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Research article

How introduction of deposit-refund system (DRS) changes recycling of non-drinking bottle PET wastes

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Abstract. Under the era of circular economy, the deposit-refund system (DRS) for *e.g.* polyethylene terephthalate (PET) is thought to be a good choice to achieve a more structured plastic recycling. The present research has the aim to make a comprehensive description and a practical guideline in order to evaluate how collection and separation system influence the quality and efficiency of mechanical recycling of PET. The DRS has been symbolized by manually collected bottles with (BCL) and without (B) caps and labels. Samples have been given from the selective income (SI) and the sorting residue (SR) of a manual selective waste sorting plant and PET fraction of refuse derived fuel (RDF). Based on preliminary qualification results such as melt flow indices (MFI), PET bottles are worth selecting into the main colours like water clear, blue, and all the others together, referred to as PET-A, PET-B, and PET-D fractions of the sorting plant. The SR seemed to be a beneficial raw material for PET recycling as both mechanical and rheological properties were proper enough. PET separated from the Mechanical Biological Treatment (MBT) plant as RDF showed the worst processing and mechanical properties, but both can be improved with deeper precleaning. X-ray tomography (CT) scans have shown a correlation between the source of waste and the gas void structure which influence the macroscopic mechanical properties.

Keywords: *recycling, mechanical properties, rheology, computed tomography, deposit-refund system*

1. Introduction

1.1. Regulation of waste management in Hungary

Among environmentally harmful wastes plastic is one of the most challenging ones on a world scale [1–7]. Packaging dominates the usage of plastics accounting for nearly 40% of plastic consumption [8–10]. The European Union (EU) continuously makes huge efforts both to reduce the amount of plastic packaging waste and to recycle post-consumer waste. One of the acts was the legislative framework to control plastic packaging waste. The latest big step of the European Commission toward a circular economy was acceptance of the Plastic Strategy in 2018 [11]. Along with the European Union Action Plan for the circular economy which allocates the entire action plan of increasing the quality of plastics recycling the 'Plastics Waste' EU strategy forces to make all plastic packaging recyclable by 2030 [11, 12]. For achieving goals of the European Union Action Plan for a circular economy appropriate institutions, systems, and incentives appear to be crucial in order to define and build a holistic approach to increase the quality of plastics recycling [11, 13, 14]. Additionally, secondary raw materials were stated by the European Green Deal to be mandatory for the market from 2019 [15]. According to the Single-Use Plastic (SUP) directive, polyethylene terephthalate

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(PET) drinking bottles will be demanded 25% recycled plastic from 2025 and 30% from 2030 [16]. The European Union boosts all the countries to create and introduce innovative models for plastic production and consumption to achieve circularity in a shorter time frame. The deposit-refund system (DRS) for wastes such as plastics and glass is only one of the models. Furthermore, the European Parliament is encouraged to establish the national DRS schemes destined for the unionization of models under EU law for the EU single market [17]. Hungary as a member of the EU harmonized the waste regulation in the country. The Hungarian Parliament accepted the amendment of the Waste Law [18] which contributed to the domestic introduction of the DRS in the next years. In accordance with the aforementioned the National Waste Management System is currently under major reconstruction with respect to fit requirements for a more sustainable circular economy [19]. In Hungary, the DRS system is going to be extended for PET drinking bottles either besides aluminium beverage can and glass bottles. In the return system planned in Hungary, the disposed drinking water and beverage bottles are collected in commercial units as cleanly as possible. It is important to note, that the DRS is destined for the compilation of drinking bottles, meanwhile, other PET products than bottles such as detergent, soap, and rinsing flasks among others will remain in the selective waste or regrettably in the municipal solid waste [20].

1.2. Waste management in the North-Balaton region

Waste management in Hungary is under a huge transformation as MOL Plc. won the Hungarian state concession tender for municipal waste management services. The concession agreement is for 35 years with a commencement date of July 1, 2023. The North-Balaton Waste Management Nonprofit Ltd. (NBWN Ltd., in Hungarian: Észak-Balatoni Hulladékgazdálkodási Nonprofit Kft.) is one of the 28 Hungarian regional waste management public service companies. NBWN Ltd. is responsible for waste management in more than 158 settlements in Veszprém country consisting of more than 126 000 households. NBWN Ltd. contributes to six other waste management public services accounting for local waste collection, separation, and temporary storage. Avar Ajka Nonprofit Ltd. is in charge of waste management in 48 towns and villages. The enterprise is competent in collecting household waste from more than 20 000 sources and as a public service collects both the selectively disposed household wastes and the selective waste yards on the streets. Households are required to collect only the packaging wastes such as paper, cardboard, plastic, and metal in a 'yellow or blue' bin. The glass packaging wastes are compiled in containers for glasses on the streets. Avar Ajka Ltd. operates a manual sorting site in Ajka with a capacity of 7630 tonnes/year for packaging wastes such as plastic, paper, cardboard, glass, metal and also the mix of them. The incoming waste is weighed and then placed into the manual sorting conveyor and separated in the 12-position manual sorting line (Figure 1).

The recyclable fractions are paper and cardboard, aluminium, PET, high-density polyethylene (HDPE) and polypropylene (PP), foil furthermore magnetizable metals. PET wastes are separated into three fractions, namely coloured PET as PET-D, water clear PET as PET-A, and bluish PET as PET-B. The sorted PETs are only drinking bottles, other PET wastes are ended in sorting residue. The highest quantity in weight in the income household selective waste is paper, cardboard, newspaper fraction (34%), 20% is glass waste, 8% is PET (all types), 6% is foil and 26% is sorting residue. The other sorted fractions count at most 3%. Values were calculated for the 2021 base year, however, the last five years gave the same rates [21].

