

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

**Mechanical recycling of post-consumer polyethylene packaging
waste**

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Cover:

Washed flakes, compounded pellets and shaped 4L-container using a post-consumer rigid polyethylene packaging waste, illustrating the mechanical recycling process.

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ABSTRACT

The properties of mechanically recycled post-consumer flexible and rigid polyethylene packaging waste have been studied, using material collected and sorted on a large scale from two sources. The influence of processing conditions during washing and compounding were investigated using material unwashed and washed on a laboratory scale and also washed and further processed on a larger scale. The melt-compounding was done using a co-rotating intermeshing twin-screw extruder. The pellets produced on a laboratory scale were injection moulded, and the pellets produced with an upscaled washing and compounding procedure were blow moulded on an industrial scale.

Washing was necessary to remove the surface contaminants and, as expected, it improved the properties in both waste streams, but washing on a large scale at a high temperature with harsh chemicals such as sodium hydroxide made the flexible polyethylene material more susceptible to further degradation during melt-compounding, especially when a high temperature was used, which was shown by a significant lowering of the oxidation induction temperature and a deterioration in mechanical properties. This was not, however, observed in the rigid polyethylene material. On both a laboratory scale and a larger scale, the washing medium had a greater influence on the properties of both streams than the washing temperature. The properties of the unwashed and washed flexible polyethylene materials were, in general, not influenced by the screw configuration during compounding, but the compounding temperature led to some changes especially in the case of the washed samples. Rheological characterizations suggested that different levels of chain branching occurred in the rigid polyethylene samples compounded with different compounding temperatures and with the application of vacuum, whereas the water stripping during compounding had no influence.

Overall, the results indicated that both post-consumer flexible and rigid polyethylene packaging waste have useful applications in new products, such as non-food packaging. 4L-containers were successfully produced by blow moulding on an industrial scale using 100 % recycled rigid polyethylene material. However, the results of this work indicate that the washing and compounding conditions should be optimized for each stream since they are affected in different ways.

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

LIST OF APPENDED PAPERS

This thesis is based on the work reported in the following papers:

PAPER I Mechanical and thermal properties of mixed PE fractions from post-consumer plastics packaging waste

Boz Noyan, E.C.; Venkatesh, A.; Boldizar, A.

ACS Omega, **2022**, *7*, 45181-45188. doi: 10.1021/acsomega.2c05621

PAPER II Washing post-consumer flexible polyethylene packaging waste

Boz Noyan, E.C.; Venkatesh, A.; Boldizar, A.

Recycling, **2022**, *7*, 90. doi: 10.3390/recycling7060090

PAPER III A comparison between laboratory-scale and large-scale high-intensity washing of flexible polyethylene packaging waste

Boz Noyan, E.C.; Boldizar, A.

Polymer engineering and science, **2024**, *64*, 1877-1886. doi: 10.1002/pen.26674

PAPER IV Rheological and functional properties of mechanically recycled post-consumer rigid polyethylene packaging waste

Boz Noyan, E.C.; Rehle, F.; Boldizar, A.

Materials, **2024**, *17*, 1855. doi: 10.3390/ma17081855

PAPER V Blow moulding of mechanically recycled post-consumer rigid polyethylene packaging waste

Boz Noyan, E.C.; Boldizar, A.

Polymer engineering and science, **2024**. doi: 10.1002/pen.26962 (early view)

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

PAPER VI The thermo-oxidative durability of polyethylene reinforced with wood-based fibres

Forsgren, L.; Boz Noyan, E.C.; Vega, A.; Yarahmadi, N.; Boldizar, A.

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CONTRIBUTION REPORT

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

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1 INTRODUCTION

1.1 BACKGROUND

Thanks to their versatility, durability, light weight, low cost and many other attractive properties, plastics have found applications in areas such as agriculture, farming and gardening, automation, electricity and electronics, packaging, building and construction, houseware and many others.^{1,2} In the last couple of years, the production of plastics seems to have stabilized in European Union and in Norway, Switzerland and the United Kingdom (EU27+3). In 2022, it reached 54 million tonnes (Mt) in Europe, but it continues to increase globally and reached 400 Mt.² This progressive increase led to an increased concern in society regarding environmental problems caused by a significant amount of littering and misuse of plastics as well as by challenges in waste management.³

For a long time, packaging has been the largest application area for plastics, representing almost 40 % of the total plastics use, comprising plastic products typically having a short lifetime such as bottles, caps, trays, bags, films, wrapping foil, disposable cutlery, cosmetic containers, hygiene products, beverage and food packaging.^{2,4,5} Thus, packaging plastics is also the major component of post-consumer plastic waste, 57 wt.% of the 32 Mt of plastics waste collected in EU27+3 in 2022.² As a result, recycling has been favoured in the last few years with ongoing legislation and an EU Directive on packaging and packaging waste which has recently been updated with proposal to include the use of a recycled material content between 10 and 35 % in plastic packaging by 2030.^{6,7}

Although different aspects of mechanical recycling have been studied, there are still many problems remaining due to a lack of proper collection schemes, the heterogeneous composition of the waste, limitations in sorting technologies, difficulties in washing, and the molecular degradation occurring during service and in the recycling process^{1,8-20}, and these factors have been studied in the present work.

1.2 PLASTIC PACKAGING AND THEIR MANUFACTURE

Plastic packaging is usually divided into flexible and rigid plastic packaging. In general, polyolefins, i.e. polyethylene (PE, 22-26 %) and polypropylene (PP, 15-19 %), are the main polymers used in packaging applications, followed by poly (vinyl chloride) (PVC, 9-12 %), poly (ethylene terephthalate) (PET, 5-6 %), polystyrene (PS, 5 %) and polyurethane (PUR, 5 %).²¹ Plastic bags, films and foils are examples of flexible packaging, and low-density (LDPE) and linear-low-density polyethylene (LLDPE) account for most of the polymers used in these applications.²² Bottles, containers, trays, caps and lids are examples of rigid packaging with high-density polyethylene (HDPE), PP and PET being mainly used in these products.²²

PVC and PS are used in both flexible and rigid packaging but to a lesser extent than the other polymers.^{1,5,22}

To facilitate manufacture and to enhance the long-term durability and other properties, polymers are usually compounded with plasticizers, antioxidants, fillers, colorants, heat- and UV-stabilizers to prepare plastic base resins.^{5,23,24} The compounding is generally done in an extruder where polymer and additives are melted and mixed under heat, pressure and shear and are then solidified and pelletized.²⁵ An extruder can be used for shaping into a final product and is also a component of film-blowing, injection-moulding and blow-moulding machines.²⁵ Film blowing is one of the main techniques for producing flexible packaging films, foils and bags, where the plastic resin is melted in an extruder and formed into a hollow tube via a spiral or annular die while air is blown simultaneously to inflate the tube into a “bubble” which is then cooled. Using coextrusion, lamination or coating, multilayer structures which are often used in flexible packaging can also be made.²⁵ Injection moulding is used mainly to produce the caps and lids with a molten plastic resin, which is injected by the forward movement of a reciprocating screw in the extruder into a mould, where it is held under pressure for a certain time, cooled and then ejected.²⁶ Rigid plastic bottles, containers and jars are manufactured by blow moulding, where a so-called “parison”, i.e. hollow plastic tube, is formed either by extrusion or injection moulding and then inflated inside a closed mould to form a hollow product.²⁷ The required properties of the plastic resin differ and many grades of the same polymer have been developed. Structural properties, such as molecular weight and molecular weight distribution, flow properties, such as melt-mass flow rate and viscosity, melt properties, such as melt strength and drawability, and mechanical properties, such as stiffness, strength and toughness, vary allowing each grade to be tailored to give specific products using appropriate processing methods.

