THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Material recycling of post-consumer flexible polyethylene packaging waste

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Gothenburg, Sweden, 2022

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Technical report no: IMS-2022-11

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Cover:

Schematic representation of material recycling process, from waste to new recycled products.

Printed by Chalmers Reproservice

Gothenburg, Sweden 2022

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ABSTRACT

The mechanical and thermal properties of recycled post-consumer flexible polyethylene packaging waste was studied, using material collected and sorted on a large-scale from two sources. Unwashed, laboratory-scale washed, industrial-scale washed, and industrial-scale washed and melt-compounded (industrially recycled) materials were used. The unwashed and washed flakes were melt-compounded on a laboratory scale with a twin-screw extruder using two different temperature profiles and two screw configurations. The pellets were then injection moulded.

Washing reduced the polymer molecular mass and the melt viscosity, making the material susceptible to further degradation during melt-compounding and more so at a higher temperature. The Young's modulus and tensile strength were affected by the washing but not by the compounding temperature or screw configuration, while the elongation-at-break were affected somewhat both by the washing and compounding temperature but not by the screw configuration. The moulded samples made of unwashed, laboratory-washed and industrial-washed materials had a stiffness, a tensile strength and an elongation-at-break as expected of conventional polyethylene grades available. The industrially recycled samples, however, had a lower stiffness, a slightly higher tensile strength and a significantly greater elongation-at-break. This significantly different mechanical properties of the industrially recycled material were probably due to the melt-filtration and possibly also to the additives in industrial melt-compounding. The overall results indicated that post-consumer flexible packaging waste had useful properties for further applications in new products, such as for non-food packaging.

Keywords: plastics recycling, polyethylene, mechanical properties, degradation, washing, melt-compounding, post-consumer flexible packaging waste

LIST OF APPENDED PAPERS

This licentiate thesis is based on the work contained in the following papers:

PAPER I **Boz Noyan, E.C.**; Venkatesh, A.; Boldizar, A. Mechanical and thermal properties of mixed PE fractions from post-consumer plastics packaging waste. Manuscript accepted (*ACS Omega, 2022*).

PAPER II **Boz Noyan, E.C.**; Venkatesh, A.; Boldizar, A. Washing of post-consumer flexible polyethylene packaging waste. Manuscript submitted.

The author also published the following paper that is not included in the thesis:

PAPER III Forsgren, L.; **Boz Noyan, E.C.**; Vega, A.; Yarahmadi, N.; Boldizar, A. The thermo-oxidative durability of polyethylene reinforced with wood-based fibres. *Polymer Degradation and Stability*, 2020, *181*, 109374.

CONTRIBUTION REPORT

PAPER I Main author. The study was planned together with the co-authors. The experimental work was executed by the author and A.V., while the characterizations by the author. The work was carried out under scientific supervision of A.B., and the results were analysed together with the co-authors. The first draft of the manuscript was written by the author and it was finalized with input from the co-authors.

PAPER II Main author. The study was planned together with the co-authors. The experimental work was executed by the author and A.V., while the characterizations by the author. The work was carried out under scientific supervision of A.B., and the results were analysed together with the co-authors. The first draft of the manuscript was written by the author and it was finalized with input from the co-authors.

PAPER III Shared main author. The study was planned together with the L.F. and A.B. The experimental work and most of the characterizations were executed by the author together with L.F. The mechanical testing was done by the author with an assistance of A.V. and L.F. The work was carried out under scientific supervision of A.B., and the results were analysed together with L.F. The first draft of the manuscript was written by the author together with L.F. and it was finalized with input from all the co-authors.

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1 INTRODUCTION

1.1 BACKGROUND

The ecological environment is of considerable concern in today's society. Parts of this concern relates to the use of resources, recycling and littering, and to the significant use and misuse of synthetic polymeric materials (plastics), especially packaging plastics, which are a significant contributor to the post-consumer waste stream.[1] The importance of managing plastics packaging waste is evident in the targets for recycling set by EU directives for the coming years.[2] Plastic packaging products, typically flexible film packaging, has been the largest application area for polymeric materials for many decades, constituting a major source of waste in society. For material recycling, some processing difficulties are expected related to the origin and character of the feedstock.[3-5]

The difficulties in recycling flexible plastic packaging can be attributed firstly to the heterogeneous composition of the waste, such as multi-layer materials, non-plastic components, a broad range of additives and a high level of contamination of food residues and other impurities.[6-8] The molecular degradation occurring during service and in the recycling process may also lead to significant changes in material properties, and the influence of impurities, collection schemes, blending and washing of mostly rigid plastic packaging waste has been investigated by several researchers,[9-17] and others have studied flexible plastic packaging waste.[18-20] Overall, it has been found that the properties of recycled materials are affected by many factors, but most of the materials can nevertheless be shaped into new products. However, publications on the recycling of post-consumer flexible plastic packaging waste are rather scarce. With this work, we hope to contribute to a better understanding of the development of properties of recycled flexible mixed polyethylene.

1.2 AIM

The purpose of this work was to study the thermal and mechanical properties of mixed polyethylene obtained from large-scale sorting of plastic packaging waste. The fraction studied was the 2D grade, consisting of the thin film fraction, one material originating from source-separated household packaging waste (SSHHPW) in Sweden and a second material of the same fraction originating from mixed municipal solid waste (MSW) in Norway. Although the whole range of polyethylenes may be present in the sorted fractions, the major part was probably lower density polyethylenes such as low-density polyethylene (LDPE) and linear low-density polyethylene (LLDPE). The influences of washing, compounding and injection moulding on the thermal and mechanical properties were included in the study. The washing and compounding processing were done on both a laboratory and an industrial scale, for comparison.

2 POST-CONSUMER PLASTIC PACKAGING WASTE

Plastics have been used in a broad range of application areas from packaging to building, transportation, electrical, household, agricultural and many other sectors. Plastics packaging has for a long time been the largest application sector for plastics, such as for food, beverage, personal care and household use and also in transport, retail and textiles. Packaging products mostly have a short lifetime, typically one year, for instance for bottles, trays, bags, films, disposable cutlery, food packaging and wrapping foil.[21,22] Materials in greater use are polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polystyrene (PS) and polyvinylchloride (PVC).[23] Plastic packaging represented 40 % of a total of 49.1 million tonnes (Mt) of plastics demand in EU 27 + 3 in 2020, while the second largest application sector was for building and construction representing 20 %.[23] For 2018 in EU 28 + NO/CH, it was reported that 61 wt.% of the total collected post-consumer plastic waste was packaging waste,[24] and thus a major part of the post-consumer waste stream.[1] This provided a strong basis for the ambitious recycling targets set by EU regarding plastic packaging waste in Directive (EU) 2018/852, which are 50 % by 2025 and 55 % by 2030.[2]

2.1 FLEXIBLE PLASTIC PACKAGING WASTE

Within plastic packaging applications, flexible materials account for the fastest growing segment but they are considered to be the most challenging for material recycling.[25] It was reported in 2017 that more than 50 % of the household waste in Norway and Sweden was PE films.[4] Later studies found that PE was the main polymer in a plastic packaging waste stream and specifically that LDPE and LLDPE were dominating in flexible packaging waste.[8,18,26] Flexible packaging can be designed as both monolayer and multilayer products, but the multilayers increase the complexity by combining different types of polymers such as PE, PP, PET, polyamide (PA), ethylene vinyl alcohol (EVOH), ethylene vinyl acetate (EVA) in different layers and also metallized layers.[27,28] The multilayers also add to the multitude of additives present in the materials. Such additives are typically introduced in melt-processing e.g., antioxidants, photo-stabilizers, flame retardants, heat stabilizers, plasticizers, compatibilizers, fillers, dyes and pigments.[3] The greater heterogeneity of both polymers and additives makes the recycling more challenging, possibly leading to low recovery rates especially for flexible plastic packaging waste.