Sorting residues are grouped and transported to another resides of the NBWN Ltd. into Királyszentistván. In Királyszentistván a waste separation plant with 120 000 tonnes/year household solid waste capacity is working to enrich the wastes energetically utilizable, for instance, refuse derived fuel (RDF). Mechanical biological treatment (MBT) is carried out on the non-hazardous waste inside the plant. The first step of the mechanical processing is the weighing and shredding of the waste into 250–350 mm particle size. Afterwards, the magnetizable metals are separated. The next step is a rotary sieve wherein the biological waste under 80 mm particle size is removed for further treatment. Residue over 80 mm is usually divided into two parts by an air-separation unit. The heavy fraction containing mostly stones and bricks goes to the landfill directly. The light and middle fractions are driven into an eddy current sorter to recover the aluminium wastes. The next process polyvinyl chloride (PVC) removal from the light and medium PET

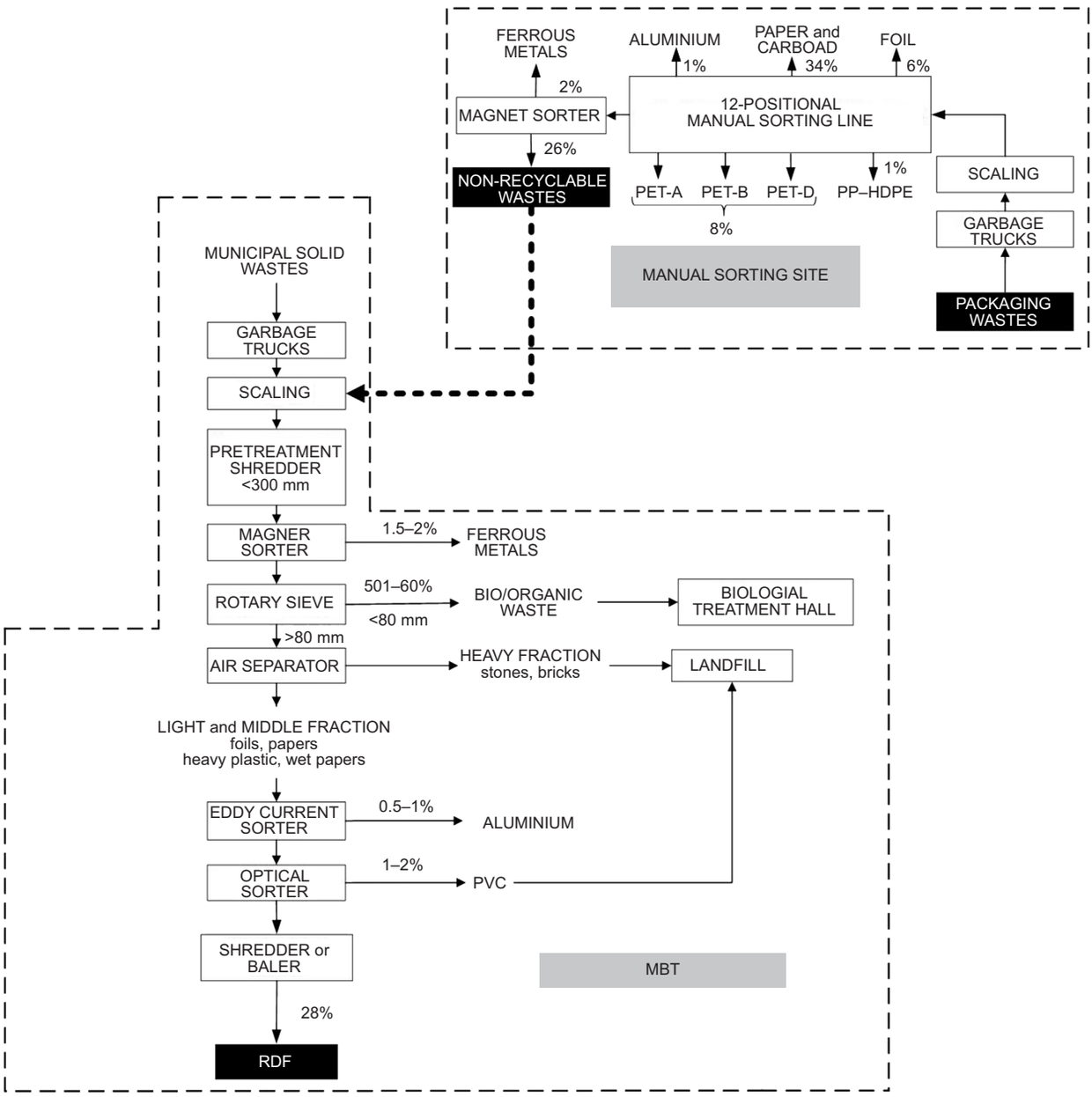

Figure 1. The waste streams through the manual sorting site (Ajka) and MBT plant (Királyszentistván), the source of PET samples for the research was highlighted in black.

fractions remained by an optical sorting machine. The sorted PVC is deposed into the landfill either. The halogen-free residue for RDF is balled or shredded and balled. 92% of the total plant income is directed to the MBT site whereby 77% of it is going through the separation line. In the end, 28% of RDF is generated from the separated household waste. Values were also calculated for the 2021 base year, however, the last five years gave the same rates either [21].

1.3. PET recycling from different waste sources

The composition of plastic mixtures originated from an open-loop recycling process as described in Figure 1 could be rather complex; therefore, chemical incompatibility and inhomogeneity of various plastics had to be taken into account. The drawbacks of recycling of the plastic mixtures can be successfully managed by the proper separation as one of the crucial steps in plastic recycling. Moreover, secondary raw materials gain higher economic values when the concentration of contaminants, *e.g.* other types of plastics, are kept at lower levels [22]. However, contaminants are not the only cause for the reduction in the properties of the recycled plastic product resulting in lower economic values. The other crucial step is the reprocessing. It is well-known, that reprocessing of plastic waste with various types of

