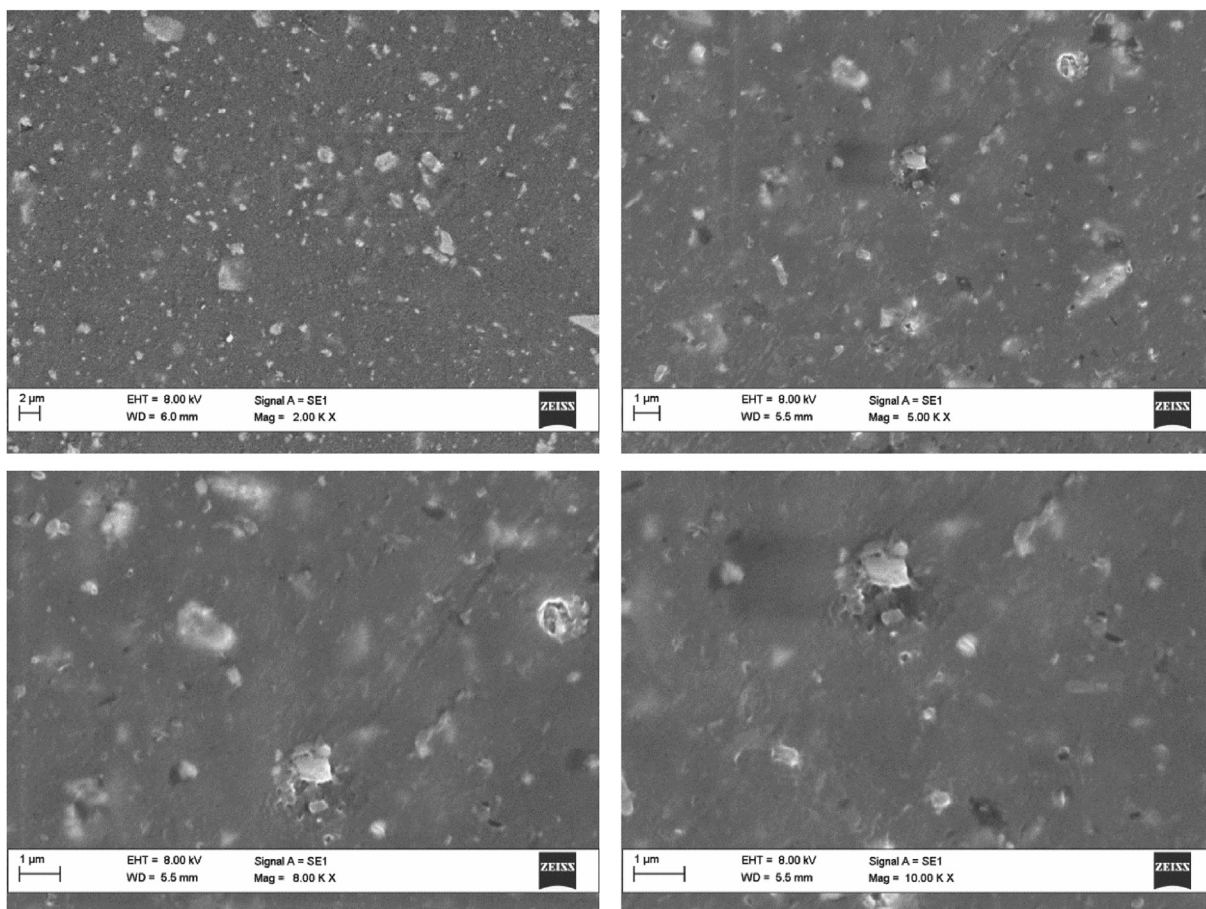


Fig. 9. SEM images of samples made from big chips (>8 mm).