3 RECYCLING OF POLYMERIC MATERIALS

The concept of resource efficiency is generally understood as making an extended use of products and materials to fulfil the total needs of society, to the benefit of a reduced environmental impact as a result of the reduced extraction and production of new materials and new products through the reuse of products, recycling of material, chemical recycling and energy recovery.[29] Recycling would thus improve the management of plastic waste. Here, materials recycling is expected to include all measures to prepare a useful raw material. After material recycling, a further recirculation scheme may include chemical recycling, where polymer chains are degraded, converted into low molecular weight products and used as a feedstock to produce new polymers or petrochemicals.[30-34] In this thesis the focus is on material recycling, in some cases referred to as mechanical recycling or as primary and secondary recycling.

3.1 MATERIAL RECYCLING

Material recycling is a well-known technology for transforming polymeric waste into new raw materials, including waste collection, sorting, shredding, washing, extrusion melt-mixing (compounding) and granulation of extruded material.[4,31,33,35,36]

It is well understood that a developed waste collection system is important for plastic waste to be treated effectively.[21] Waste collection and management schemes can vary between countries, as reported in several review papers.[4,37] In general, post-consumer plastic packaging waste from households is collected either as source-separated or as a part of the co-mingled household waste. The co-mingled household waste is generally termed mixed municipal solid waste (MSW) or mixed municipal residual waste (MRW), and in some cases it also includes organic waste such as food residues. In most cases, the source-separated plastic packaging waste from households includes both flexible and rigid plastics.[4,21,29,37-40]

Separation of the plastics packaging waste from the co-mingled waste stream is generally done in large-scale material recovery facilities, and it then joins the source-separated plastic packaging waste stream for further sorting into specific polymer streams, such as flexible PE, rigid PE and PET trays.[4,41] The sorting involved can be both manual and automated, depending on the purpose of the separation, the character of the feedstock and the intended subsequent application.[33,34,42,43] The main sorting techniques commonly involved are wet or dry gravitation and electrostatic, magnetic-density, froth-flotation and sensor-based sorting such as making use of near-infrared spectroscopy.[42,43] The sorting usually involves a combination of several technologies.

The next step after the sorting is usually washing to remove surface contamination,[44] proceeded by shredding to make the washing process more efficient. The main techniques commonly used to remove contaminants use water, a solvent or friction.[19] Wet friction washing has recently

come into use, involving intensive mechanical agitation at an elevated temperature, typically 70-90 °C, with added detergents, caustic soda (NaOH) and a surfactant.[40,44] A wet density sorting follows the wet-washing process to provide fines sorting, such as static or dynamic sink-float separation. Dry-washing is mainly done via friction using fast-rotating blades followed by dry density sorting, e.g. air classification.[40]

Extrusion compounding is commonly used for melt-mixing the washed plastic flakes in order to distribute and disperse constituents, to obtain homogeneity, but also to pressurize, melt-shape with a die and pelletize the material.[22,34,35] Melt-filtration can be applied in compounding to eliminate non-melting particles.[40] To adapt recycled raw materials for certain applications, compatibilizers, stabilizers, plasticizers or virgin polymers may be added at this stage.[15] The pellets produced can then be shaped into new products using conventional manufacturing methods such as extrusion and injection moulding.[30] Typical products made of flexible plastic packaging waste are plastic bags, trash cans and plastic lumber, but such products are generally not allowed for use in contact with food.[45]

3.2 POLYMER DEGRADATION

The polymer degradations include mechanical degradation due to deformations, photodegradation, or thermal degradation and chemical degradation by oxidation and hydrolysis.[46] Several degradation types in combination can induce early degradation and accelerate the polymer degradation,[47,48] but the main degradation mechanisms are oxidation and hydrolysis, both accelerated by temperature.[49] All the material recycling processes have moderate or high temperatures and ambient oxygen present, and this makes the thermo-oxidative degradation at both low and high temperatures of great interest.[7,50-52] It is also known that polyolefins are susceptible to chain scission, branching and cross-linking as a result of thermo-oxidative degradation during both processing and use.[22,39,53] Chain scission, branching and cross-linking may occur simultaneously and compete in the case of polyethylene degradation whereas chain scission dominates in the case of polypropylene degradation.[22,53-55] In general, the degradation mechanism is a complex phenomenon influenced by the compounding temperature,[50,51] the number of extrusion cycles,[56,57] the washing conditions,[13] the content of unsaturations in the polymer chains[52,58] and the molecular structure.[9] The thermo-oxidative degradation has been of particular interest in the present work.

3.3 CHALLENGES

Despite progress in the recycling of post-consumer plastic packaging waste and the findings of several studies,[12,18,39,59] there are still significant problems remaining in the contamination of the collected plastic waste and in the methods used in the recycling process. The municipal waste collection schemes for mixed waste are hampered by a high level of non-polymer impurities and a high moisture content, which reduces the efficiency of the sorting processes and lowers the recycling rates compared to those of source-separated collection.[18,21] The near-

infrared (NIR) spectroscopy, which is increasingly applied in the sorting, has limitations regarding the detection of black or dark parts, and of multilayer or coated plastics, and this leads to faulty-sorting and thus a lower purity level.[4,33,44,60] As a result, collected and sorted flexible plastic packaging waste is very heterogeneous, and after sorting is still contaminated by different polymer types and by non-polymeric coatings, adhesives, labels, dust, soil, grease and organic residues. The state of degradation of the heterogeneous plastic waste varies, however. A low thermo-oxidative degradation during the material recycling process is extremely interesting and of benefit for high and consistent properties of recycled raw materials.[26,31,61,62] Each step of the material recycling has its own problems, however, and these influence the efficiency of the recycling process and the properties of the resulting end-products.

4 MATERIALS AND METHODS

4.1 MATERIALS

Two bales, ca. 700 kg each, of sorted post-consumer flexible PE packaging waste (PE-2D) were received from large-scale plants, one from a sorting plant in Sweden and the other from a sorting-washing-compounding plant in Norway. The former originated from source-separated Swedish household plastic packaging waste (HHPPW) and the latter from Norwegian mixed municipal solid waste (MSW). From Norway, ca. 60 kg of industrially washed flakes and ca. 10 kg of industrially washed, compounded and melt filtered pellets were also received, henceforth called industrially washed flakes and industrially recycled pellets, respectively. All the materials were supplied in the spring of 2021 and shown in Figure 1.