equipment of melt processing techniques such as extrusion and injection moulding can lead to the deterioration of product properties because of molecular weight reduction and degradation effects [23]. PET has become one of the most popular packaging materials and the main problem is the large volume of waste produced within an extremely short time. Huge efforts have already been made towards recycling PET, especially recycling into higher value-added products since landfilling is not an option for disposal anymore [24]. Melt extrusion is one of the general processes for the mechanical recycling of plastics resulting in PET pellets without significant changes in the chemical structure of the polymer chain. Within the EU, PET bottles are recycled by over 50%, becoming the most recyclable product up-to-date [7]. PET-based products other than bottles, such as films and fibres, do not achieve such high recycling rates as PET bottles. Most of them end in the RDF fraction of waste which is the most difficult to recycle into higher value-added products. Because important mechanical properties usually reduce due to degradation during reprocessing designation mechanical recycling of PET as down-cycling often occurs [25]. PET upcycling applications started with Coca-Cola to reuse beverage bottles [26]. The recycling industry started with mechanical PET transformation into value-added products such as clothes. Different end products such as containers, films, nonwoven fabrics, and other plastics can also be produced from waste PET [26]. Plenty of researchers applied PET waste as a replacement for aggregate in cement concrete as a reinforcement in the form of fibers or even as raw material for the production of activated carbon [24, 26]. Numerous publications describe opportunities for the synthesis of polyester resin on a PET waste basis either [24]. There is increased interest in the application of recycled plastic materials in pavement and PET was a widely used candidate in many studies as a partial replacement for the aggregate [27–33]. For example, the highest resistance to permanent deformation was obtained by using 20% of PET as a partial replacement moreover, PET as a modifier improved fatigue life either [30, 34]. Contaminants in PET waste cause serious problems, *e.g.* PVC decomposes and hydrochloric acid gas releases resulting in the colour-changing and degradation of molecules. Energy consumption and costs are saved through recycling if plastics are reprocessed without separation from other materials or cleaning steps can be minimized [22, 24]. Alsewailem and Alrefaie [23] investigated only the influence of paper contaminants on the mechanical properties and processability of post-consumer PET. One of their most important results was that the toughness of reprocessed post-consumer PET bottles drastically decreased in the presence of paper. The other important result was that the presence of PE as caps or cap rings and labels did not seem to alter its moldability and mechanical properties. Researchers also paid attention to minimizing the number of melt processing steps of reprocessing post-consumer PET in order to avoid deterioration of its mechanical properties [23].

Since choices of consumers can either support or hinder the circularity of plastic reuse, our study basically aims to present the values of PET waste since its most important properties are good enough for higher value-added products, moreover, the research concentrates on wastes of PET products other than bottles either aimed to fill a research gap. Real PET is not a model PET raw material for demonstrating the waste origin, but such a PET waste stream, that comes from everyday life from humans as waste either selectively collected or not. The study focused on processing opportunities and mechanical properties of different real PET wastes containing polyolefins as contaminants. Experiments were carried out with waste PETs derived from the selective collection and different stages of the separation line of plastic sorting plants. Although the material structural analysis used to be applied to determine the properties of composites or structured materials [35], in the case of waste materials the effects of contamination (*e.g.* organic residues), as well as the preparation processes (*e.g.* melt currents, viscosity, pressure condition, temperature), can be followed exactly by X-ray tomography which is a novel and untraditional analysis technique in the field of polymer recycling researches.

2. Experiments

2.1. Raw materials

The ‘selective income’ sample (SI) was a typical ‘yellow/blue bin’ waste which is originated from the input of the Avar Ajka Ltd. plant (Ajka, Hungary). The second sample was the ‘sorting residue’ of the manual sorting plant of Avar Ajka Ltd. signed as SR (Ajka, Hungary). That kind of residue is transported to the waste plant of Királyszentistván. The third

sample was derived from the RDF waste supplied by the sorting plant of Királyszentistván (Hungary). The waste samples were sorted for PET by an optical sorting machine (Black UNISORT P1000 R, Steinert GmbH, Cologne, Germany). The optical sorting machine is working in the near-infrared wavenumber range (1000–1700 cm^{-1}). The machine is unique because its spectrum library is editable with spectra of new or unknown materials of the software either. The equipment is also able to separate the black materials because the separation of the matters is achieved beyond the conveyor. The sensor unit is arranged above the conveyor and the beam source that lights the conveying material stream is also built into that module. The emitted beam is infrared light and the reflected light by the materials is absorbed and measured by the camera system. The PC processes and evaluates the measured spectra and the results are directed to the controller unit. At end of the belt, 80 pieces of pneumatic nozzles are positioned responsible for the separation of the detected objects. The composition of real waste samples for the experiments was identified by the near infrared (NIR) apparatus first. The investigated input sample (SI) of the manual sorting plant consisted of paper and cardboard (22%), glass (11%), and other plastics mainly PP, low-density polyethylene (LDPE), HDPE, polystyrene (PS) (44%), and PET waste (23%). In that sample, the PET wastes were typically drinking bottles. In the sorting residue sample (SR) the initial content was 10% metal, 69% other plastic waste and the PET was 21% wherein the bottles of washing and shower gels were significant. In the RDF fraction 20% was waste glass, 21% resulted in PET and 59% was other plastics. After optical sorting, the SI and SR samples contained 10% impurity which is mostly HDPE as bottle cap and PP label of the bottle.

In the RDF sample, the label appears as contamination nearby the HDPE bottle cap and PP bottle label (Figure 2). As a raw material for experiments two other waste PET samples were sorted by hand from the selective collection of only PET drinking bottles symbolizing the bottles from DRS to be introduced. One sample 'B' (Veszprém, Hungary) was just the PET bottle without any cap or label. The second one 'BCL' (Veszprém, Hungary) was the whole PET bottle with label and cap.

After separation, PET samples were shredded in a shredder (GRS 183A9, DIPRE, San Giorgio delle Pertiche, Italy) and classified by a sieve with a

Figure 2. NIR separated and shredded PET samples from real waste a) SI, b) shredded SI, c) SR, d) shredded SR, e) RDF, f) shredded RDF (SI – selective income; SR – sorting residue; RDF – refuse derived fuel).

diameter of 5 mm (Figure 2). The shredded samples were soaked for a day in tap water. The process was carried out two times. The shredded samples were dried at 60 °C for 16 hours before filament extrusion, the moisture content was below 0.02 wt% for all samples after the procedure.