Before a plastic packaging reaches the market, labels with inks, adhesives and other plastic/non-plastic components are often incorporated in the product, and this increases the heterogeneity and complexity of the plastic packaging products which already include a broad range of additives, various grades of the same polymer, and cross-contamination from other types of polymer in a single product e.g., multilayer packaging and different polymers for caps and lids compared to the bottle or container.^{8,28-37} As a result, collected plastic packaging waste is expected to be very heterogenous in composition.

1.3 MECHANICAL RECYCLING, CURRENT STATE AND CHALLENGES

Resource efficiency requires the extended use of materials and products to meet society's needs and reduce the environmental impact by minimizing the extraction and production of new materials and products through the reuse of products, the recycling of waste and energy recovery.³⁸

Mechanical recycling is the most relevant strategy if reuse is not possible, and it is more efficient both economically and environmentally than other recycling technologies to reach the EU's recycling targets for plastic packaging waste.^{3,5,6,39} It includes waste collection, sorting, washing, compounding and pelletizing to transform polymeric waste into new raw materials.^{1,40-45} The first step in mechanical recycling is the collection of the plastic packaging waste which requires a proper and developed system and can vary between countries and between municipalities.^{1,4,46} The municipal waste consists of mixed waste or separately collected waste, e.g. paper and cardboard, glass, metals, plastics etc., from households and other sources such as small commercial businesses and public institutions.⁶ The plastic packaging from households is collected either separately or as a part of the co-mingled mixed municipal solid waste (MSW) or mixed municipal residual waste (MRW), where the flexible and rigid plastic packaging is collected together with residues of food, cosmetics and other chemicals.^{1,4,38,46-49}

The collected plastic packaging waste then goes for sorting to a material recovery facility (MRF)¹, which usually combines several technologies such as magnetic and eddy-current separation to remove ferrous and non-ferrous metals, size-based separation via trommel screens, density-based separation such as a ballistic separator, wind sifter to separate flexible and rigid, sensor-based separation using near-infrared spectroscopy, and sometimes also manual sorting depending on the purpose of the separation, the character of the feedstock and the intended subsequent application.⁵⁰⁻⁵³ In a co-mingled waste stream, the plastic packaging waste is first separated from materials such as metals, paper and glass and then sorted into separate polymer streams such as flexible PE, rigid PE, rigid PP, PET bottles and PET trays etc.¹

The sorted plastic packaging waste is then washed to remove surface contamination.⁴⁰ The contamination is usually very complex and the sorted plastic waste contains potential health risks unless it is properly cleaned.^{54,55} Washing is, therefore, an important step to create clean recyclates that meet the guidelines on using recycled materials especially in food-contact packaging.^{32,56} A washing line commonly involves size-reduction with a shredder, followed by pre-washing at room temperature, wet grinding, hot washing with some chemicals, rinsing at room temperature, followed by wet-density-separation by means of a static or dynamic sink-float separation technology, dewatering and drying. The hot washing step usually involves intensive mechanical agitation at an elevated temperature, typically 70-90 °C, with added detergents, caustic soda (NaOH) and a surfactant.^{49,54,57} Dry washing is another approach that uses friction by fast-rotating blades, usually followed by dry density sorting, e.g. air classification.⁴⁹ However, a more advanced cleaning procedure is required if the recycled materials are intended to be used for food-contact packaging.⁵⁶

The next step is compounding in an extruder to melt, mix, homogenize and pelletize the material.^{5,41} During compounding, melt filtration is usually applied to eliminate non-melting particles. Compatibilizers, stabilizers, plasticizers or virgin polymers may be added depending on the subsequent applications.^{5,49} The sorted, washed and compounded post-consumer plastic packaging waste can then be used as a new feedstock for further shaping into new products using conventional manufacturing methods such as extrusion, injection moulding, film blowing and blow moulding.⁴⁰

The latest report by Plastics Europe² states that 32 Mt of post-consumer plastic waste was collected in EU 27+3 in 2022, almost half of it by mixed waste collection and the other half by separate waste collection. Plastic packaging accounted for 57 wt.% of this post-consumer plastic waste, of which almost 38 wt.% was recycled, 45 wt.% energy was recovered and 17 wt.% was landfilled.² It was also stated that 12 wt.% of the 54 Mt of plastic products and components put onto the market in EU in 2022 were made of mechanically recycled post-consumer plastics used mainly in agriculture, farming and gardening, building and construction and packaging.² The same report illustrated the evolution of waste management for post-consumer plastic packaging waste in EU 27+3 between 2006 and 2022, with a 130 % increase in recycling rate, a 100 % increase in energy recovery and a 60 % decrease in landfilling.

Nevertheless, the expected increase in both municipal waste and plastic waste makes the targets set by the EU Directive⁶ on plastic packaging waste difficult to reach, due to the remaining challenges in the mechanical recycling process.^{58,59} The recycling rate of plastic packaging waste collected separately was 60 % higher than that collected in a mixed waste which was used either for energy recovery or landfill.⁶ The content of dirt, moisture and non-polymeric impurities also differs when the packaging is collected separately or in a mixed waste, and this also reduces the efficiency of the sorting processes.^{4,18} Near-infrared (NIR) spectroscopy, which is the technology currently being predominantly used, still has certain limitations, often resulting in faulty sorting and lower purity levels^{1,52,60}, although some improvement has been achieved by combining NIR with other technologies.^{61,62} Different aspects of washing have been studied usually on a laboratory scale, such as the influence of temperature and washing medium on cleaning efficiency, odour-removal, and degradation^{10,17,57,63-65} and the results of these studies indicate that there is no a common washing procedure that fits all types of sorted plastic packaging. Each specific stream requires optimization. Conventional washing is not sufficient and an advanced deep cleaning process is required if the recycled material is to be used in food-contact-packaging applications. Such cleaning procedure have been in use and are quite mature in the recycling of post-consumer PET bottles due to their inert character. Even though highly diffusive polymers like PE are considered harder to clean, there are promising studies using similar deep-cleaning approaches but these require further developments.^{32,55,66} In the extrusion compounding of plastics, it is well-known that thermo-oxidative degradation occurs at a high temperature, high shear and in the presence of oxygen, leading to a deterioration in the final properties.⁶⁷⁻⁶⁹ This makes the compounding of sorted and washed

plastic packaging waste critical, due to its degradation both after use and after washing, in addition to its heterogeneity and complexity due to a great variety of additives, different grades of the same polymer and cross-contamination by other polymers.^{29,32-34,36,37}

1.4 POLYMER DEGRADATION

Degradation during the mechanical recycling process has an important influence on the final properties of the recycled raw materials.^{28,43,70,71} It can be initiated in several ways, depending on the molecular structure and the environment to which the materials are exposed, external forces and deformation, photodegradation as a result of light radiation, thermal degradation due to high temperature especially when melting occurs, and chemical degradation initiated by oxygen or water which is usually referred to as oxidation or hydrolysis, respectively.⁷² In some cases, several of these mechanisms can co-exist leading to an early accelerated degradation.^{73,74} The thermo-oxidative degradation is of significant concern, occurring both during the lifetime at low temperatures with a high content of oxygen and during processing at high temperatures in the presence of oxygen.^{15,67,68,75} The degradation mechanism can be quite complex depending on the molecular structure and morphology of the polymer, on the method of synthesis, on chemical residues, and on processing conditions such as compounding temperature and multiple compounding.^{9,17,67,68,75-80} In the case of polyethylenes, chain scission, chain branching and crosslinking occur simultaneously during thermo-oxidative degradation.^{5,48,67,77,78,81,82}

The composition of plastic packaging products is, as mentioned previously, very heterogeneous from the beginning. Then when these products become waste after use, additional contaminants such as degradation products, food and chemical residues and greases occur. As a result, the sorted fraction of plastic packaging waste contains a great variety of contaminants and impurities, different grades of polymer, cross-contamination due to the original design of the product and due also to faulty sorting. This makes the expected degradation caused by subsequent processing, i.e. washing and compounding, even more complicated.