Figure 1. The received materials, from left the sorted unwashed PE-2D bale, the industrially washed flakes and the industrially recycled pellets.

4.2 METHODS

4.2.1 Washing and compounding

Before further washing or compounding the plastics received were shredded using a Rapid Granulator 300-45 with a screen size of 17 mm. The Swedish (S) sorted PE-2D was washed in the laboratory for comparison with the unwashed state. The shredded flakes were first soaked in water with mild agitation at a solid-to-liquid ratio of 1/60, followed by a machine washing with a Vortex M6, SDL Atlas (USA) in 0.5 wt.% NaOH solution at 40 °C. The solid-to-liquid ratio in the washing was kept at 1/40 and 1 kg of material was washed per cycle. The washing cycle took about 40 minutes, comprising washing, rinsing and spinning. The washing and rinsing lasted for about 15 minutes each and the rinsing was done at 25 °C. The washing was followed by drying for at least 24 hours at 60 °C in a Moretto SX201 dryer.

The industrially washed flakes from Norway were used as received. According to the supplier, the industrial-scale washed material had been shredded with a screen size of 60-80 mm, then screened with a magnet, pre-washed at room temperature, further shredded with a wet-grinder

and friction hot washed with added NaOH and defoamer at 70-80 °C, followed by rinsing at room temperature, centrifuging and finally drying, the whole process taking approximately 30 min.

The unwashed and washed flakes, both laboratory-scale and industrial-scale, were compounded using a Werner & Pfleiderer ZSK 30 M9/2 co-rotating intermeshing twin-screw extruder (TSE), with a screw length and diameter of 969 mm and 30 mm, respectively. Two different temperature profiles were used, 100-150-200-200-200-210 °C and 100-150-200-240-240-250 °C and the influence of mixing was assessed using two different screw configurations, as shown in Figure 2. The first screw configuration (SC1) had only transport elements whereas the second configuration (SC2) had four mixing elements per screw shaft. The unwashed flakes were screened before compounding using a magnet grid to remove magnetic metal particles. This was not required for the washed flakes due to the separation achieved in the pre-soaking stage. All the materials were fed manually into the extruder and compounded at a screw rotation rate of 80 rpm resulting in an average throughput of 1.2 ± 0.4 kg/h. The compounded strands were granulated to obtain about 2 mm long pellets, ready for further shaping.

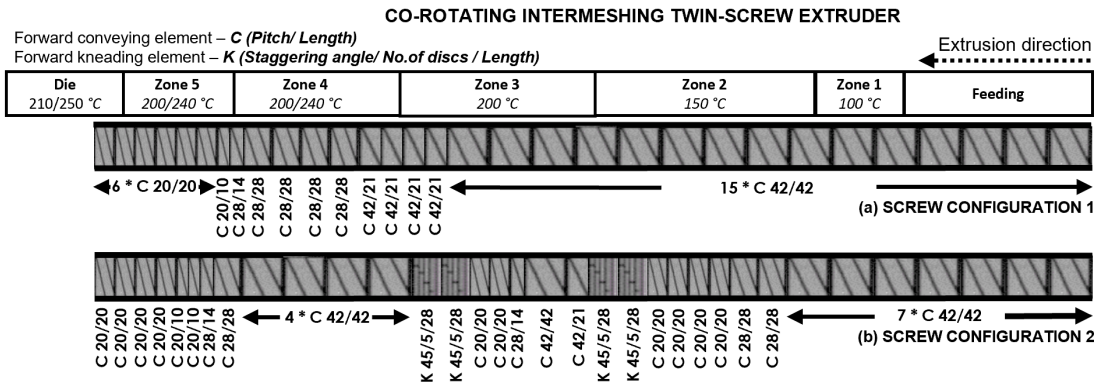

Figure 2. The co-rotating intermeshing twin-screw extruder screw configurations used for compounding; **(a)** Screw configuration 1: no mixing elements, **(b)** Screw configuration 2: with 4 mixing elements.

4.2.2 Shaping processing

The industrially recycled pellets and all the pellets compounded on a laboratory-scale were moulded into a frame, as shown in Figure 3, using an Arburg Allrounder 221M-250-5 injection moulding machine. The frame shape was chosen in order to assess the mechanical properties of different material structures commonly found in conventional injection moulded products, the gate region (G) with a mixed molecular orientation, the simple flow region (SF) with unidirectional flow and the weld line region (WL) where two flow fronts meet, as shown in Figure 3.

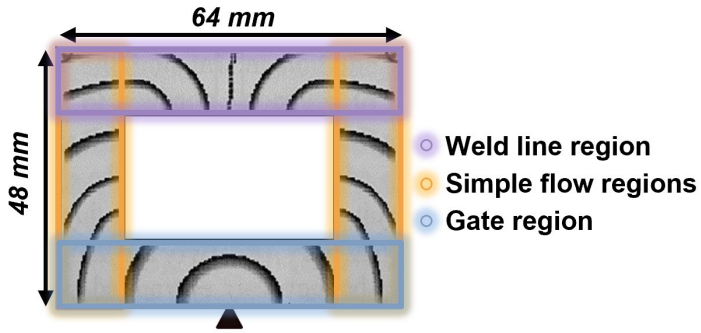

Figure 3. The filling pattern in the frame mould, producing a material with a thickness of 2 mm.

The injection moulding was carried out with a temperature profile of 120, 170, 200, 220, 220 °C and injection and holding pressures of 500 and 700 bar, respectively. The injection volume was adjusted for each material type, to achieve at least an 80 % meeting of the weld line width before the holding pressure was applied. Table 1 shows the sample codes used and Figure 4 shows examples of pellets and IM samples.

Table 1. Sample overview with details of origin, treatment and processing

		Compounding parameters		
Origin	**Treatment**	**Screw design**	**T profile (°C)**	**Sample code**
Swedish source-separated HHPPW	Unwashed	SC1	100-150-200-200-200-210	S_UW_SC1_200
	Unwashed	SC1	100-150-200-240-240-250	S_UW_SC1_240
	Unwashed	SC2	100-150-200-200-200-210	S_UW_SC2_200
	Unwashed	SC2	100-150-200-240-240-250	S_UW_SC2_240
	Laboratory-scale washed	SC2	100-150-200-200-200-210	S_LW_SC2_200
	Laboratory-scale washed	SC2	100-150-200-240-240-250	S_LW_SC2_240
Norwegian mixed MSW	Unwashed	SC1	100-150-200-200-200-210	N_UW_SC1_200
	Unwashed	SC1	100-150-200-240-240-250	N_UW_SC1_240
	Unwashed	SC2	100-150-200-200-200-210	N_UW_SC2_200
	Unwashed	SC2	100-150-200-240-240-250	N_UW_SC2_240
	Industrial-scale washed	SC1	100-150-200-200-200-210	N_IW_SC1_200
	Industrial-scale washed	SC1	100-150-200-240-240-250	N_IW_SC1_240
	Industrial-scale washed	SC2	100-150-200-200-200-210	N_IW_SC2_200
	Industrial-scale washed	SC2	100-150-200-240-240-250	N_IW_SC2_240
	Industrial-scale washed and compounded	N/A	N/A	N_rLDPE