2.2. Methods

PET samples were prepared in a twin screw laboratory extruder (LabTech LTE 20-44, LabTech Engineering Ltd., Samutprakarn, Thailand). The screw speed was 100 rpm for each sample, and the temperature profile varied between 245 and 265 °C depending on the origin of the waste. Tensile properties were measured under laboratory conditions at room temperature and at 48% of relative humidity with an universal tensile testing machine (INSTRON 5967, Instron, Norwood, Massachusetts, USA) with a cross-head speed of 90 mm/min. The extruded filament samples were conditioned at 60 and at 105 °C for 4 hours before measurements. Filaments with dimensions of 2 mm averaged diameter and 150 mm averaged length were prepared for mechanical tests. Tensile strength and elongation at break results were taken from the average of a minimum of 11 parallel measurements. Rheological properties of the preconditioned granulated filaments were measured with a capillary rheometer (CEAST Smart Rheo 2000, CCSi, Inc., Akron, Ohio, USA) at 255 °C after three minutes of preheating time and shear rate below 10 000 1/s. Five PET waste-based extruded samples were scanned at a resolution of approximately 6 µm using CT scanner (Nikon XT H 225 ST, Nikon Metrology, Leuven, Belgium). The average time per scan was 21 min and 1250 projections images were captured in two frames per projection mode. The samples were scanned a voltage of 165 kV and a current of 65 µA. For the analysis and visualization, the VG Studio Max 2022/2 software pack was applied. These measurement settings were able to show the microporosity as well as the inclusion content of the extruded rods, which gives useful information about the inner structure of the scanned bodies.

All DSC measurements were done on a reaction calorimeter (C80 D, Setaram, Mougins, France). The PET waste samples were weighted on a Mettler Toledo analytical scale (ML204 NewClassic, Columbus, Ohio, USA) and was put into an aluminium foil sample holder (weight differences between the sample and reference holders were ±5.0 mg). A standard stainless-steel cell was used for the measurements without PTFE joints. The sample was first pre-heated to 30 °C using 0.1 °C/min heating rate and was held at that temperature for 2.0 hours. The analysis was done using 0.8 °C/min heating rate from 30 to 300 °C. The observed reproducibility was within 0.5 °C of the specified parameters throughout the measurements. DSC results were taken from the average of three parallel measurements. The results were analysed using the Calisto software (version 1.076).

3. Results and discussion

3.1. Raw material characterization

All the colours of PET bottles from the selective collection at the Department were separately collected for at least a two-year period then ground and different batches were identified with melt flow index (MFI) (Figure 3) values. An MFI tester (CEAST 7024000, CCSi, Inc., Akron, Ohio, USA) was used to determine melt flow indices of the PET samples at 255 °C under a load of 2.16 kg. Waste PET bottles had MFI values between 12 g/10 min and 20 g/10 min dependent on the colour of the bottle (Figure 3e). MFI of batches of water clear (Figure 3a) and green colours (Figure 3b) were close to the average values but occasionally with high standard deviation (SD). MFI of the blue PET bottles was the most unbalanced (Figure 3c). Collecting waste PET bottles in mixed form without any separation based on colours resulted in MFI within a wide range of average values and extremely high standard deviations (Figure 3d).

In accordance with the confidence of MFI values and the colour of PET, the separation of drinking bottles by colour is beneficial from the aspect of recycling. In further experiments, DRS was symbolized by blue colour bottles based on the MFI values that were all the same as of mixed colour bottles.

Shredded samples originated from hand selection and collection systems showed similar behaviour toward parameter change with the purity. Furthermore, because of the corresponding signals became less intensive certain thermal properties could not be identified to an exact point by differential scanning calorimetry (DSC) (Figure 4, Table 1). Glass transition temperature (T_g) of the RDF sample elevated by 12.5 °C compared to the SI. Similarly, for the hand selected samples the T_g increased by 5.4 °C for the BCL sample compared to the B sample. Based on