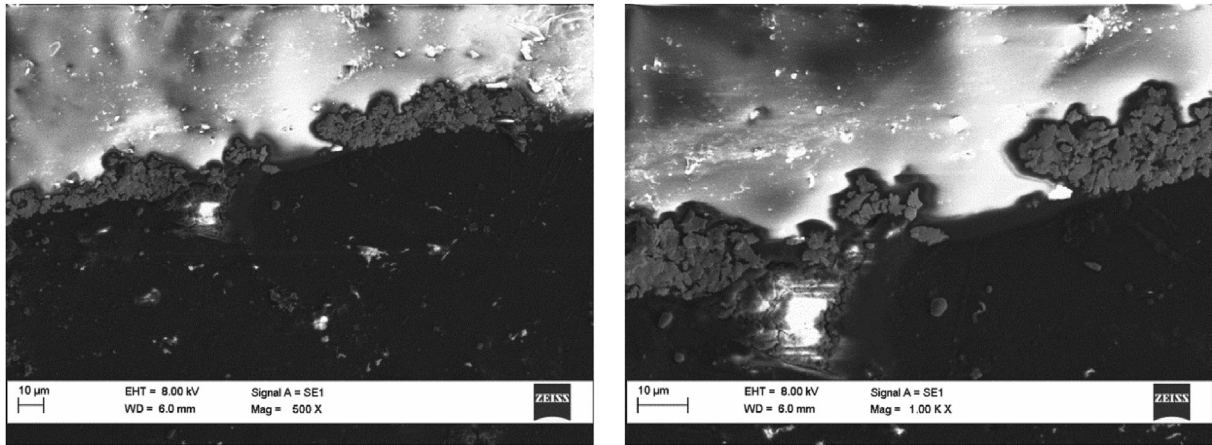


Fig. 10. SEM images of samples made from granules (<2 mm).

larger chips, with a variation range of 16.57% and 30.46% respectively with chip size ranging from 8 to 2 mm. Both flexural modulus and strength are significantly greater for smaller sample thicknesses, Fig. 5(e), with a variation of 14.4% for the flexural modulus, and of 35.62% for the strength, for sample thickness varying between 1 and 3 mm. Fig. 5(f) shows that both flexural modulus and strength decrease as the rubber content increases from 0 to 9%, with the sharpest reduction occurring for rubber content between 5 and 7%.

#### 4.4. Comparisons of properties of S1, S2 & MEP54

For this comparison, each of the samples of S1, S2 and MEP54 were moulded at the same conditions: moulding temperature – 190 °C; moulding time – 5 min; moulding pressure – 10 MPa; wall thickness – 2 mm; 0% rubber content; and with the same chip sizes. The results are presented in Fig. 6 and show that sample S1 has a tensile modulus of approximately double that of both S2 and MEP54. On Fig. 6, letters 'T' and 'F' stand for Tensile and Flexural,