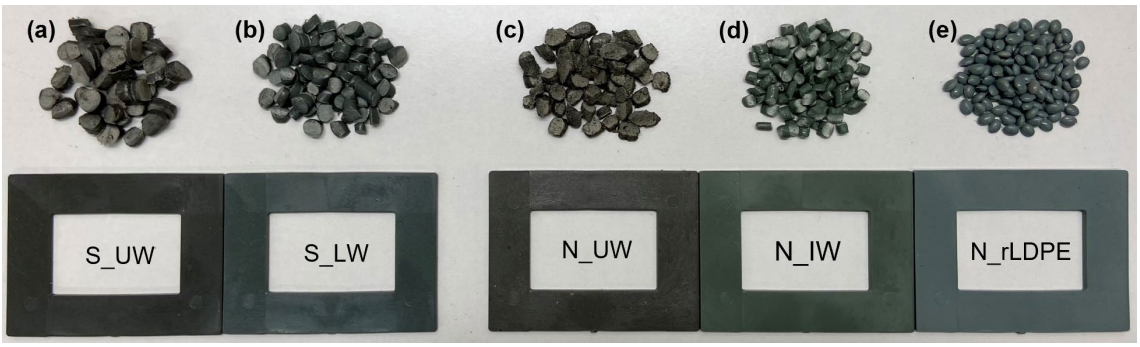

Figure 4. Examples of compounded pellets and injection moulded samples: **(a)-(d)** compounded in the laboratory using SC2 at 240 °C, **(e)** compounded in industry with melt filtration.

4.2.3 Characterization

To investigate the polymer composition of the Swedish sorted PE-2D fraction, a sampling scheme was applied over a five-week period, sampling a random bale two or three days per week, in weeks 47, 49 and 51 in 2020. A 200-litre sample was taken from each bale, resulting in eight bags of sorted plastic pieces. 100 pieces from each of the eight bags of sorted PE-2D fraction were analysed using a hand-held NIR analyser type microPHAZIR-Thermo Scientific and by measuring the mass using a laboratory balance from Sartorius AG. The weight percentages (wt.%) of the different type of polymers were reported.

Differential scanning calorimetry (DSC) was used for the thermal characterization, which included both the thermal transitions and the oxidation induction temperature (T_{ox}), determined according to ISO11357-1 and ISO 11357-6, respectively, with a Mettler-Toledo DSC 2. Both compounded pellets and injection-moulded samples were characterized. Circular sections, with a thickness of 0.65 ± 0.1 mm, were prepared for the T_{ox} measurements and the thermal transitions were assessed on samples with a weight of at least 5 mg. Air was used for purging when determining the T_{ox} and nitrogen when determining the thermal transitions, both at a heating rate of 10 °C/min. Duplicate measurements were made on each type of material and mean values were calculated. For the measurement of change in enthalpy ΔH, the baseline was taken from 60 to 132 °C. The results were reported for the first heating cycles.

The ash content of samples milled into a powder was measured by thermogravimetric analysis (TGA) using a TGA/DSC 3+ Star system from Mettler Toledo. The 3 ± 1 mg sample was heated from 25 °C to 650 °C at a rate of 10 °C/min in air at a flow rate of 50 mL/min. Duplicate measurements were made on each type of material and mean values were calculated. The reported ash content values were taken at 550°C.

The molecular weights of selected samples were assessed by high temperature gel permeation chromatography (HT-GPC) at ITS Testing Services (UK) Limited (Redcar, UK), after dissolution at a concentration of 4 mg/ml in 1,2,4 trichlorobenzene with 200 ppm butylated hydroxytoluene (BHT) as antioxidant. The analyses were performed using Polymer Laboratories GPC220 instrument with PlOlexis and PlOlexis guard columns with lengths of 3*30 cm at 160 °C, with an injection volume of 200 µl and a flow rate of 0.8 ml/min. Data were recorded and analysed using a Polymer Laboratories Cirrus software. The results shown are the weight-average molecular mass (M_w) and polydispersity index (PDI) based on two independent measurements.

A Ceast Modular Melt Flow instrument was used to determine the melt mass-flow rate (MFR) of the pellets, using a standard weight of 2.16 kg at 190 °C in accordance with ISO 1133-1:2011. The rheological behaviour of selected samples was studied using pellets obtained after compounding in a high-pressure capillary rheometer Rheograph 20 (Göttfert) at 220 °C using a constant piston speed at each shear rate between 10^3 and 10^1 s^{-1}. Three dies were used having

a diameter of 2mm and aspect ratios (L/D) of 5, 10, 15 for the Bagley correction with respect to the ISO Standard 11443:2021, and a Weissenberg-Rabinowitsch correction was applied. The graphical results show the corrected viscosity versus the shear rate, assessed with the die with a L/D ratio of 10.

The tensile properties were measured with a Zwick/Z2.5 instrument equipped with a 2 kN load cell. Test bars were cut from the three different regions of the moulded frame using an Elastocon EP 04 ISO 37-2 cutting die, corresponding to specimen type 5A in ISO 527-2, and kept in a conditioned environment at of 23 ± 2 °C and 50 ± 10 % relative humidity for at least 24 hours prior to the tensile tests to measure the Young's modulus, tensile strength, and elongation at break at a strain rate of 1 s^{-1}. The reported average values and standard deviations are based on five independent measurements.

5 MAIN RESULTS AND DISCUSSION

The compositions of the PE streams sampled in the three separate weeks are shown in Figure 5. Significant contents of PE, PP and PET were detected and other polymers and non-plastics as classified as “other”. W1 and W3 are each average of three successive days while W5 is the average of two days. The bars indicate the standard deviations.