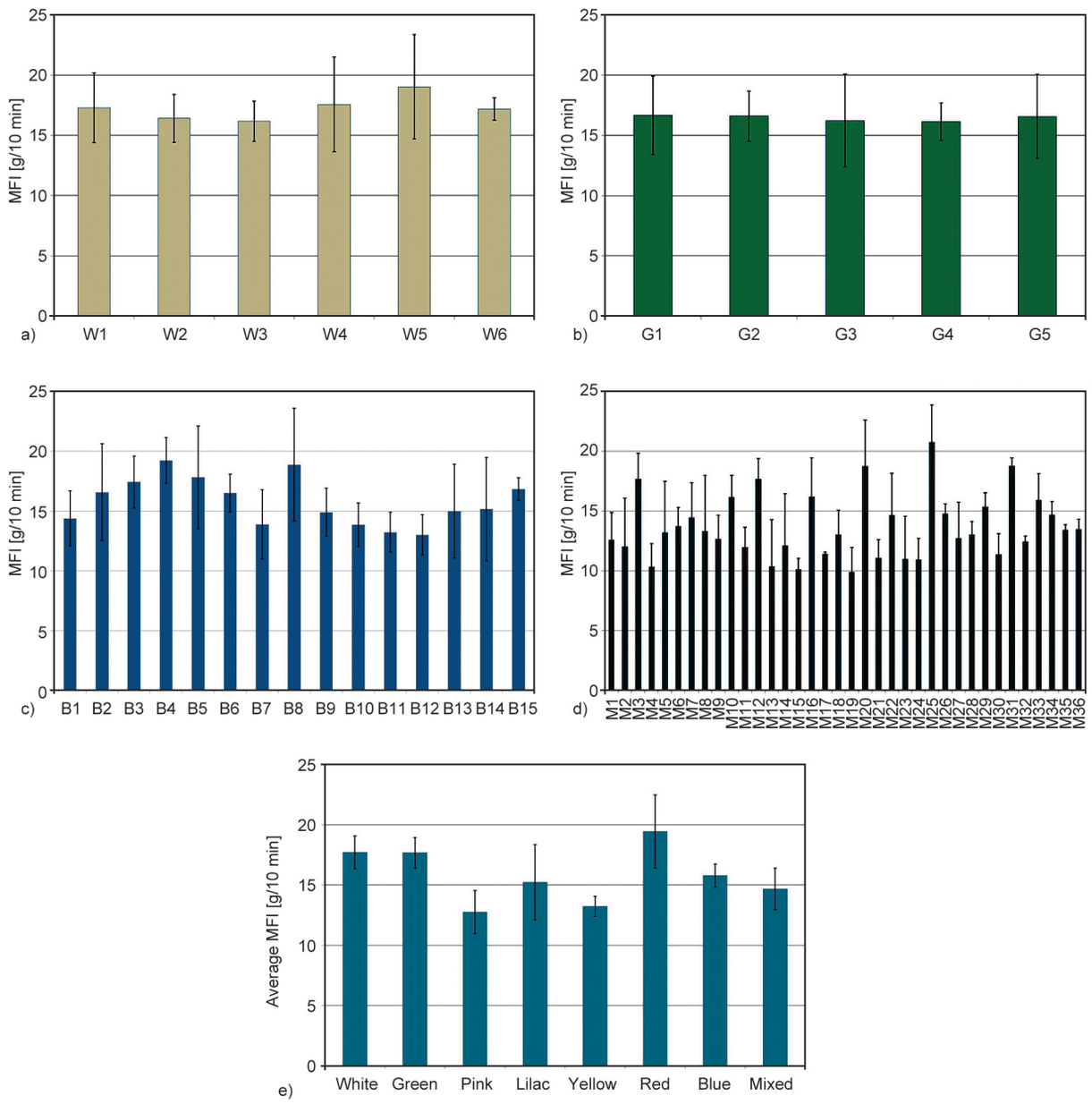

Figure 3. MFI values of waste PETs selected by the colour of the bottles a) water clear (W), b) green (G), c) blue (B), d) mixed (M), e) averaged.

the level of impurities, minor endothermic peaks and an elongated exothermic baseline shifts were observed between 134.9 to 239.6 °C indicating the melting and different phase transitions of the polyolefin content. Both the cold crystallization (T_{cc}) and the melting temperature (T_m) decreased by a small extent within 3.0 °C overall depending on the level of impurity. However, no T_{cc} value appeared in the RDF sample indicating that the impurities mask the phase transition.

Table 1. Main data from the DSC analysis.

Sample	(1) – T_g [°C]	(2) – T_{cc} [°C]	(5) – T_t [°C]
SI	74.9	115.5	256.9
SR	85.5	113.9	256.0
RDF	87.4	N/A*	253.8
B	70.9	117.1	260.3
BCL	76.3	116.3	258.1

*Phase transition masked by impurities

3.2. Melt flow index of the waste PETs

MFI values of waste PET from the different origins at 255 °C and under the load of 2.16 kg were represented in Figure 5. Wastes were identified as ground material. Waste PET from the selective collection of mixed bottles had the lowest MFI (15 g/10 min), indicating the highest molecular weight among the waste PETs. Both selective income (SI) and sorting

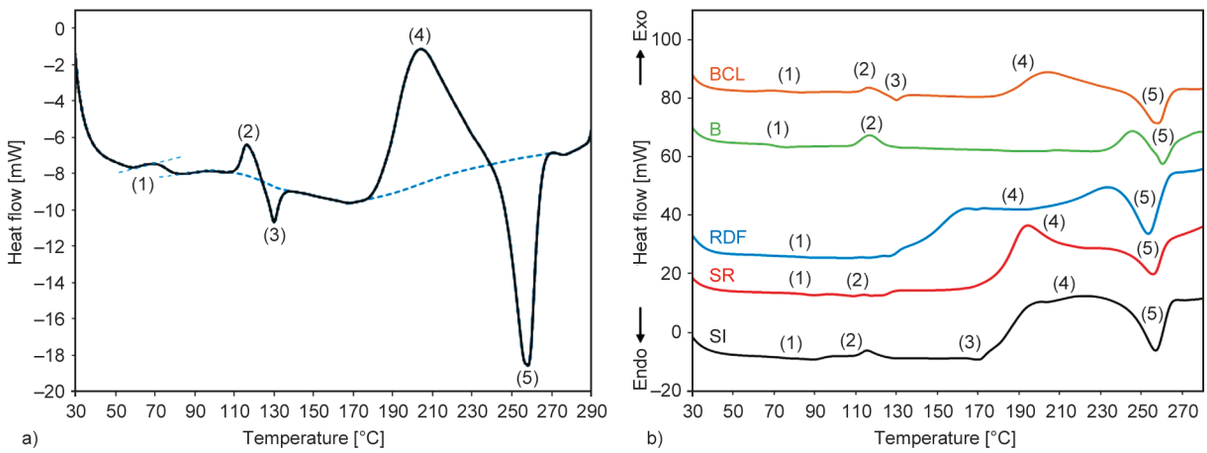

Figure 4. The DSC analysis and evaluation (BCL (a)) of the shredded PET waste samples (b). (1): T_g – glass transition; (2): T_{cc} – cold crystallization; (3): T_{endo} – endotherm peak, polyolefin component melting; (4) T_{exo} – exothermic peak, polyolefin component phase transition and (5): T_m – melting.
(SI – selective income; SR – sorting residue; RDF – refuse derived fuel; B – bottle; BCL – bottle with cap and label).

residue (SR) were the same and had higher (~30 g/10 min) MFI than that of only PET bottles. MFI of SI and SR were 50% higher than waste PET bottles ground with the caps and labels (~20 g/10 min) together, demonstrating that the polyolefin content of those real waste PETs shifted the MFI advantageously to the same direction and extent. Waste PET sorted from the RDF had outstandingly high MFI (~50 g/10 min) also with extremely high standard deviation (~10 g/10 min), as expected. A wide quality range of waste PET was supposed to be the consequences of degradation effects during the lifetime via the lower molecular weight of the molecules.