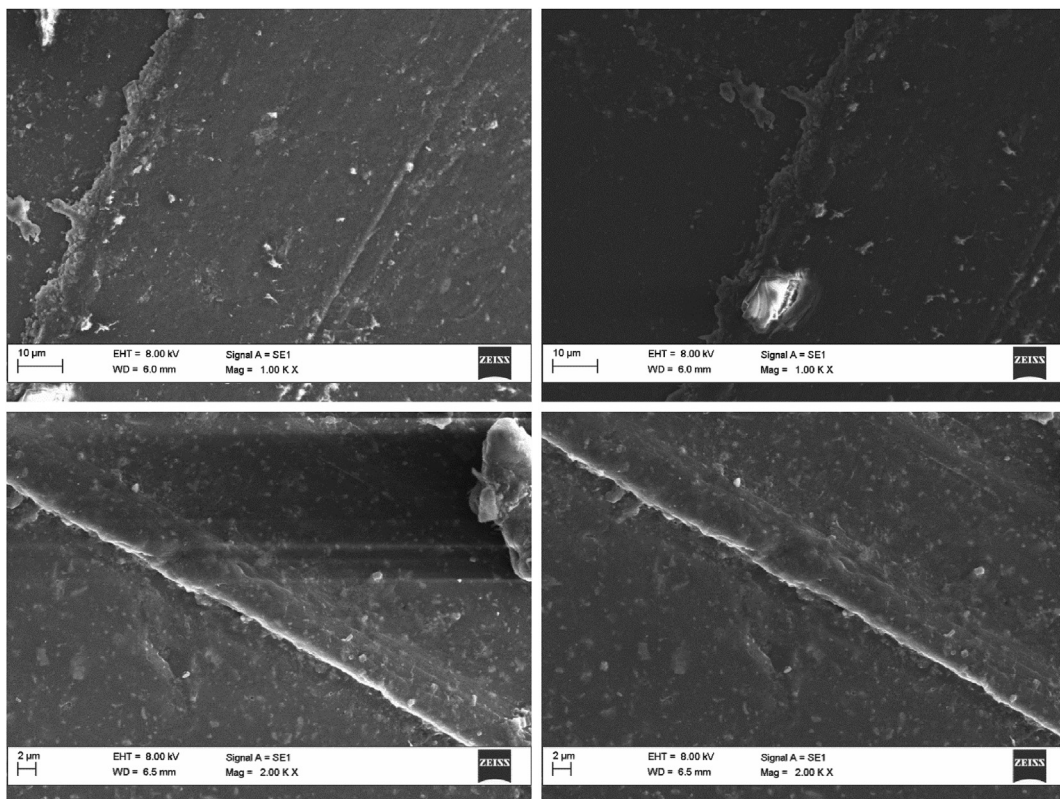


Fig. 11. SEM micrographs showing mechanical bonds on the surface of the samples.