1.5 PROCESSING, DEGRADATION AND RHEOLOGICAL CHARACTERIZATION

The process-related degradation of polymers has been extensively studied. In many cases, the polymers were extruded multiple times and different methods have been applied to assess the degradation-related changes in physical, chemical and mechanical properties as well as the changes in molecular structure.⁸³ Fourier-transform infrared spectroscopy has been used to evaluate changes in carbonyl and vinyl groups, and changes in molar mass have been studied by size exclusion chromatography. Gas- and liquid-chromatography coupled with mass spectrometry have sometimes been used to assess the formation of low molecular weight compounds.⁸³ The thermo-oxidative durability of polymers and antioxidant stabilization have been studied mainly by oxidation-induction-time and -temperature using differential scanning calorimetry.

Rheological properties especially in the dynamic shear mode measured by rotational rheometry are sensitive to the molecular structure of polymers, and Van Gorp and Palmen⁸⁴ developed an approach to verify the time-temperature superposition principle by plotting the phase angle versus the complex shear modulus, later referred to as a van Gorp-Palmen plot (vGP-plot) used to establish a relation between the shape of the curve and molecular characteristics of the polymer e.g. chain branching, branching topology, and the length and amount of branched structure.⁸⁵⁻⁹⁹ In addition, properties such as the complex viscosity and crossover frequency, where the storage and loss modulus curves intersect, can also be used to compare polymers with different levels of chain branching, different molecular weights and different molecular weight distributions.^{77,86,94,98} Time-sweep rheometry in a dynamic shear mode has been also used to study the degradation of polymers¹⁰⁰⁻¹⁰², and the shear viscosity in capillary rheometry has been used to examine degradation-related changes in the polymer. Chain scission decreases the viscosity, for example, whereas chain branching and crosslinking increase the viscosity.⁷⁷ In addition to other characterization methods, rheological characterization offers a great potential for assessing the degradation-related structural changes that may occur during mechanical recycling.

Rheological characterization can also be used to evaluate the melt properties of polymers, which play an important role during e.g. extrusion, injection moulding, film blowing and blow moulding. The entrance pressure losses in capillary flow, for example, can be indirectly related to the melt elasticity of the polymer, which influences processing aspects such as die-swell ratio, bubble and parison formation and stability, haze formation and other appearance-related issues.^{27,103-114} Rheotens type measurements to measure the melt strength and drawability, which are important in the case of bubble and parison formation, can be used to study melt instability.^{27,103,105,109,113,115-117} The recycled post-consumer packaging waste should ideally have a potential for use in products similar to virgin grades, and the melt properties of the recycled pellets are therefore of great importance.

1.6 AIM OF THE WORK DESCRIBED IN THIS THESIS

Various aspects of recycling including the influence of impurities, waste collection, cleaning efficiency and odour removal have been studied, but studies of the impact of the whole recycling process on the material's functional properties are still scarce. The work described in this thesis has sought to clarify the influence of the process conditions in washing and compounding during the mechanical recycling of post-consumer plastic packaging waste in both flexible and rigid polyethylene streams, collected and sorted at large-scale facilities. The processing involved washing, compounding and shaping of the sorted plastic waste in order to complete the recycling cycle and evaluate the whole process. To better understand the effects of these processes, the properties of untreated and unwashed sorted waste were also studied, mainly for the flexible polyethylene packaging stream.

The influence of compounding temperature and screw configuration were assessed for the flexible polyethylene stream (**Papers I and II**) and the compounding temperature (**Paper IV**), applied vacuum and water stripping (**Paper V**) were investigated for the rigid polyethylene stream. For both streams, the influence of the washing medium and washing temperature (**Papers III, IV and V**) were evaluated. In general, the effects of different processing conditions were assessed by the thermal, structural, thermo-oxidative and rheological characterization of recycled pellets. The mechanical properties of the injection-moulded and blow-moulded samples were also evaluated. Special interest was devoted to the impact of the recycling process on the thermo-oxidative stability of the materials and to the structural changes resulting from degradation during the recycling process.

2 MATERIALS AND FEEDSTOCK CHARACTERIZATION

2.1 MATERIALS

Batches of sorted flexible and rigid polyethylene packaging waste were received from two large-scale facilities, one from a sorting facility in Sweden (S) and the other from a sorting and recycling facility in Norway (N). The sorting facility in Sweden receives separated plastic packaging waste collected from households in Sweden and then sorts the mixed plastic packaging waste into mono-plastic packaging streams, e.g. flexible PE, rigid PE, PET, PP and PS, after size-separation, density-separation and NIR sorting. The sorting and recycling facility in Norway receives mixed municipal residual waste and the mixed plastic waste is first separated from other materials such as metal and paper, by means of a magnetic separator, an eddy-current separator and a set of NIR sensors. The mixed plastic waste is then sorted into mono-plastic packaging streams by density-separation and NIR sorting, and these streams are washed and compounded. From each facility, one bale, ca. 700 kg, of sorted post-consumer flexible PE packaging waste and of rigid PE were received. Washed flakes, ca. 60 kg, of each stream were also received from the facility in Norway. All the materials were supplied in early 2021 and used in laboratory-scale processing. From the facility in Sweden, two bales, 1.2 tonnes in total, of sorted post-consumer rigid PE packaging waste were received in late 2023 and used in large-scale processing. According to the supplier, these bales contained 95 wt.% PE-rigid (high density PE), 2 wt.% PE-film (low density PE), 1 wt.% other films, 0.5 wt.% PP, 1 wt.% other rigid plastics and 0.5 wt.% residues.

In the laboratory-scale washings, sodium hydroxide (NaOH) and a detergent (D) were used as washing agents. NaOH was supplied by Merck in the form of pellets. The detergent was “Via professional liquid colour, perfume free”, from Unilever Professional, and it contained 5-15 % anionic surfactants, <5 % non-ionic surfactants, soap and <1 % enzymes, phenoxyethanol, and methyl isothiazolinone.

SABIC HDPE B5421, with a density of 0.954 g/cm³ and a melt mass-flow rate (*MFR*) at 190 °C and 2.16 kg of 0.16 g/10min, was used as a reference virgin grade HDPE which is typically used for blow moulding to produce containers for detergents, cleaners, shampoos and cosmetics. According to the supplier, this grade also contains antioxidants, but the type and amount were not stated.