All kinds of waste PET were reprocessed by extrusion moulding and qualified first with MFI. Results were summarized in Figure 5. No significant differences (2–5 g/10 min) between MFIs of the ground and regranulated materials appeared, providing that extrusion parameters were set advantageously. Reprocessing made sense, at least in a narrower range of SD for the MFI. Therefore, regranulated wastes generally were more homogeneous than the only ground materials. Furthermore, SD was at least the same (~2 g/10 min) for regranulates from the SI and SR as for BCL. The average MFI (~30 g/10 min) of regranulated PET from SI and SR were the same, but 60% higher than the only PET sorted from bottles without cap and label. Therefore, the molecular weight of the PET materials from the selective collection containing not only drinking bottles (SI) was thought to be lower than that of only PET drinking

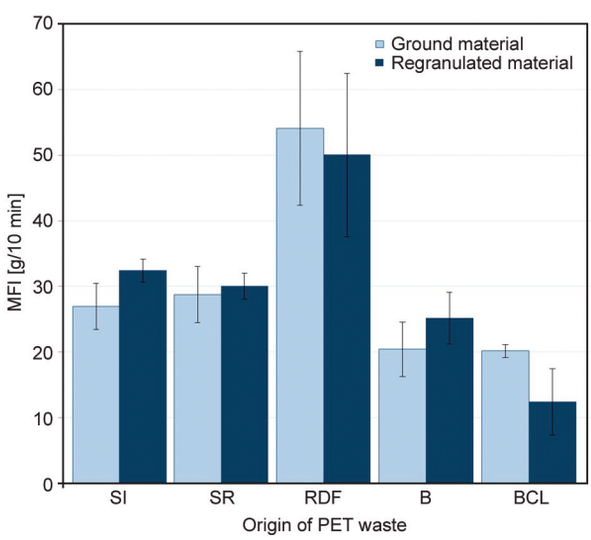

Figure 5. MFI values of PET wastes from different sources and with different pretreatment.
(SI – selective income; SR – sorting residue; RDF – refuse derived fuel; B – bottle; BCL – bottle with cap and label).

bottles. MFI of PET sorted from RDF was doubled probably because of the drastic deterioration of molecular weight as PET in RDF was degraded to a much higher extent and its quality changed in a wide range.

MFIs of regranulated SI and SR are not the same as regranulated waste PET bottles ground with the caps and labels together indicating that the polyolefin content of those real waste PETs shifted the MFI in the opposite direction. The reason behind the phenomenon was that the polyolefin content of SI and SR came not only from the caps and labels of drinking

bottles, but PET residues were also other than only bottles. Just as a short conclusion of MFI results, it was stated that PET A/B/D fractions are worth selecting because of their higher values as secondary raw materials since PET wastes (SI and SR) had much higher MFI, therefore lower molecular weight values. The wide range of MFI of real PET waste makes both the reprocessing and the quality control more difficult. Caps and labels beside the PET did not cause such significant deterioration that might be difficult to manage during reprocessing. Alsewailem and Alrefaie arrived at the same statements [23].

3.3. Results of capillary rheological measurements of the waste PETs

During the investigation of reprocessing conditions of waste PETs from different origins, capillary rheological measurements were carried out at the same temperature and in the same shear rate range in order to make a correct comparison of the results. Viscosity curves (Figure 6) were identified in the typical shear rate range not only for extrusion moulding but also for injection moulding. Viscosity curves of waste PETs showed the same trends in the whole shear rate range investigated. SI and SR samples had not only the same trend of viscosity but also the same values. The lowest viscosities were given by the waste PET derived from RDF in sync with the high MFI values aforementioned. Differences in the viscosities among the waste PETs disappeared in the shear rate range of injection moulding; therefore, processing parameters during injection moulding could be rather independent of the origin of waste PET.

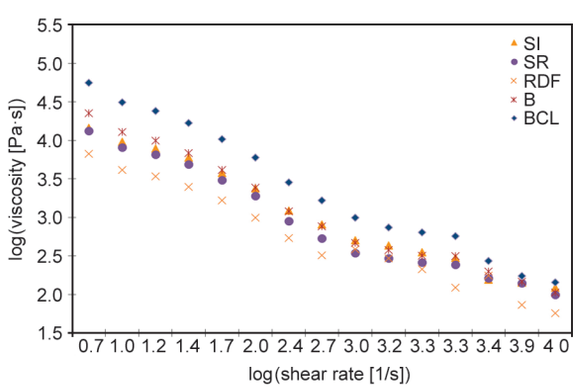

Figure 6. Viscosity of extruded PET wastes from different sources.
(SI – selective income; SR – sorting residue; RDF – refuse derived fuel; B – bottle; BCL – bottle with cap and label).

Viscosities of waste PETs (Figure 6) were the same in the region for injection moulding except the one sorted from RDF. The former phenomenon was believed to be a sign of the same processing parameters for waste PET regranulates (SI, SR and B) by injection moulding. Regranulate of PET bottles represented exactly the same viscosity curve as the SI and SR regranulates. PET bottles are worth sorting as selectively collected PET bottles with cap and label (BCL) had higher viscosities than the other PET waste streams in the shear rate region of extrusion moulding, forecasting also better mechanical properties of the extruded filaments. Reprocessing of waste PET materials caused an average viscosity increase of 5–7%, which can be partially drawn back to the measurement standard deviation.