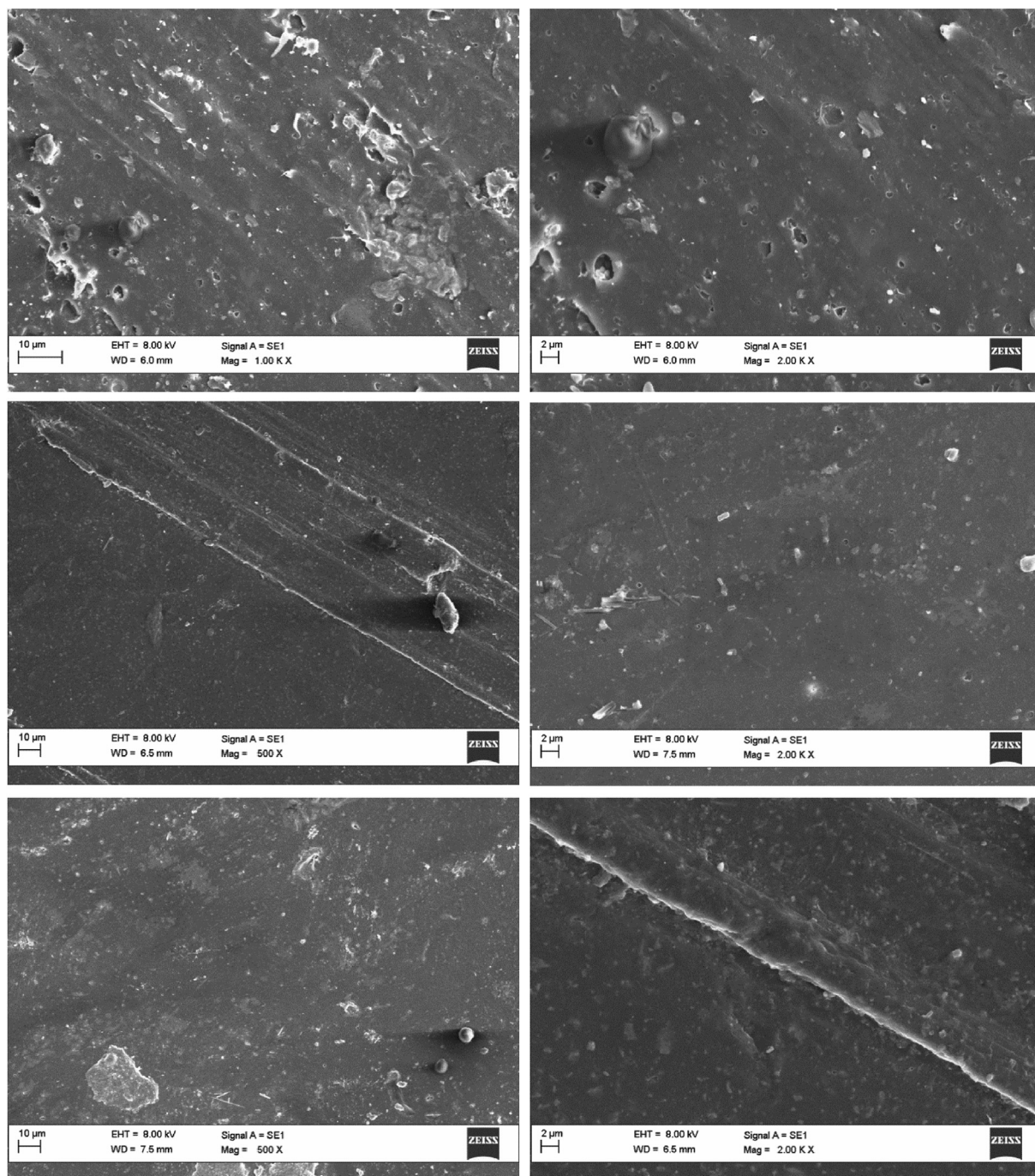


Fig. 12. SEM images of the surfaces of the samples moulded at 170 °C (top), 200 °C (middle) and 210 °C (bottom).

respectively. The same trend is present for the tensile strength comparing S1 with S2, but the tensile strength of MEP54 is somewhat less. This pattern is not repeated for the flexural modulus. Here the flexural moduli of the three materials are similar, but with that of MEP54 being noticeably greater. The flexural strengths of S1 and S2 are approximately equal, while the flexural strength of MEP54 is about half of that value.

#### 4.5. Rheological properties

Typically, the rheological properties of polymers are quite complex in nature and depend on shear and viscoelasticity. In this

study, two different types of rheological tests were performed. Fig. 7(a) shows the effect of shear loading frequency on the viscosity and storage/loss modulus of MEP54. The complex viscosity decreases with increasing frequency, exhibiting the nature of a pseudo-plastic fluid, *i.e.*, shear thinning. The results of the temperature ramp test for MEP54 are shown in Fig. 7(d). This represents the variation of viscosity, modulus and tangent  $\delta$  with the temperature. Two viscosity reduction points are clearly visible at about 100 °C and 165 °C. There is also a significant reduction in  $\delta$  at about 110 °C. Fig. 7(b–c) and (e–f) show the results obtained for S1 and S2, for complex viscosity, shear storage modulus ( $G'$ ) and shear loss modulus ( $G''$ ). The fluctuations shown in Fig. 7(e) between 50

and 75 °C should be related to some initial tuning of the test set-up and hence can be ignored and also it is out of the temperature range of interest for this study.

#### 4.6. Thermal properties

Fig. 8 shows the DSC results for the three material types, MEP54, S1 and S2. All of these tests were carried after cleaning the chips to remove any possible impurities. In each DSC test, at least five chips from each constituent polymer (i.e. 6 constituent polymers for each test) of the same batch of recycle were used. From these tests, the glass transition temperatures,  $T_g$ , were observed, and these are given in Table 2. This table also shows the mean and standard deviation values for each  $T_g$ .

#### 4.7. Image analysis

Scanning Electron Microscopy (SEM) was used to analyse the nature of the material and their processing behaviour (the figures presented here are for MEP54). Fig. 9 shows SEM micrographs of compression moulded samples made from big chips (>8 mm), with magnification levels from 2.00 to 10.00 kX. SEM micrographs of samples made from granules (<2 mm) are shown in Fig. 10, with magnification levels of between 100× and 1000×. Here the boundaries between two neighbouring different constituents are clearly visible. In Fig. 11, the images reveal the physical nature of the bonding formed between different polymer granules of recycled polymers. Fig. 12 shows that for samples moulded at low temperature there are more nodules and craters on the surface than for those moulded at high temperature.

### 5. Discussion

#### 5.1. Discussion concerning the test results for manufacturing process parameters

The tensile and flexural testing of the MEP54 samples manufactured with different process parameters raised some unexpected results. In both the tensile and flexural tests, it was found that samples made from larger chips had superior properties to those made from small chips. This opposes the assumption that the smaller chip size would lead to the creation of more connecting bonds within the samples, which might be expected to make mechanical properties better. This result implies that although smaller chip size does bring more connecting bonds to the sample, these bonds themselves are weaker than original bonds that were already in the chips, so that the bonds between chips are, in effect, defects in the samples. This assumption is confirmed by the observation that almost every specimen failed at the interface between chips: as the individual chips differ in colour, it is easy to recognize failure at the interface by naked eye. As defects lead to stress concentrations, so both Young's modulus and tensile strength decrease sharply for the smaller chip size and larger number of defects. These findings support the case that the bonds generated in the material during the original forming stage are stronger than the newly formed connecting bonds and goes some way to explain the observation that material properties deteriorate through the life cycle [14–16].