2.2 FEEDSTOCK CHARACTERIZATION

Over a five-week period in late 2020, a random bale from each stream (flexible PE and rigid PE) at the facility in Sweden was sampled on two or three days during weeks 47, 49, and 51, i.e. a total of eight bales of each stream. From each bale a 200-litre sample was taken and 100 pieces from each 200-litre were identified using a hand-held NIR analyzer, type microPHAZIR-Thermo Scientific. From a bale of sorted rigid PE from the facility in Norway, a 200-litre sample was taken and 100 pieces were similarly identified. The weight percentages (wt.%) of the different types of polymer were reported.

3 PROCESSING

3.1 WASHING

Before washing or compounding on a laboratory scale, the sorted plastic streams were shredded using a Rapid Granulator 300-45 with a screen size of 17 mm. The laboratory-scale washing procedure was basically the same for both flexible and rigid PE streams, except for differences in washing medium and temperature. A 1 kg sample of the shredded plastic waste was soaked in 60 L of tap water at room temperature, the plastic flakes being manually agitated to improve wetting, and allowed to soak for 7 min. Two batches of the floating fraction were used in each washing cycle in a Vortex M6, SDL Atlas machine using 72 L of water. Each washing cycle lasted for 45 min and included 15 min agitation at 100 rpm, draining, 15 min rinsing with fresh water at 25 °C, draining and 4 min spinning at 600 rpm. The soaking, washing and rinsing times were chosen based on earlier studies.^{17,65}

The sorted flexible PE received from the facility in Norway was washed in three different media; with water without any added agent, with 0.5 wt.% of NaOH in 72 L of water, and with 0.4 wt.% of detergent in 72 L of water, and each type of washing was done at both 25 °C and 40 °C. The sorted flexible PE received from the facility in Sweden was washed at only 40 °C and with only 0.5 wt.% of NaOH in 72 L of water. After the washing, three or four batches of the flakes of each type were dried in a Moretto SX201 dryer at 60 °C for at least 18-20 h until a humidity of 4 % RH was recorded by a EL-USB-2-LCD Lascar Electronics hygrometer inside the dryer. The sorted rigid PE waste received from Norway and Sweden was washed with water without any added agent and with 0.4 wt.% of detergent in 72 L of water at 25 °C, followed similarly by drying at 60 °C overnight until a humidity of 4 % RH was reached.

Flakes of flexible and rigid PE, washed in a large-scale facility in Norway, were used as received. The washing procedures for the two streams were the same and, according to the supplier, the sorted plastic waste 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, detergent, defoamer and other additives at 70-80 °C, followed by rinsing with fresh water at room temperature, further density separation in a tank centrifuge with added fresh water at room temperature, dewatering and finally thermal drying, the whole process taking approximately 30 min. In the case of the rigid PE stream, remaining flexible materials were also separated via a wind sifter after drying.

The sorted rigid PE waste received from Sweden was washed on a pilot scale at Herbold Meckesheim GmbH (Germany). The washing involved shredding with a screen size of 80 mm, pre-washing at room temperature, wet grinding, friction washing with water at room temperature, centrifugation, batch washing in a container with stirrer, friction washing with water at room temperature, centrifugation, density separation in a hydrocyclone, friction washing with water at room temperature and centrifugation, the whole process taking

approximately 35 min. Batch washing was done under three different conditions, the first with anti-fat detergent and defoamer at 40 °C, the second with NaOH, anti-fat detergent and defoamer at 40 °C, and the third with NaOH, anti-fat detergent and defoamer at 80 °C, the other steps in the procedure being kept the same.

3.2 COMPOUNDING

The laboratory-scale compounding was done using a Werner & Pfleiderer ZSK 30 M9/2 co-rotating intermeshing twin-screw extruder with a screw length to diameter ratio (L/D) of 32 and a screw diameter of 30 mm. Two different temperature profiles and two different screw configurations, SC1 and SC2, were used, as shown in Figure 1. The compounding temperature profiles used, from heating zone 1 to the die, were 100-150-200-200-200-210 °C (flexible LT) and 100-150-200-240-240-250 °C (flexible HT). The unwashed flakes of flexible PE from both Sweden and Norway and the flakes washed on a large scale from Norway were compounded with both SC1 and SC2 and both temperature profiles. The flakes of flexible PE from both Sweden and Norway washed with NaOH at 40 °C on a laboratory scale were compounded with SC2 and both temperature profiles, but the samples from Norway washed under other conditions on a laboratory scale, as described in section 3.1, were compounded with SC2 at 100-150-200-240-240-250 °C.

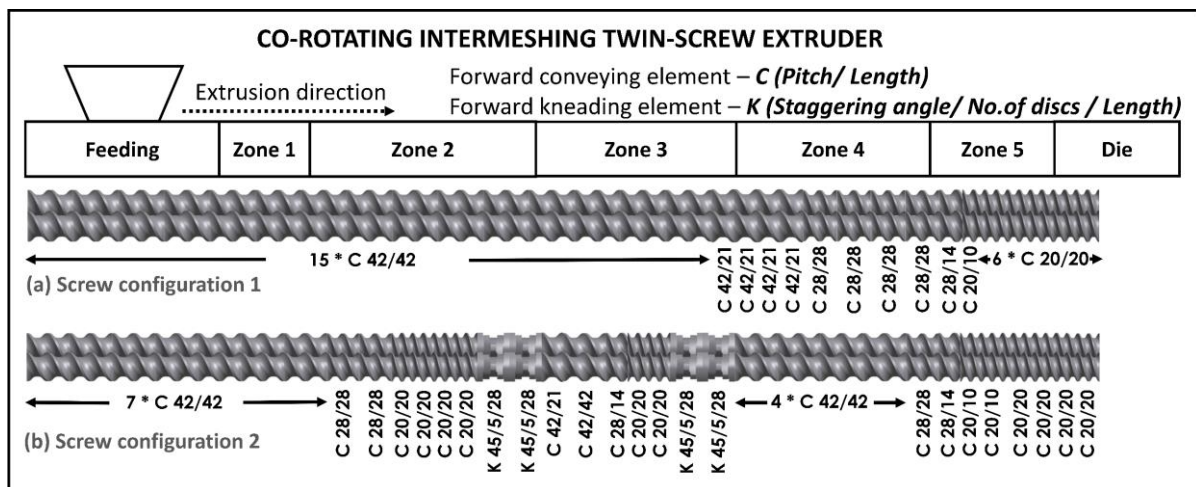


Figure 1. The co-rotating intermeshing twin-screw extruder screw configurations used for compounding; (a) screw configuration 1 (SC1): no mixing elements, (b) screw configuration 2 (SC2): with four mixing elements per screw shaft.

The rigid PE flakes were compounded only after washing using SC2. The samples from both Sweden and Norway washed on a laboratory scale with water alone and the flakes washed on a large scale from Norway were compounded with three different temperature profiles, 110-160-200-200-200-210 °C (rigid LT), 110-160-200-240-240-250 °C (rigid MT) and 110-160-210-260-260-270 °C (rigid HT). The samples from both Sweden and Norway washed on a laboratory scale with detergent were compounded only at 110-160-200-240-240-250 °C.