3.4. Assessment of mechanical properties of the recycled PETs

Not only determination of the advantageous processing parameters is essential in plastic recycling for achieving a seemingly homogeneous sample, but also the level of mechanical properties. The tensile test is one of the most informative and fast measurements to identify the possible application fields. In our research, pellets originated from different qualities of PET waste were extrusion moulded, and tensile test of the extruded filaments was carried out. Since tensile properties of filaments conditioned at both 60 and 105 °C were measured, information was collected about samples conditioned at both below and above temperature of glass transition of the PET. Samples conditioned at 105 °C were expected to be brittle [36]. Specimens were broken almost promptly after tensile stress, reflected by only a few percent but no more than 7% for the elongation at break (Figure 7a). The highest value of elongation at break (EB) was measured for the sample from selectively collected PET waste and it was stated that the more contaminated the PET waste was the lower the elongation at break became regardless of the pre-conditioning parameters. No relevant difference was found between SI and SR, but an outstandingly brittle sample was obtained by processing PET originated from RDF. The latter is revealed by elongation at break being only 1–2%. Even the EB of the same sample conditioned at 60 °C was only 2%. Elongation at break of the samples conditioned at 60 °C was higher compared to the samples conditioned at 105 °C

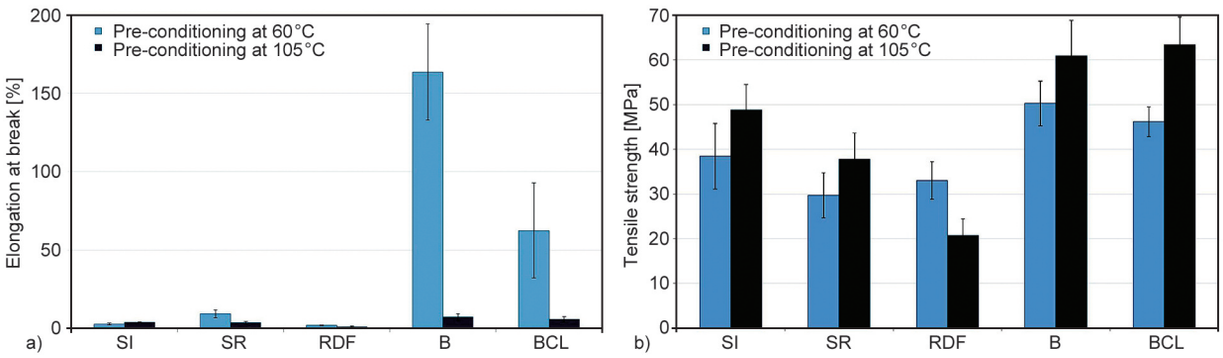

Figure 7. Elongation at break (a) and tensile strength (b) of extruded PET wastes from different sources. (SI – selective income; SR – sorting residue; RDF – refuse derived fuel; B – bottle; BCL – bottle with cap and label).

considering all of the waste PET grades, but the degree of the difference was dependent on the origin of the waste. High EB (approx. 150%) was only observed for the sample of selectively collected just PET material when pre-conditioning was carried out at 60 °C. Favourable EB (62%) was achieved in waste PET containing both cap and label meanwhile the degree of SD indicated inhomogeneity of the samples either. Close cold crystallization temperature was first thought to be responsible for the significant difference in EB of the samples with the same composition because a stiffer material with higher strength was produced, when pre-conditioning had been carried out at 105 °C (Figure 7b).

PET collected selectively (sample B) possessed the highest tensile strength (approx. 60 MPa), which was the less dependent on the pre-conditioning temperature (Figure 7b). The tensile strength of the sample from the PET bottle processed with cap and label together (BCL) decreased by only 10% compared to the one from just PET bottles collected selectively. Standard deviation also reduced, which was presumably caused by processing polyolefins accompanied to the PET [23]. The tensile strength of PET samples derived from the waste sorting plant reached only a maximum 40 MPa if pre-conditioning was at 60 °C before measurements. No significant difference at the same pre-conditioning was observed in the tensile strengths of the recycled PET samples originated from the fractions of SR and RDF. Approx. 30 MPa of tensile strength was achieved for both of the latter, which refers to the tensile strength level of general original polyolefins. Increasing pre-conditioning temperature up to 105 °C resulted in the improvement of tensile strength by an average of 20% increase, which could be basically accounted for thermal treatment closer to the cold crystallization processes mentioned before except for the RDF. A general conclusion about the homogeneity of a sample can be drawn from the level of the SD for tensile properties. Lower standard deviation referred, on the one hand, to better homogeneity of a sample produced from a waste resource, and on the other hand, to the similar extent of degradation processes within the sample caused by extrusion moulding. Inhomogeneity of a sample, *i.e.* higher standard deviations, may be caused by both quality of the waste material and the degradation reactions occurring during the processing.

Overall, it can be stated that in the later step in the sorting process, the waste PET came from less the pre-conditioning temperature affected EB. Therefore, inhomogeneity and a wide quality range of waste PET were suggested to be the crucial factors and not the cold crystallization.

3.5. Results of computed tomography investigation of the extruded waste PETs

Results shown in Figure 8 demonstrated the potential of X-ray tomography (CT) techniques to investigate the correlation between the source and properties of the wastes as well as the emerging inner structure of the samples. It can be clearly established that the uncontaminated B and BCL samples contain no gaseous inner porosity and inclusions of higher X-ray absorption coefficient (*e.g.* metal contamination) in the detectable size range (above 6 µm). These results highlight that the tensile tests show the real bulk properties of that samples without any influence of inner geometrical inhomogeneities. In the case of real waste, characterized by SI, SR, and RDF extruded rods, the structure is much more complex.