A further observation that runs counter to intuitive expectation is that the material strength increases with reducing wall thickness. As it would seem that the amount of plastic strain energy absorbed by materials during moulding, per unit weight or volume, in thin samples is greater than that in thicker ones, there may be an explanation as to increased strength arising from the larger area of overlapping bonds resulting from greater areal spreading of the

chips during forming. This effect would be expected to be more prevalent with larger chips.

Although rubber would not normally be a desirable additive in a first-use polymer, its presence in MEP54 in volume fractions of 5–7% suggests that it is probably difficult to avoid in recycled materials. As the rubber particles are usually randomly scattered within the samples, the location and size of these particles, and how they cluster, would be expected to affect the sample strength. The test results obtained here indicate the extent of the mechanical property reduction, which is an important consideration and a matter for trade-off with the cost of rubber removal.

With regards to the moulding temperature, there was a significant difference in the trend of property performance between tensile and flexural properties. The tensile properties, particularly the strength, were greatest for the lowest moulding temperature of 170 °C used during experiments, whereas for the flexural properties the same moulding temperature gave the worst flexural modulus and strength, and properties for 180 °C and above were broadly similar. In part this might be explained by the chip flattening which takes place during moulding, and through consideration of the fact that in tension, the same stress is applied across the entire specimen cross-section in the gauge section, whereas for flexural testing, the stress is greatest at the specimen surface. A further observation is that the typical melting temperature of PP is close to 170 °C. According to the composition tests, MEP54 contains of around 29% of PP by weight, hence a considerable portion of the material sample would remain only partially molten at 170 °C, and thus resistant to effective moulding. This fact was also confirmed by the DSC and the microscopy results, where it is shown that high moulding temperatures improve the bonding between granules. In this study, all materials were mixed by mechanical stirring and this may not be the ideal method for mixing.

#### 5.2. Differences between the three recycle materials: MEP54, S1 and S2

For both S1 and S2 materials, the strengths are similar, and yet significantly higher than those for MEP54. The similarity of S1 and S2 was expected as a consequence of their having a similar composition of polymer types and in similar ratios. These two recyclates have a lower impurities and PVC content compared with the MEP54. Nevertheless, it should be remembered that the results were obtained with different chip sizes, properties are not uniform, and that for all the results collected, there is no convincing overall trend. This poor predictability of behaviour is, of course, one of the main issues with the use of recycled materials in practical applications [14,15].

#### 5.3. Observations in regard to rheological and thermal properties

The rheological testing carried out on the MEP material, showed it to have shear-thinning properties. This can be explained as increasing shear force reduces the molecular chain entanglement for all the constituents contained in MEP54, which leads to a greater fluidity of the blend. The two viscosity reduction points at about 100 °C and 165 °C can be attributed to the melting of ABS, having a  $T_g \approx 102$  °C, and PP, for which the melting point is  $T_m \approx 166$  °C. The reduction in  $\tan \delta$  at about 110 °C suggests a lower gain in the viscoelastic behaviour, compared with the gain in the storage modulus. For temperatures higher than 160 °C, the values of the moduli coincide because  $\tan \delta \approx 1$ .

For S1 and S2, the results obtained showed that the shear storage modulus,  $G'$ , is higher than  $G''$ , the shear loss modulus, in the low frequency region. This was reflected in the  $\tan \delta$  curves, where  $\tan \delta$  is the ratio of loss to storage modulus. It should be

noted that  $G'$  and  $G''$  are related to the elastic and viscous responses of a material, respectively, and that a material with  $\tan \delta > 1$  will exhibit more damping than the materials with  $\tan \delta < 1$ . The temperature ramp test for S1 was different to that for MEP54 and S2 as it kept reducing continuously with increasing temperature.