Before compounding, the unwashed flakes were screened using a magnet grid to remove magnetic particles, but this was not required for the washed flakes, due to the separation achieved in the pre-soaking stage. In all compoundings, the degassing port at barrel zone 4 was kept open. Both flexible and rigid PE materials were fed manually into the extruder and compounded at a screw rotation rate of 80 rpm. 2-3 kg of each flexible PE sample were compounded at an average throughput rate of 1.4 ± 0.5 kg/h, and 3-4 kg of each rigid PE sample were compounded at an average throughput rate of 2.1 ± 0.4 kg/h. The compounded strands were pelletized using a Dreher pelletizer type SG10Ni.

The pilot-scale compounding of the pilot-scale-washed rigid PE from Sweden was done using a Coperion ZSK 58 Mc18 co-rotating intermeshing twin-screw extruder. The materials were fed automatically from a hopper into barrel zone 5 and passed through barrel zone 13, followed by a melt filter with a screen size of 150 μm , a melt pump and a pelletizer, as shown in Figure 2. The same screw configuration, including some kneading and reverse elements in barrel zone 7 with mixing elements in barrel zone 10, and the same temperature profile, as shown in Figure 2 which is considered as medium-temperature profile (MT), were used for all the materials. The number and type of the special elements were not shared by the Coperion due to confidentiality.

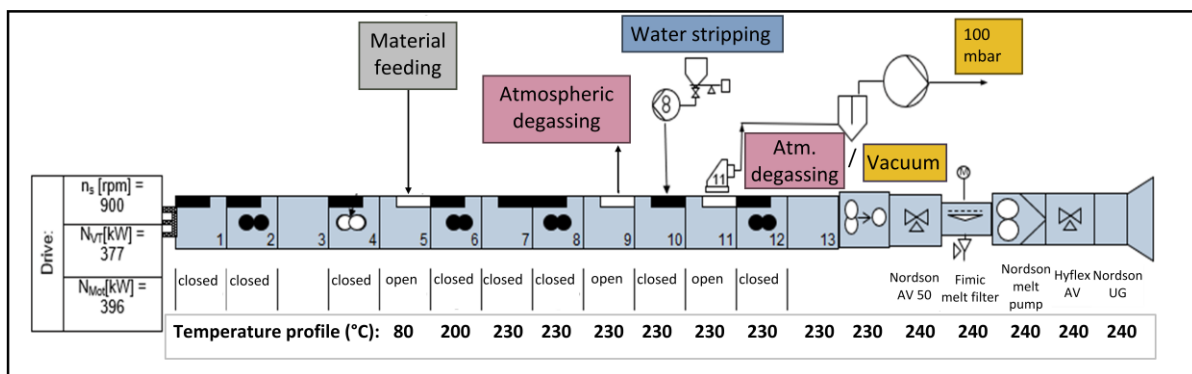


Figure 2. A schematic representation of the pilot-scale compounding, with the temperature profile used.

Each type of sample washed under different conditions was compounded with 100 mbar vacuum applied in barrel zone 11, but the samples washed with NaOH at 40 °C and 80 °C were also compounded with atmospheric degassing in barrel zone 11. The sample washed with NaOH at 80 °C was additionally compounded with water stripping at a rate of 1 kg/h in barrel zone 10, followed by an applied vacuum. All the samples were compounded at a screw rotation rate of 300 rpm with an average throughput rate of 200 kg/h. An underwater pelletizer BKG AH2000 with a 15 x 2.8 mm die plate was used to pelletize the compounded materials.

The sorted rigid PE waste received from the facility in Sweden, that was used for pilot-scale processing, was also shredded and batch-mixed without washing for comparison. The shredding was done using a Rapid Granulator 300-45KU with a sieve size of 6 mm and the mixing was carried out with a Brabender internal mixer AEV 330 having counter rotating screws W50 at 40 rpm for 5 min at 190 °C. The molten polymer was manually flattened and cut into small pieces after solidification.

3.3 SHAPING

All the materials compounded on a laboratory scale were injection moulded into a frame using an Arburg Allrounder 221M-250-5 injection moulding machine. In order to assess the mechanical properties of material structures commonly found in conventional injection moulded products, the frame had a gate region (G) with a mixed molecular orientation, a simple flow region (SF) with unidirectional flow and a weld line region (WL) where two flow fronts meet, as shown in Figure 3. The SF region would correspond well to the structure and properties of a standard tensile test bar.

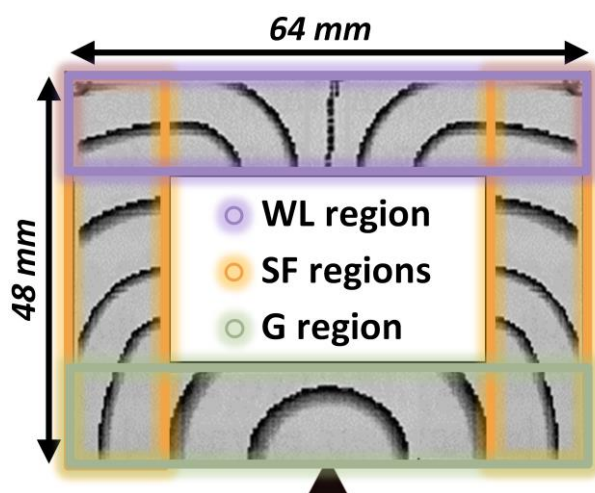


Figure 3. The filling pattern in the frame mould, producing a material with a thickness of 2 mm.

For the flexible PE samples, 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. For the rigid PE samples, the moulding was done with a temperature profile of 120-160-200-240-240 °C and injection and holding pressures of 500 and 900 bar. 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.

The rigid PE materials compounded on a pilot scale were blow moulded to form 4L-containers, Figure 4, on an industrial scale using an Uniloy Milacron UMS 12s machine equipped with W. Müller's torpedo and spider-leg mandrel die head at Emballator AB (Mellerud, Sweden). The temperature profile was the same for all the materials with feeding zone 1 set to 30 °C, heating zones 2-12 set to 200 °C and the die head set to 190 °C. The screw rotation rate and the die-opening were adjusted for each material to produce bottles weighing 145 ± 5 g.

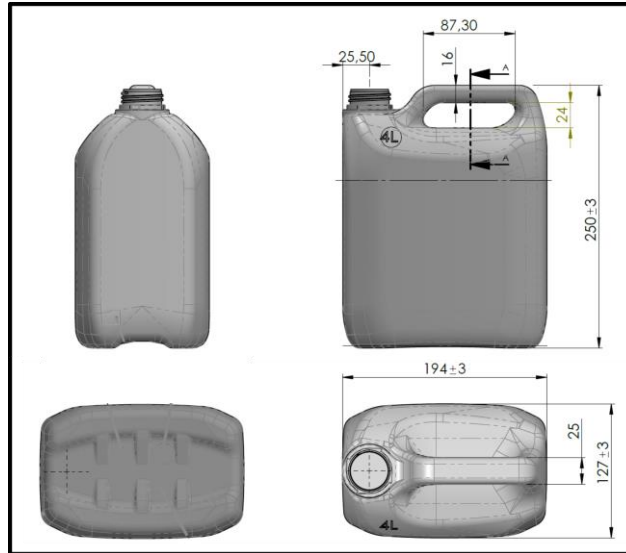


Figure 4. Drawing of the 4L-container with dimensions in mm.