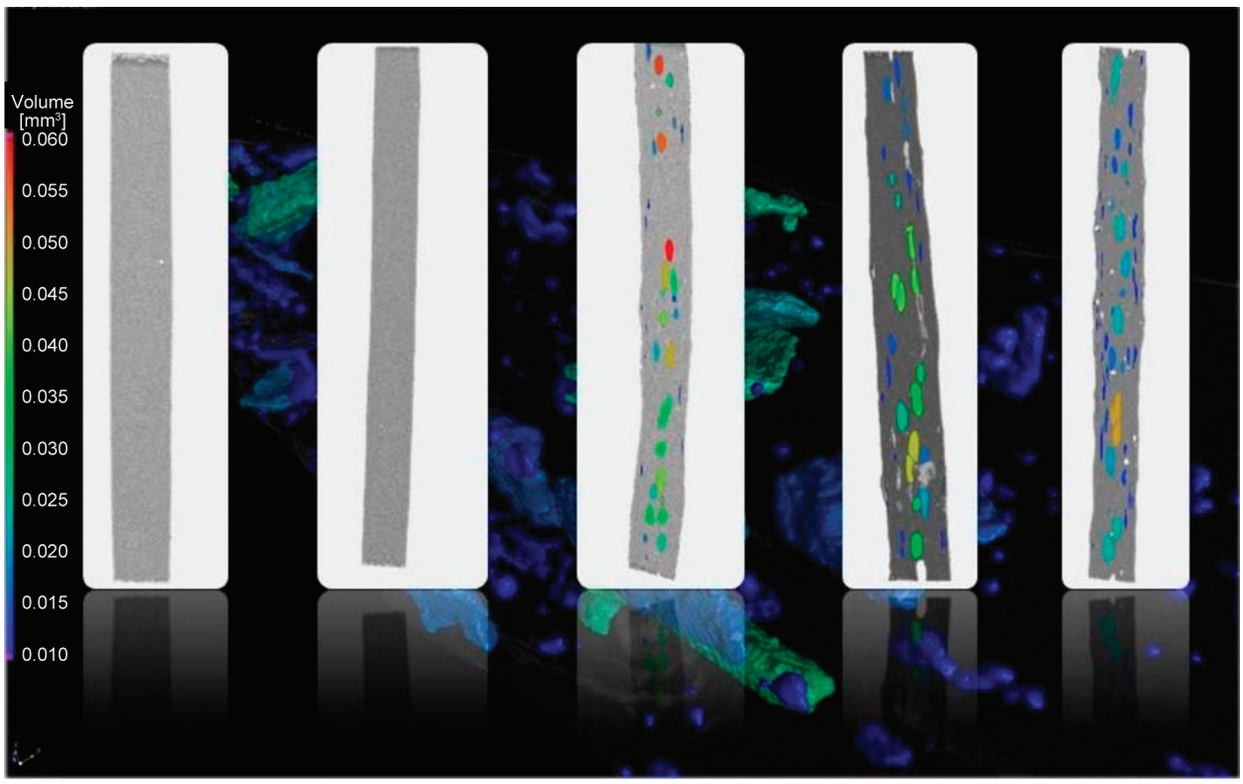

Figure 8. 2D slices produced from the 3D CT measurement of extruded samples. Coloured surfaces characterize the microporosity of the samples, while bright surfaces (white colour areas) belong to the inclusions. The image used as background is the 3D visualisation example of inclusions in the SR sample. (2D images characterize the results of B, BCL, SI, SR and RDF samples from left to right).
(SI – selective income; SR – sorting residue; RDF – refuse derived fuel; B – bottle; BCL – bottle with cap and label).

They contain simultaneously gaseous pores and inclusions of higher atomic numbers (*e.g.* metal particles). Therefore, it is proven that these material systems have to be treated as composite materials during the generally performed material testing techniques. While the material inhomogeneities can enhance the mechanical properties as a particle, a fibre, or plate reinforcement because of the appropriate adhesive forces due to its original task (the background image of Figure 8), the material vacancies can decrease the material cross-section making weak points in the structure. The voids have a certain degree of crack-stopping role, but investigating elastic material systems does not have a significantly positive influence on the mechanical properties. Summarized, it was proven and supported by the structural analysis, that the organic contamination content of the real plastic wastes produces gaseous voids in the structure because of the heat treatment during the recycling. Furthermore, the inhomogeneities of higher atomic numbers (metals) can be detected with various morphologies depending on the origin of waste. These parts significantly influence the properties of the produced recycled materials.

4. Conclusions

Due to the effort of the European Union to reduce the amount of single-use plastics and increase the rate of recycled plastic products, especially PET bottles, introduction of a deposit-refund system for PET drinking bottles has been urged. The aim of that study was to investigate recycling of PET wastes, particularly of non-drinking bottle types. The PET wastes were sampled from different real waste streams such as from a selective waste sorting plant, namely its selective income (SI) and its sorting residue (SR). PET waste was also separated from RDF from the MBT plant. The deposit-refund system (DRS) was symbolized by collected in-home bottles with (BCL) and without (B) caps and labels.

Since PET drinking bottles possess higher mechanical strength and elongation than the secondary raw material consisting of all kinds of PET wastes (SI),

in consequence, the introduction of a deposit-refund system on PET drinking bottles is highly recommended. PET bottles are worth selecting into PET-A, PET-B and PET-D fractions. The selective income (SI) containing all kinds of PET wastes and the sorting residue (SR) containing PET wastes except drinking bottles can be extruded at the same temperature profile and extrusion speed. Samples from both aforementioned waste PETs (SI, SR) had the same rheological and mechanical properties indicating on the first hand, that the absence of the PET drinking bottle waste did not cause significant property change and on the second hand, that PET selected from SR can be at least as advantageous secondary raw material as the SI. PET wastes derived from RDF showed the worst processing and mechanical properties. That behaviour was dedicated to the foreign organic compounds bonded to the PET waste unless it was carefully washed and conditioned before extrusion moulding. Based on the results, more serious washing pretreatment was suggested for achieving better performance of PET waste derived from the RDF fraction. X-ray tomography measurements showed a correlation between the source of waste plastic (purity) and the gas void structure (quantity and morphology) which influence essentially the obtained macroscopic mechanical properties.

In conclusion, based on our experimental results, it can be determined that valuable PET waste suitable for recycling is available beyond the drinking-bottles. However, this raises several other questions that need to be studied in the future, such as those summarized in the followings. The willingness of the citizens to engage in selective waste collection, the development of programs related to incentivizing selective waste collection, which can involve both educational activities and financial incentives (*e.g.* PAYT (pay-as-you-throw), DRS etc.), and the situation of the vulnerable segment of the population involved in collecting recyclable materials to earn some income are all points that need to be examined. These aforementioned aspects are planned to be studied within the framework of a comprehensive study in the future.

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