Although the chips tested may not be recycled from one product, they showed little variability in the DSC tests, with the exception that there were two different  $T_g$  values for PC/ABS at around 87 °C and 145 °C. PC/ABS is a blend of PC and ABS, and since the  $T_g$  of PC is known to be about 145 °C, then if the two polymer types are not mingled well in the blend, a  $T_g$  of around 145 °C for a PC/ABS blend is to be expected.

The amorphous polymers (ABS, HIPS, Nylon, and PC/ABS) show  $T_g$  values that are lower than 100 °C. The crystalline polymers, PP with 20% Talc, and PP-C, have  $T_g$  values which are significantly higher, and therefore these materials cannot be melted at temperatures below 170 °C. This result explains the sharp rise observed in flexural properties from 170 to 180 °C, as these materials would not be fully molten.

#### 5.4. Areas for further investigation

Adding different constituents of recycled polymers one by one to the mixture should help to discover the effect of different polymers on the properties of final material mixture. The results from a DSC test can only show a  $T_g$  of single kind of polymer/constituent, not the  $T_g$  of the blend made, because the size required for DSC is too small. Thus, the  $T_g$  of the blend should be obtained. Besides, it is better to investigate how some compatibilizers can improve the properties of these types of blends. Also, more investigation should be carried out to understand more on how to form blends of materials to achieve desired properties. Once different types of materials were separated through recycling, different materials can be mixed based on their compatibility and the desired properties of the target application. Moreover, material separation is also an important area which requires further improvements. Then, some impurities such as rubber can be separated to have clean recycled materials, and so to achieve good performance.

Since recycled material is frequently used for packaging and insulation material, an in-depth investigation of their impact strength and insulation properties would be informative. Based on the observed tensile and flexural properties, the impact strength performance of these materials might be expected to be compromised compared with virgin polymers, but this deserves to be tested.

#### 5.5. Implications of testing and of future trends in technology

The re-manufacturing trials described and discussed above have demonstrated a process in which recyclable material is extracted from a batch of recyclate that included a proportion of unusable material. Various manufacturing process parameters were varied, and samples of the processed material were then tested. Within the constraints of this set of trials, some trends in the performance can be identified: it is important to note that while some significantly more advanced and capable re-manufacturing process could produce samples that would have radically better properties by improving the quality of the bonding between chips, the present trial actually presents a good paradigm of a real re-manufacturing factory process. We should recognize a distinction between a recycling process that takes low grade scrap and produces useful

but mundane products, one in which well sorted scrap is recycled for use in highly engineered safety critical components, and a range of product types between those two extremes.

The process that has been explored here has identified areas in which the value of the recyclate “raw” material could have been improved. Better pre-treatment to remove unusable materials from the recyclate would add value to the recyclate product and improve the value proposition for the recyclate supplier. The identification and separation of undesirable content, for example PVC or rubber, and the sorting of material by chip size could also add to the value proposition. Furthermore, as the strength and stiffness indications of this trial would seem to indicate that bonding between chips is largely through mechanical adhesion rather than molecular bonding, improvements to the strength of those bonds could be made by changing the form of the chips. Chips that have been remoulded into flattened discs or strings would have greater contact area when subsequently processed and might be expected to superior properties even when processed under basic moulding processes.

## 6. Conclusions

In this study, three different recyclate materials were investigated, and processed to form samples which were then tested. The processing carried out was a paradigm of a basic polymers re-manufacturing factory making new products within the constraints of moulding at a limited range of temperatures and pressures. The aim of the study was to show the process of manufacturing process optimisation, and to use this experience to help identify ways in which the recycling circular economy could be improved. In taking this approach we were able to identify areas in which the value proposition of the recyclate supplier could be increased, firstly, by making some changes and improvements to the sorting of chip material, and secondly by carrying out further processing on chip material to change its shape.

Three different polymers were studied under varying processing conditions. Test samples were prepared by compression moulding, and then subjected to mechanical, rheological, and thermal tests, and SEM analysis. The mechanical testing showed that moulding temperature, sample thickness and chip size were key parameters: samples made from the bigger chips (>8 mm), moulded at 210 °C, and moulded to the smallest thickness (1 mm), exhibited the best mechanical performance. Rubber content and impurities in the material lead to a reduction in the mechanical performance of the samples. Rheological studies indicated that the materials exhibited the characteristics of a pseudo-plastic fluid. A DSC investigation demonstrated that the  $T_g$  of dominant constituents should be considered as an indicator for the choice of set moulding temperature. In support of this, SEM observations showed more nodules and craters on the surface of samples moulded in low set temperature conditions. Overall, the results highlighted the complexity of processing behaviour and the unpredictable nature of the properties of the recycled materials.