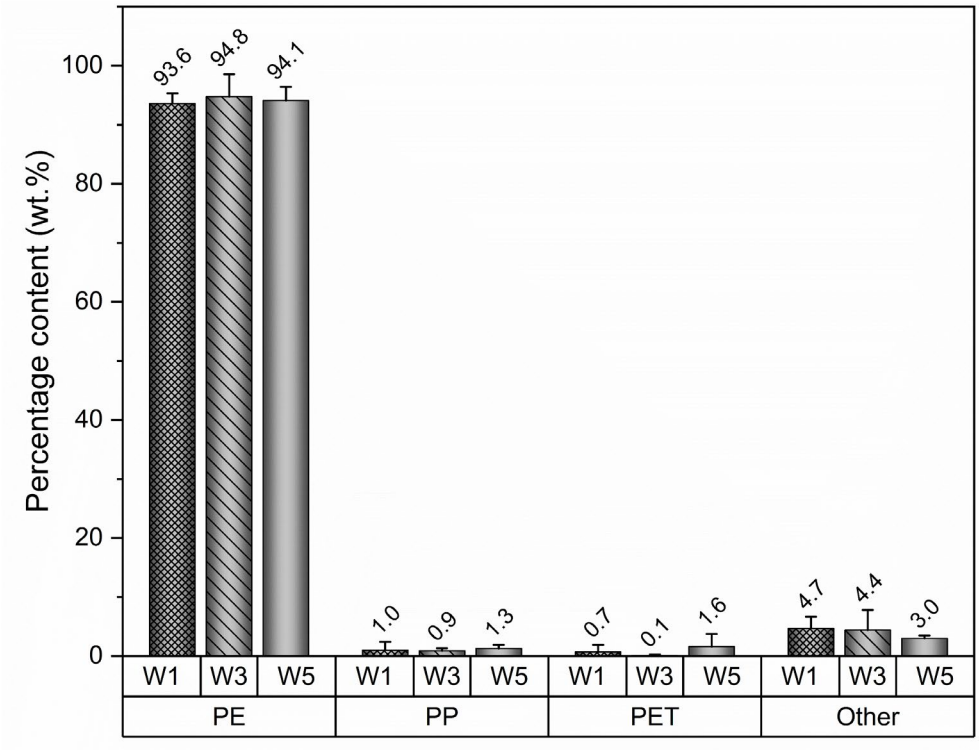

Figure 5. The weight proportions of different plastics in the flexible PE fractions sampled in three different weeks in Sweden.

The PE content was roughly the same on different days of the week and in different weeks during the sampling period, resulting in an overall average of 94 ± 2 wt.% at the 95 % confidence level. The level of contamination by other polymers, especially PP, was in agreement with values reported in the literature.[63,64]

The thermal transitions, oxidation induction temperatures (T_{ox}), ash contents, melt mass-flow rates (MFR), weight-average molecular masses (M_w) and polydispersity indices (PDI) of the samples are given in Table 2.

Table 2. The thermal, structural and rheological properties of the samples

Sample	T_{p1} (°C)		T_{p2} (°C)		T_{p3} (°C)		ΔH (J/g)		T_{ox} (°C)		Ash content (%)	M_w (g/mol)	PDI	MFR (g/10min)
	P	IM	P	IM	P	IM	P	IM	P	IM				
S_UW_SC1_200	111	110	125	126	161	161	76	73	222	222	5	N/A	N/A	0.7
S_UW_SC2_200	112	110	127	125	162	161	78	72	224	224	5	N/A	N/A	0.7
S_UW_SC1_240	112	110	126	126	161	161	76	70	216	214	5	N/A	N/A	0.8
S_UW_SC2_240	112	112	126	125	162	161	77	72	216	217	5	115500	4.5	0.8
S_LW_SC2_200	112	113	125	123	161	161	86	89	211	214	3	N/A	N/A	0.7
S_LW_SC2_240	111	110	125	123	162	161	94	89	210	211	3	114500	4.6	0.6
N_UW_SC1_200	111	111	125	125	161	161	66	63	224	223	11	123000	4.8	0.5
N_UW_SC2_200	111	112	125	126	162	161	68	66	232	231	11	124500	5.2	0.5
N_UW_SC1_240	111	112	124	126	161	161	67	63	226	226	10	N/A	N/A	0.6
N_UW_SC2_240	111	112	125	127	161	160	69	64	231	232	11	122000	5.0	0.6
N_IW_SC1_200	113	111	125	122	161	161	76	71	192	195	5	N/A	N/A	1.9
N_IW_SC2_200	113	111	126	123	161	161	76	71	195	194	5	87000	4.1	2.4
N_IW_SC1_240	113	111	123	124	159	160	76	70	180	182	5	N/A	N/A	4.8
N_IW_SC2_240	113	110	123	123	160	160	76	71	184	184	5	71500	4.4	3.9
N_rLDPE	118	109	125	125	161	161	74	69	188	193	5	103500	4.0	14.6

*P: pellets after compounding, IM: injection-moulded samples

There were no major differences in thermal transitions between the samples. The DSC curves of pellets compounded using SC2 at 240°C and of the injection moulded samples are shown in Figure 6. The samples had a main peak at 122-127 °C with a shoulder at 110-113 °C and a small peak at 159-162 °C, given in more detail in Table 2. The main peak (T_{p2}) was associated with a range of PEs (MD, LLD, and HD), the shoulder peak (T_{p1}) with the LDPE and the third peak (T_{p3}) with the PP, all typical melting temperature ranges.[12,65] In general, the effects on the melting peaks of compounding temperature, screw configuration and washing, whether laboratory-scale or industrial-scale, were negligible. The industrially recycled material had similar transition temperatures. The results showed small differences in the enthalpy of melting (ΔH), however, suggesting similar contents of PE and PP in the waste material from the two sources. The ΔH of the PP peak indicated that the Norwegian source (2.1 J/g) contained about twice as much PP as the Swedish source (1.1 J/g).

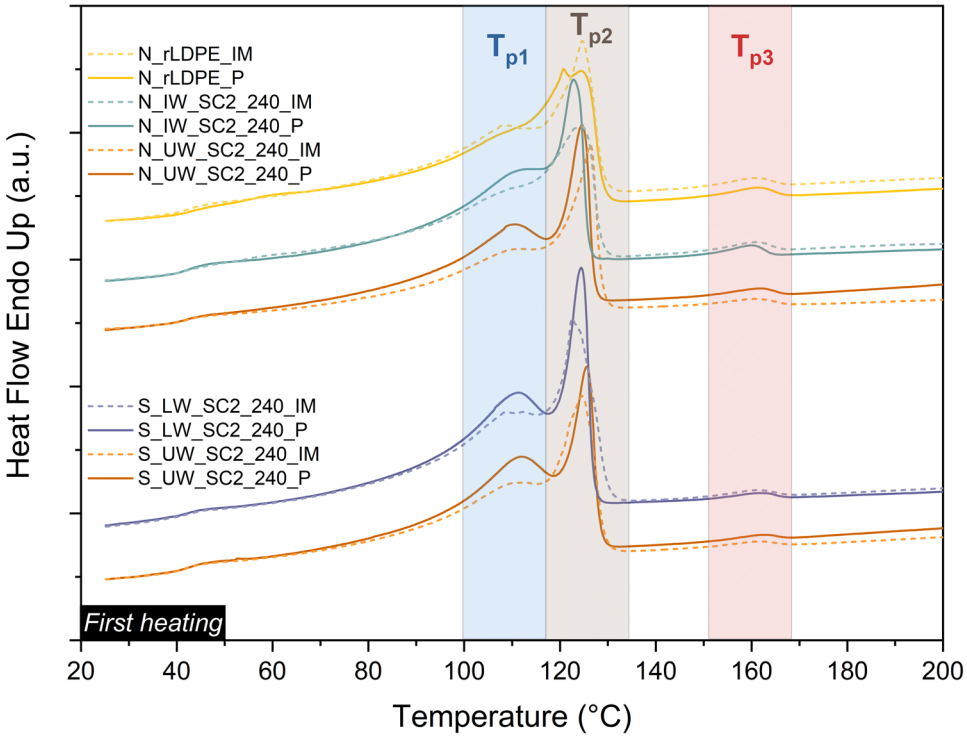

Figure 6. The first heating curves of the selected samples. The solid curves show the pellets (P) after compounding and the dashed curves the injection-moulded (IM) samples.