During the blow-moulding, the operating window limits were investigated. All the process parameters were kept fixed except for the die-opening which was varied between 0.4 and 8 mm, and the screw rotation rate which was varied between 10 and 40 rpm, resulting in a pressure range at the end of the extruder between 25 and 160 bar. The parisons shaped under different die-openings and extrusion pressures were examined visually to observe pleating or other melt instabilities.¹¹⁸ The occurrence of pleating or melt instability was rated subjectively from level-0 (none) to level-3 (very severe), as shown in Figure 5, to give a relative comparison between the processing of virgin grade HDPE (VG-HDPE) and recycled rigid PE.

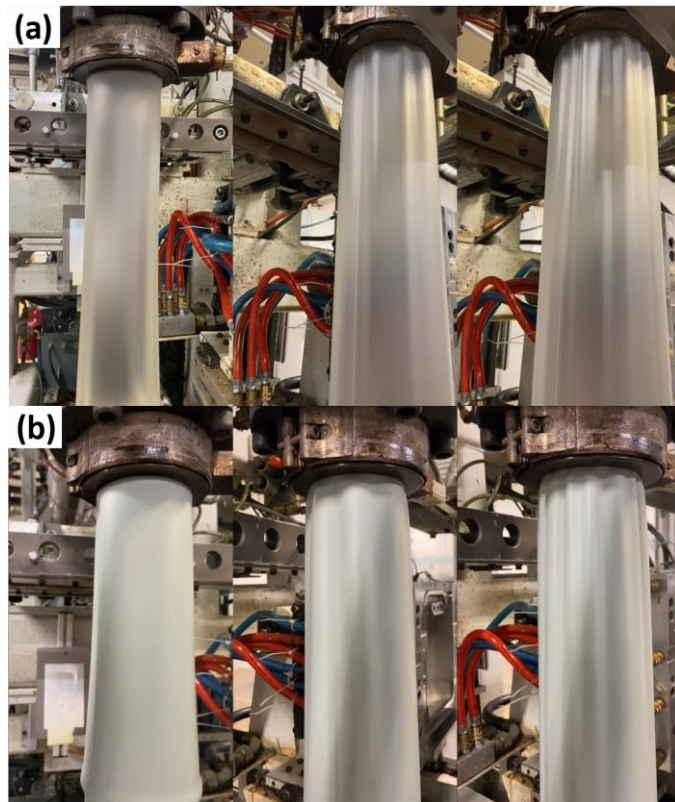


Figure 5. Pleating level-0 (left), -1 (middle) and -3 (right) for (a) VG-HDPE, (b) S-R_PW-NaOH40_Vac.

Figure 6 shows a summary of all the processing, and Table 1 lists the samples with processing conditions and notations.

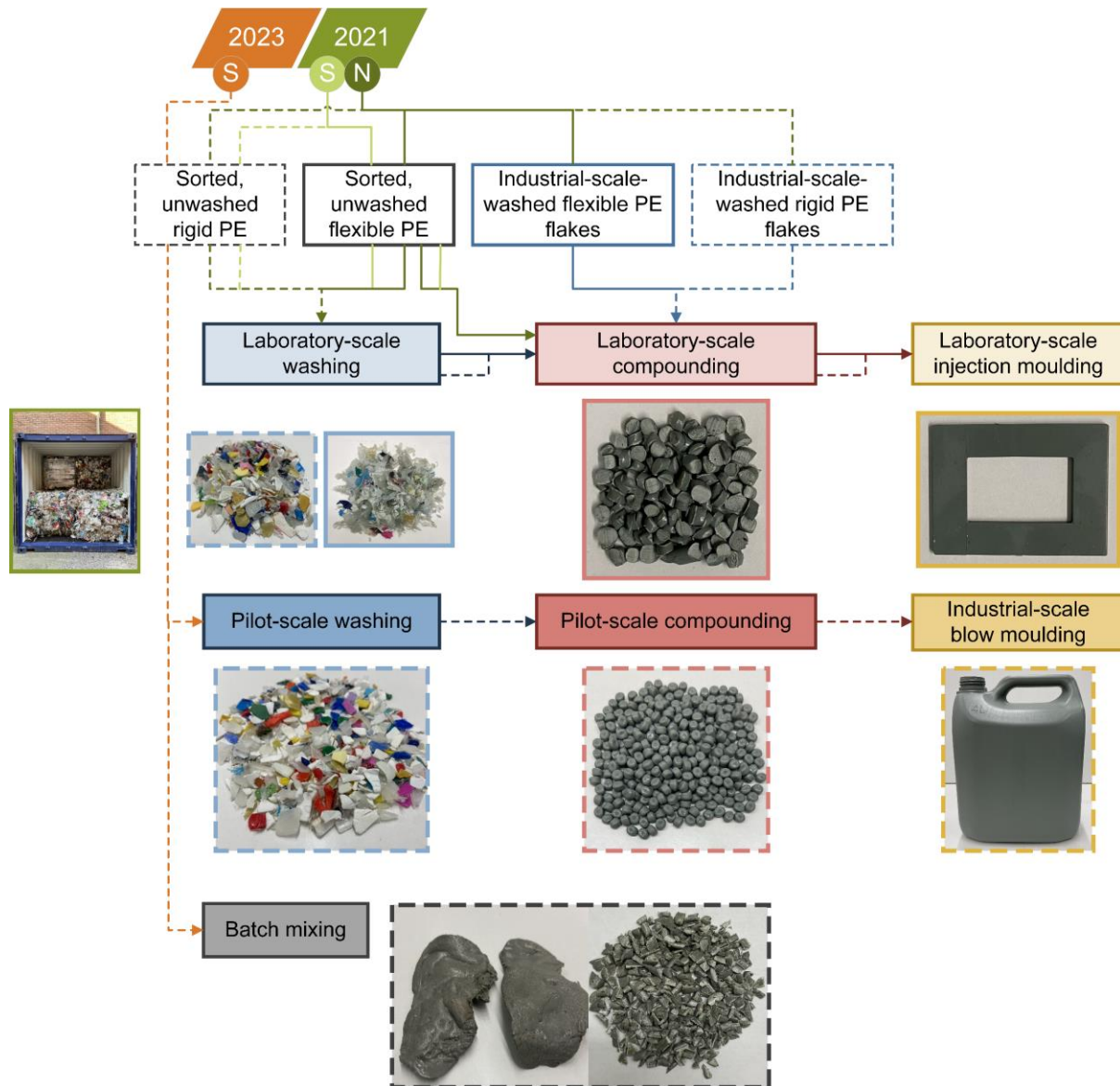


Figure 6. Schematic representation of materials, processing and examples of produced samples.