In summary, better separation methods should be used to separate materials. To make future recycling more effective, industries making products containing recyclable polymer should be encouraged to mark them so that the polymers can be readily identified. Standards for polymer recycling should be introduced to support the process, to avoid the mixing of waste with deleterious additives with other recyclates. In the longer term, development of technologies for marking materials at the nanoscale should be encouraged.

More importantly, if we want to tackle this waste issue effectively, there is an urgent need of implementing an improved global recycling framework which would be adhered by all the nations across the globe; otherwise there will be no short-cuts to tackle the escalating global waste problem including polymers/plastics. If we do not act together, timely and in more organized ways, there will be garbage patches not only in Pacific Ocean but all over this planet. Failing to do this in timely manner in right scale, will definitely bring catastrophic consequences to the health and wellbeing of the planet Earth.

### Conflicts of interest

The authors declare that there is no conflicts of interest.

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## Nomenclature

ABS: Acrylonitrile Butadiene Styrene  
 ASR: Automotive shredder residues  
 ADC: Authorised dismantling centre  
 BBP: Butyl benzyl phthalate  
 BFR: Brominated flame retardant  
 CFC: Chlorofluorocarbon  
 CRT: Cathode ray tube  
 DBP: Dibutyl phthalate  
 DFD: Design for dismantling  
 DIBP: Diisobutyl phthalate  
 DSC: Differential Scanning Calorimetry  
 EoLV: End-of-life Vehicle  
 EEE: Electrical and Electronic Equipment  
 EPD: Environmental Product Declaration  
 EPR: Extended Producer Responsibility  
 EVB: Electric vehicle batteries  
 FDM: Fused Deposition Modelling  
 FFF: fused filament fabrication  
 FML: Fibre metal laminates  
 FTIR: Fourier Transform InfraRed  
 GC: Gas Chromatography  
 GDP: Gross Domestic Product  
 GFRP: glass fibre-reinforced plastic  
 GHP: Guarded Hot Plate  
 GQD: Graphene Quantum Dot  
 HDT: Heat Distortion Temperature  
 HHW: Household waste  
 HIPS/PPE: Polyphenylene Ether blended with High Impact Polystyrene  
 HTTP: human toxicity potential  
 ICP-MS: Inductively coupled plasma mass spectrometry  
 IW: Industrial waste  
 LCA: life cycle analysis  
 LCI: life cycle inventory  
 LIBS: Laser induced breakdown spectroscopy  
 MANOVA: Multivariate analysis of variance  
 MFA: material flow analysis  
 MFI: Melt Flow Index  
 MINLP: mixed integer non-linear programming  
 MOD: Ministry of defence  
 MSW: municipal solid waste  
 NIR: Near InfraRed  
 OEM: original equipment manufacturer  
 Opex: operational expenditure  
 PBB: Polybrominated biphenyls  
 PBM: Population balance model  
 PC: Polycarbonate  
 PC: personal computer  
 PCB: polychlorinated biphenyl  
 PCB: printed circuit board  
 PEF: Polyethylene furanoate  
 PET: Polyethylene terephthalate  
 PGM: Precious group metal  
 PRO: producer responsibility organization  
 PU: Polyurethane  
 PV: Photovoltaic  
 QMIP: Quadratic mixed integer programming  
 REE: Rare earth element  
 RPA: Recycled Plastic Aggregate  
 RoHS: Restriction on hazardous substances  
 RRR: reuse recycling recovery  
 SEM: Scanning Electron Microscopy  
 SIC: Standard Industry Classification  
 SSD: solid state disks  
 SSSP: Sliding spark spectroscopy  
 TCLP: toxicity characteristic leaching procedure  
 TGA: Thermogravimetric Analysis  
 TIS: Technological innovation system  
 TIS: technological innovation systems  
 WEEE: Waste from Electric and Electronic Equipment  
 XRF: x-ray fluorescence  
 XRT: X-ray transmission