The melting peaks showed no great variations, but the ΔH values increased slightly with both laboratory-scale and industrial-scale washing, indicating that the degree of crystallization increased and that the content of impurities was reduced by washing,[66] and possibly also that some degradation occurred during washing, as the greater mobility of smaller polymer chains may lead a higher degree of crystallinity.[67] The industrially recycled sample had a level of crystallinity similar to that of the industrially washed material. The ΔH values of unwashed samples from the Swedish source were slightly higher than those from the Norwegian source, which may indicate different levels of contaminations due to different waste collection schemes.[18]

The T_{ox} values were similar for all unwashed samples, with those from the Norwegian source being slightly higher than those from the Swedish source. In the case of the unwashed samples from the Swedish source, the temperature of compounding had more influence than the screw configuration, whereas the opposite was observed in the case of the Norwegian samples. In general, washing led to a lower T_{ox} in both groups, implying that the washing possibly caused some degradation.[13] The T_{ox} reduction was much greater after industrial-scale washing, indicating that the conditions were relatively more degrading. The industrially recycled sample had a low T_{ox} value similar to that of the industrially washed samples, but the T_{ox} results suggested that the unwashed and the laboratory-washed samples still contained a significant amount of residual active stabilizer, since the T_{ox} for the un-stabilized virgin PE was expected to be 180 ± 5 °C.[68]

The ash contents of the samples were reduced by both laboratory-scale and industrial-scale washing, and more so by the latter. The industrially recycled sample had the same ash content as the industrially washed samples, which was expected since the samples were presumably washed in a similar manner. The values found before and after washing were similar to those reported by Gala et al.[3] and Gall et al.[11] In general, samples from the Norwegian source had a higher ash content, indicating a higher level of contamination.[18]

The molecular weight distributions of selected samples showed that the industrial washing reduced both M_w and PDI and that chain scission was the dominating degradation mechanism.[54,67] Increasing the compounding temperature reduced the M_w of industrial-scale washed samples, indicating increased degradation. The industrially recycled samples had M_w values intermediate between those of the unwashed and industrially washed samples, possibly indicating that thermo-oxidative stabilizers had been added in the compounding; these are commonly applied in the recycling industry.[22,67] In the case of the laboratory-scale washing of samples from the Swedish source, the washing led to a slight decrease in M_w and a slight increase in PDI. The samples from the Norwegian source had higher M_w values than the samples from the Swedish source. The unwashed samples from the Norwegian source had M_w and PDI values unaffected by the screw configuration or compounding temperature.

The viscosities are shown in Figure 7.

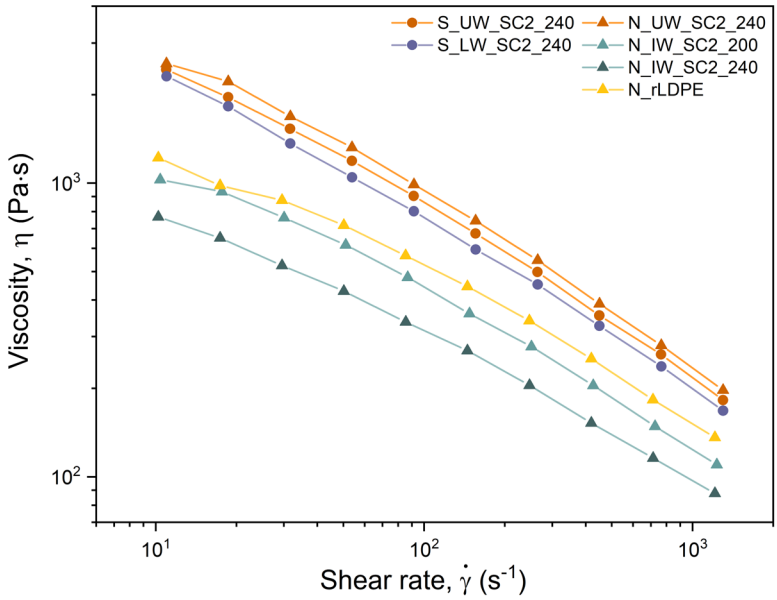

Figure 7. Viscosity as a function of shear rate for the different samples.

The viscosity data corresponded well with the GPC results,[50] but there are several disagreements between the MFR values in Table 2 and the viscosity curves in Figure 7, probably because the MFR measurement represents a poorly defined single point on the viscosity curve. The viscosity results agree more with the GPC results than with the MFR values. The effect of laboratory-scale washing on the viscosity was negligible, but the industrial-scale washing had a greater effect. The MFR values were similar for the unwashed

samples from both the Swedish and Norwegian sources, the former having slightly higher MFR values and slightly lower viscosities. The MFR values of both unwashed and washed samples were typical of blown film, pipe extrusion or extrusion blow moulded materials but lower than those of injection-moulded virgin materials. The industrially recycled material had a higher MFR value than the other materials, corresponding to that of a typical material for injection moulding to produce flexible products such as caps, toys, houseware and the like.[62,69]

The Young's modulus (MPa), tensile strength (MPa) and elongation-at-break (%) are shown in Figures 8, 9 and 10 respectively. The properties were measured in three different regions of the IM-samples, viz. in the weld line, in the gate and in the simple flow region, as illustrated in Figure 3.