Table 1. Sample processing history with given sample codes.

| Type | Washing parameters | | | Compounding parameters | | | Sample code |
|----------------------------------|--------------------|-------------------|------------------|--------------------------|--------------------------|-----------------------|------------------------------|
| | Washing scale | Washing medium | Temperature (°C) | Screw design | Temperature profile (°C) | Degassing | |
| Flexible PE, Sweden, 2021 | Unwashed | N/A | N/A | SC1 | Flexible LT | Atmospheric | S-F_UW_SC1_200 |
| | Unwashed | N/A | N/A | SC1 | Flexible HT | Atmospheric | S-F_UW_SC1_240 |
| | Unwashed | N/A | N/A | SC2 | Flexible LT | Atmospheric | S-F_UW_SC2_200 |
| | Unwashed | N/A | N/A | SC2 | Flexible HT | Atmospheric | S-F_UW_SC2_240 |
| | Laboratory | NaOH | 40 | SC2 | Flexible LT | Atmospheric | S-F_LW-NaOH40_SC2_200 |
| | Laboratory | NaOH | 40 | SC2 | Flexible HT | Atmospheric | S-F_LW-NaOH40_SC2_240 |
| Flexible PE, Norway, 2021 | Unwashed | N/A | N/A | SC1 | Flexible LT | Atmospheric | N-F_UW_SC1_200 |
| | Unwashed | N/A | N/A | SC1 | Flexible HT | Atmospheric | N-F_UW_SC1_240 |
| | Unwashed | N/A | N/A | SC2 | Flexible LT | Atmospheric | N-F_UW_SC2_200 |
| | Unwashed | N/A | N/A | SC2 | Flexible HT | Atmospheric | N-F_UW_SC2_240 |
| | Laboratory | NaOH | 40 | SC2 | Flexible LT | Atmospheric | N-F_LW-NaOH40_SC2_200 |
| | Laboratory | NaOH | 40 | SC2 | Flexible HT | Atmospheric | N-F_LW-NaOH40_SC2_240 |
| | Laboratory | NaOH | 25 | SC2 | Flexible HT | Atmospheric | N-F_LW-NaOH25_SC2_240 |
| | Laboratory | D | 40 | SC2 | Flexible HT | Atmospheric | N-F_LW-D40_SC2_240 |
| | Laboratory | D | 25 | SC2 | Flexible HT | Atmospheric | N-F_LW-D25_SC2_240 |
| | Laboratory | W | 40 | SC2 | Flexible HT | Atmospheric | N-F_LW-W40_SC2_240 |
| | Laboratory | W | 25 | SC2 | Flexible HT | Atmospheric | N-F_LW-W25_SC2_240 |
| | Industrial | NaOH, D, other | 70-80 | SC1 | Flexible LT | Atmospheric | N-F_IW_SC1_200 |
| | Industrial | NaOH, D, other | 70-80 | SC1 | Flexible HT | Atmospheric | N-F_IW_SC1_240 |
| | Industrial | NaOH, D, other | 70-80 | SC2 | Flexible LT | Atmospheric | N-F_IW_SC2_200 |
| Industrial | NaOH, D, other | 70-80 | SC2 | Flexible HT | Atmospheric | N-F_IW_SC2_240 | |
| Rigid PE, Sweden, 2021 | Laboratory | W | 25 | SC2 | Rigid LT | Atmospheric | S-R_LW-W25_SC2_200 |
| | Laboratory | W | 25 | SC2 | Rigid MT | Atmospheric | S-R_LW-W25_SC2_240 |
| | Laboratory | D | 25 | SC2 | Rigid MT | Atmospheric | S-R_LW-D25_SC2_240 |
| | Laboratory | W | 25 | SC2 | Rigid HT | Atmospheric | S-R_LW-W25_SC2_260 |
| Rigid PE, Norway, 2021 | Laboratory | W | 25 | SC2 | Rigid LT | Atmospheric | N-R_LW-W25_SC2_200 |
| | Laboratory | W | 25 | SC2 | Rigid MT | Atmospheric | N-R_LW-W25_SC2_240 |
| | Laboratory | D | 25 | SC2 | Rigid MT | Atmospheric | N-R_LW-D25_SC2_240 |
| | Laboratory | W | 25 | SC2 | Rigid HT | Atmospheric | N-R_LW-W25_SC2_260 |
| | Industrial | NaOH, D, other | 70-80 | SC2 | Rigid LT | Atmospheric | N-R_IW_SC2_200 |
| | Industrial | NaOH, D, other | 70-80 | SC2 | Rigid MT | Atmospheric | N-R_IW_SC2_240 |
| | Industrial | NaOH, D, other | 70-80 | SC2 | Rigid HT | Atmospheric | N-R_IW_SC2_260 |
| Rigid PE, Sweden, 2023 | Pilot | D, defoamer | 40 | N/A | MT | Vacuum | S-R_PW-D40_Vac |
| | Pilot | NaOH, D, defoamer | 40 | N/A | MT | Vacuum | S-R_PW-NaOH40_Vac |
| | Pilot | NaOH, D, defoamer | 40 | N/A | MT | Atmospheric | S-R_PW-NaOH40_NoVac |
| | Pilot | NaOH, D, defoamer | 80 | N/A | MT | Vacuum | S-R_PW-NaOH80_Vac |
| | Pilot | NaOH, D, defoamer | 80 | N/A | MT | Atmospheric | S-R_PW-NaOH80_NoVac |
| | Pilot | NaOH, D, defoamer | 80 | with water stripping, MT | | Vacuum | S-R_PW-NaOH80_W+Vac |

Abbreviations: PE: polyethylene, NaOH: sodium hydroxide, D: detergent, W: water, LT: low-temperature, HT: high-temperature, MT: medium-temperature

The virgin grade HDPE is henceforth referred as VG-HDPE and the batch-mixed unwashed rigid PE waste as S-R_UW.

4 CHARACTERIZATION

4.1 THERMAL PROPERTIES AND MOLECULAR STRUCTURE

The thermal transitions were assessed by differential scanning calorimetry (DSC) using a Mettler-Toledo DSC 2 or 5+. The samples were prepared according to ISO 11357-1:2016 and the measurements were made in a nitrogen atmosphere with a flow rate of 50 ml/min. The heating and cooling rates were 10 °C/min. Duplicate tests were made and the average results for the first heating cycles were reported. For the measurement of the heat of fusion (ΔH), the baseline was taken from 60 to 134 °C for the flexible PE and from 65 to 144 °C for the rigid PE.

The ash content was determined by thermogravimetric analysis (TGA) using a Mettler-Toledo TGA/DSC 3+. A sample weighing ca. 3 mg, 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 for each type of material, and the ash content values were determined at 550 °C for the flexible PE and at 600 °C for the rigid PE samples.

The weight average molecular mass (M_w) and the polydispersity (PD) were determined with high temperature gel permeation chromatography (HT-GPC) using a Polymer Laboratories GPC220 instrument with PlOlexis and PlOlexis guard columns with lengths of 3x30 cm at 160 °C. About 30-40 mg of material were dissolved in 10 ml 1,2,4 trichlorobenzene with 200 ppm butylated hydroxytoluene as antioxidant. The injection volume was 200 μ l and the flow rate was 0.8 ml/min. The results shown are based on two independent measurements except for the N-F_LW-NaOH40_SC2_240 and S-R_LW-D25_SC2_240 samples.

4.2 THERMO-OXIDATIVE STABILITY

The oxidation-induction-temperature (T_{ox}) was assessed using a Mettler-Toledo DSC 2 or 5+ instrument. The samples were prepared according to ISO 11357-6:2018 and the measurements were made in air with a heating rate of 10 °C/min. Duplicate measurements were made and the average values are reported.

The oxidation-induction-time (OIT) for the rigid PE samples washed and compounded on a pilot scale was measured by DSC using Mettler-Toledo DSC 5+ and by time-sweep rheometry using an Anton Paar MCR702 rotational rheometer. The measurements were made at 200 °C and 240 °C in both cases. For the DSC measurements, the samples were prepared according to ISO 11357-6:2018 where they were first heated to the measurement temperature in a nitrogen atmosphere at a rate of 20 °C/min, and then allowed to rest for 3 min before the atmosphere was changed to air. The gas flow rate was 50 ml/min in all cases. For the time-sweep rheometry, a 25 mm parallel plate geometry was used, and the materials were therefore prepared as 25 mm discs using an Xplore Micro Injection Moulder IM12. The measurements were made with a shear strain of 1 %, a frequency of 0.159 Hz and a gap of 1 mm. The time between the loading

of the sample and the beginning of the measurements, including the melting, compressing and trimming of the sample, was 9 min. Duplicate measurements were made and the average values are reported.