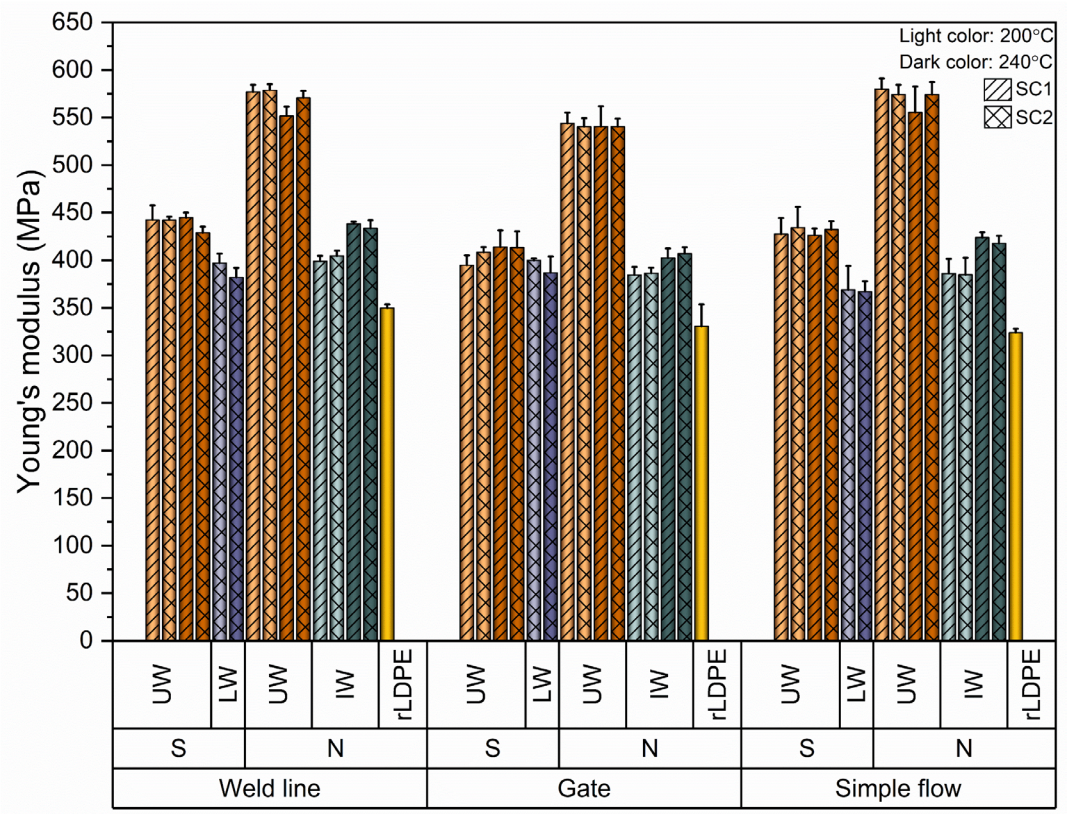

Figure 8. Young's modulus of the samples at different regions in the IM frame. The light colours show the samples compounded at 200 °C and the dark colours the samples compounded at 240 °C. Two different patterns correspond to the different screw configurations.

In general, neither the compounding parameters nor the structure of the moulded sample had a major influence on the Young's modulus. For the unwashed samples from the Swedish source, the average modulus value was 430 MPa, whereas for those from the Norwegian source it was 560 MPa. This difference in Young's modulus was probably due to the regional differences and to the level of contamination, although the composition of the PE-2D stream of the Norwegian source was not known in detail. The laboratory-scale washed samples had an average modulus of about 380 MPa, the industrial-scale washed samples about 410 MPa, and the industrially recycled samples about 340 MPa.

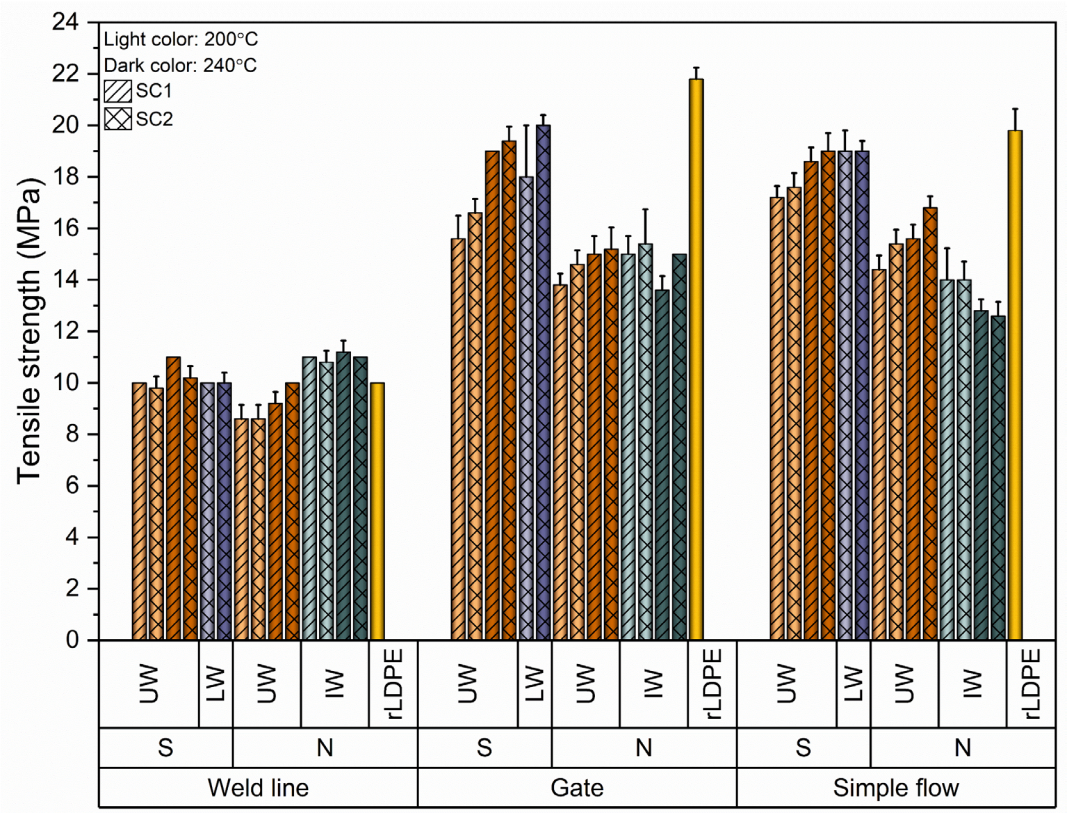

Figure 9. The tensile strength of samples at different regions in the IM frame. The light colours show the samples compounded at 200 °C and the dark colours the samples compounded at 240 °C. Two different patterns correspond to the different screw configurations.

The samples taken at the weld line had the lowest tensile strength of ca. 10 MPa, as expected, but the strength was nevertheless unexpectedly high and close to the reported value (13 MPa) for virgin PE-LLD.[70] In the other two regions of the moulded sample, the strength varied between the material from the Swedish and Norwegian sources. In the gate region (i.e. with a mixed molecular orientation) the samples from the Swedish source had an average tensile strength of 18 MPa for unwashed and 19 MPa for laboratory-scale washed, whereas both the unwashed and industrial-scale washed samples from the Norwegian source had an average strength of 15 MPa. In the simple flow region (i.e. the unidirectional flow region) the unwashed and laboratory-scale washed samples from the Swedish source showed the same average values as the samples from gate region, 18 and 19 MPa, respectively. Unwashed samples from the Norwegian source had the same average value of 15 MPa as in the gate region, but the industrially washed samples showed slight decrease to 13 MPa. The industrially recycled sample had the highest strength in both the gate and simple flow regions, 22 and 20 MPa, respectively.

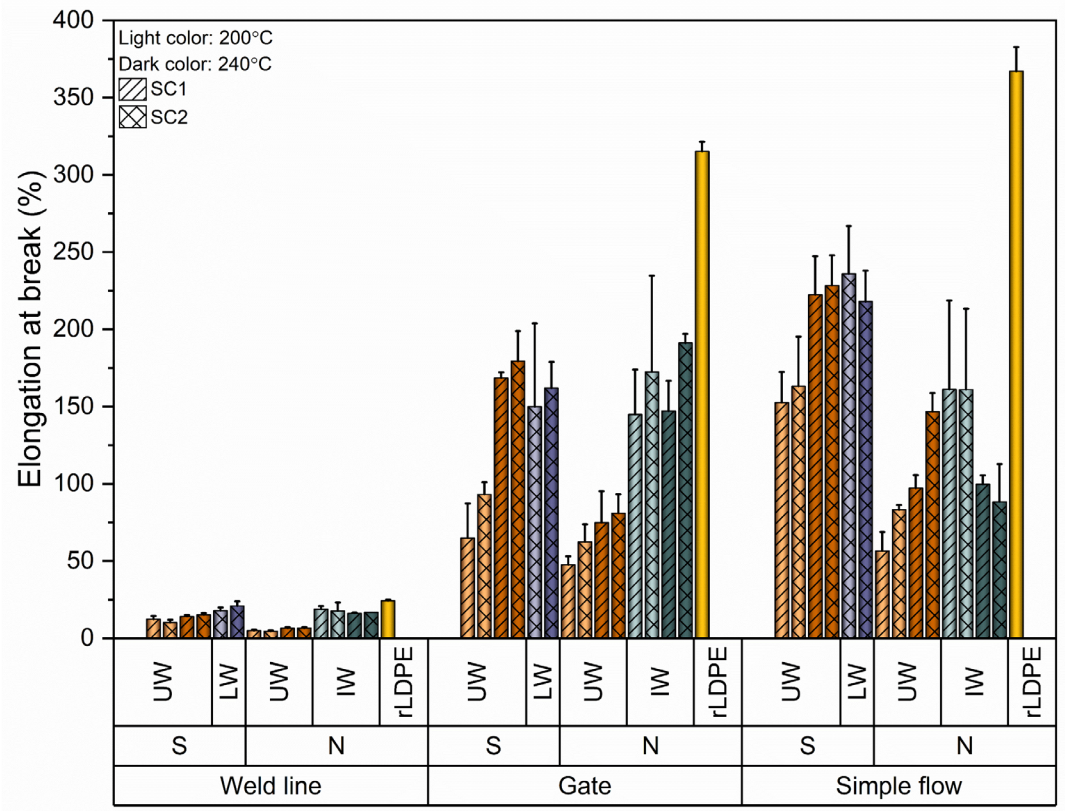

Figure 10. The elongation-at-break at samples taken from different regions in the IM frame. The light colours show the samples compounded at 200 °C and the dark colours the samples compounded at 240 °C. Two different patterns correspond to the different screw configurations.

The elongation-at-break was the lowest in all cases for the samples at the weld line. The average elongation-at-break for the unwashed and the laboratory-scale washed samples from the Swedish source was 13 and 19 %, and for unwashed, industrial-scale washed and industrially recycled samples from the Norwegian source it was 6, 18 and 24 %. In all regions of injection-moulded samples, the unwashed samples from the Norwegian source showed the lowest elongation-at-break while the industrially recycled sample had the highest value, with averages of 315 and 367 % respectively in the gate and simple flow regions.

In general, the influence of temperature during compounding on the elongation-at-break values was found to be greater than that of the screw configuration. In the gate region of the material from the Swedish source, the unwashed samples had averages of 79 % and 174 % when compounded at 200 and 240 °C, respectively, while the laboratory-scale washed samples had values of 150 and 162 %. From the Norwegian source, the unwashed samples had 55 and 78 % whereas the industrial-scale washed samples had 159 and 169 % when compounded at 200 and 240 °C, respectively. In the simple flow region, the unwashed samples from the Swedish source had an average elongation-at-break of 158 % when compounded at 200 °C and of 225 % when compounded at 240 °C. The laboratory-scale washed samples had values of 236 and 218 %, respectively. The unwashed samples from the Norwegian source had an average elongation-at-break of 70 and 122 %, while the industrial-scale washed samples had 161 and 94 % when compounded at 200 and 240 °C, respectively.

6 CONCLUSIONS

The sorted flexible PE packaging waste studied was found to have a consistent composition during the sampling period of five weeks, with a PE content of about 95 %. The compounding parameters, screw configuration and compounding temperature, had no great influence on the stiffness and strength of moulded samples and no significant influence on the melting temperature or crystallinity of the samples.

Thermo-oxidative degradation apparently occurred in the industrially washed samples, as shown by the reduced oxidation induction temperature and reduced molecular mass. Industrially washed materials were also prone to further degradation when compounded at a high temperature. The unwashed and laboratory-washed materials were not significantly degraded during washing and not significantly further degraded during compounding.

The differences in molecular mass and viscosity, in some cases small, indicated that polymer chain scission was the dominating degradation mechanism, although chain branching and crosslinking cannot be excluded.

The elongation at break increased with increasing mixing during the compounding of unwashed materials, particularly after the compounding temperature was increased and to a minor extent when the high-shear screw configuration was used. This was not however seen in the washed materials, possibly because the mixing effect competed with the degradation effect.

Moulded samples made of the industrially recycled material had somewhat lower modulus, were significantly stronger and had a greater elongation-at-break, probably because the melt-filtration reduced the particle contamination and because of the incorporation of additives in the large-scale compounding.

7 FUTURE WORK

The significant reduction in the thermo-oxidative stability of some of the samples implied that a better understanding is required regarding the degradation occurring during the material recycling process. In this respect, two important processing steps are the washing and the compounding. An investigation of the influence of washing parameters e.g., temperature and washing agent, is therefore essential. The aspects of compounding would also be interesting to expand upon, and to study the effects of melt-filtration, additives and odour reduction, in view of design and manufacturing aspects. Further studies on the shaping processing of recycled materials would also be of interest, to compare with the processing of new materials.

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ACKNOWLEDGEMENTS

I would like to express my extreme gratitude to my supervisor Antal Boldizar, for his guidance, time, endless support and patience. I am grateful for all the fruitful discussions and for your generous sharing of your knowledge. I have learnt so much and am looking forward to the coming years.

I would like to thank my co-supervisor Giada Lo Re and my examiner Christer Persson for their assistance whenever needed.

The Plastkretsen AB:s Stiftelse för Forskning and the Norwegian Retailers' Environment Fund are gratefully acknowledged for their financial support for this work. I would also like to thank Swedish Plastic Recycling AB for their firm support, with special thanks to Rickard Jansson for cooperation and interesting discussions. The RISE Research Institutes of Sweden is gratefully acknowledged for experimental support.

I wish especially to thank my co-author Abhijit Venkatesh for sharing your knowledge and for your constant support whenever I needed it.

I thank Roger Sagdahl and Eric Tam for their help in the laboratory and with technical issues. Blagica Smilevska is especially thanked for help with transport-related issues regarding the tonnes of plastic waste.

Thanks also go to all my colleagues at the Department of Industrial and Materials Science for the best possible environment and all the interesting talks during "fika" breaks. Special thanks go to my officemate, Claudia, for her support and care. All the members of the polymer group are especially thanked for their support, great conversations and the joyful times we spent together. I especially thank Lilian Forsgren, my former mentor and colleague, for all the knowledge transfer and assistance.

Anthony Bristow is gratefully acknowledged for the linguistic revisions.

I thank my dear family for their limitless love and support, I am genuinely thankful to have grown up in such a loving family. Many thanks to my friends who have always been there for me. Finally, my deepest gratitude to my husband Onur. Thank you for believing in me and for your love, patience, encouragement, care, support and thousands more. There are no words to describe how grateful I am to share my life with you…