The values of T_{ox} and OIT measured using DSC were retrieved from the intercept point determined by the tangent method described in ISO 11357-6:2018, and the same was applied for the time-sweep rheometry using the plot of storage modulus (G') versus time. The steepest linear slope observed in the exotherm (DSC) or G' (time-sweep rheometry) was extrapolated by drawing a tangent, and its intercept with the extended baseline was taken as T_{ox} and OIT .

4.3 RHEOLOGICAL PROPERTIES

The melt mass-flow rate (MFR) was determined using a Ceast Modular Melt Flow instrument. Flexible PE samples were measured at 190 °C and rigid PE samples at 240 °C, both with a weight of 2.16 kg in accordance with ISO 1133-1:2011. The moderate processing temperature used in this study, 240 °C, was chosen to determine the MFR of the rigid PE samples because 190 °C was not sufficient to get the required minimum mass described in the standard.

The melt viscosity was evaluated using a Göttfert high-pressure Rheograph 20 capillary rheometer. The measurements were made at a constant piston speed at shear rates from 10^3 to 10^0 s⁻¹. The measurement temperature was 220 °C for flexible PE, 240 °C for laboratory-scale-compounded rigid PE and 200 °C for pilot-scale washed and compounded rigid PE samples. Three dies were used with a diameter (D) of 2 mm and aspect ratios (L/D) of 5, 10 and 15. The Bagley and the Weissenberg-Rabinowitsch shear-rate corrections (ISO 11443:2021) were applied and the graphs of corrected viscosity versus shear rate and of the entrance pressure losses based on the Bagley plots were given for the die with a L/D ratio of 10.

The melt strength and drawability were assessed using a haul-off unit, consisting of a strand wheel connected to a force transducer and a take-off wheel, coupled with the Göttfert high-pressure Rheograph 20 capillary rheometer. ISO 16790:2021-02 was used as a guideline for the measurements made using a capillary with a diameter of 2 mm and a length of 20 mm in an ambient environment of 23 ± 1 °C and 25 ± 5 % RH. For the flexible PE samples, the measurements were made at 220 °C, the initial velocity (v_0) of the extruded strand at the exit of the capillary being constant at 11.25 mm/s and the starting tangential velocity (v_I) of the take-off wheel being 17.5 mm/s for the unwashed and laboratory-scale-washed samples, but 25 mm/s for the industrial-scale-washed samples. The speed of the take-off wheel was increased at a rate of 1.2 mm/s². For the rigid PE samples washed and compounded on a laboratory scale, the measurements were made at 240 °C with both v_0 and v_I being 13.5 mm/s, where the former was kept constant and the latter was increased at a rate of 0.24 mm/s². For the rigid PE samples washed and compounded on a pilot scale, the measurements were made at 200 °C with v_0 constant at 13.5 mm/s and v_I being the same for all the samples except for the unwashed materials for which it was set to 9.9 mm/s. The speed of the take-off wheel was

increased at a rate of 1.2 mm/s². The force required to draw the melt was recorded together with the velocity of the take-off wheel and the time until the extended strand broke. The strain (ε) was calculated as $\varepsilon = (v_I - v_0)/v_0$.¹¹⁹ The mean values of the melt strength (force-at-break) and of the drawability (strain-at-break) were based on at least five independent measurements.

Dynamic-oscillatory shear flow measurements were made using an Anton Paar MCR702 rheometer with a 25 mm parallel plate geometry. The measurement gap was set to 1 mm and measurements were made in nitrogen at 180 °C for rigid PE samples washed and compounded on a laboratory scale and at 200 °C for rigid PE samples washed and compounded on a pilot scale. The linear viscoelastic (LVE) region for each sample was first determined with oscillatory strain sweep tests performed at a constant frequency of 1 Hz in the shear strain range of 0.001-100 %. Frequency sweep tests were then carried out at a constant shear strain of 1 % within the LVE region over a frequency range from 100 to 0.0016 Hz. The storage modulus (G'), the loss modulus (G''), the complex modulus (G^*), the complex viscosity (η^*) and the phase angle (δ) were recorded and averages of duplicate measurements were reported.

4.4 MECHANICAL PROPERTIES

Mechanical properties were measured using a Zwick/Roell Z2.5 instrument equipped with a 2 kN load cell. For the flexible PE and rigid PE samples injection moulded on a laboratory scale, test bars of type 5A in ISO 527-2:2012 were cut from the three different regions of the moulded frame. The thickness of the specimens was ca. 2 mm. For the rigid PE samples blow-moulded on an industrial scale, test bars of type 5A in ISO 527-2:2012 were cut from three different 4L-containers for each type of material, four specimens being taken in the longitudinal direction, four in the transverse direction and five from the bottom of the container including a weld-line from each 4L-container. The thickness of the specimens was ca. 1.5 mm in the weld line regions and ca. 1.2 mm in the longitudinal and transverse directions. The batch-mixed unwashed material of the same rigid PE waste was injection-moulded into tensile bars of type 1BA in ISO 527-2:2012 using Xplore Micro Injection Moulder IM12, the thickness being 2 mm. All the samples were conditioned according to ISO 527-1:2012 before the measurements were made at a strain rate of 1 min⁻¹ in an ambient environment of 23 ± 1 °C and 33 ± 5 % RH. The Young's modulus, the stress- and strain-at-yield, and the stress- and strain-at-break were determined and averages of at least five independent measurements were reported.

4.5 COLOUR AND GLOSS

The colour and gloss of the blow-moulded 4L-containers and batch-mixed unwashed rigid PE material were measured using a Konica Minolta CM-36dG spectrophotometer with an integrated ISO-2813-compliant 60° gloss sensor. The results were obtained assuming D65/10° illuminant and observer conditions, and the illumination and measurement diameters were 11 and 8 mm, respectively. The illumination was diffuse with a viewing angle of 8° and the wavelength range was 360-740 nm with a wavelength pitch of 10 nm. The colour was measured in both specular-component-included (SCI) and -excluded (SCE) modes, and the gloss measurements were made with an incidence angle of 60°. The SCE depends on the surface conditions, whereas the SCI gives the total colour appearance independent of the surface conditions. The colour was expressed in terms of the CIELAB $L^*a^*b^*$ system. For the VG-HDPE samples, the measurements were made both with the sample holder as background (black) and with a white paper background, but for the other non-translucent materials the sample holder background was used alone. Five pieces of the batch-mixed unwashed material were used and three measurements were made on each piece. Three different 4L-containers were used and six measurements were made on each container. The average L^* , a^* , b^* and gloss values were reported.

5 MAIN RESULTS AND DISCUSSION

5.1 FEEDSTOCK CHARACTERISTICS

The distribution of flexible PE, rigid PE, PP, and the total of other polymeric (PET, PS, PVC, etc.) and non-polymeric (paper, textile, etc.) “other” content are shown in Figure 7, for sorted flexible PE and rigid PE waste, where the latter also includes items which could not be identified with NIR due to their dark colour.

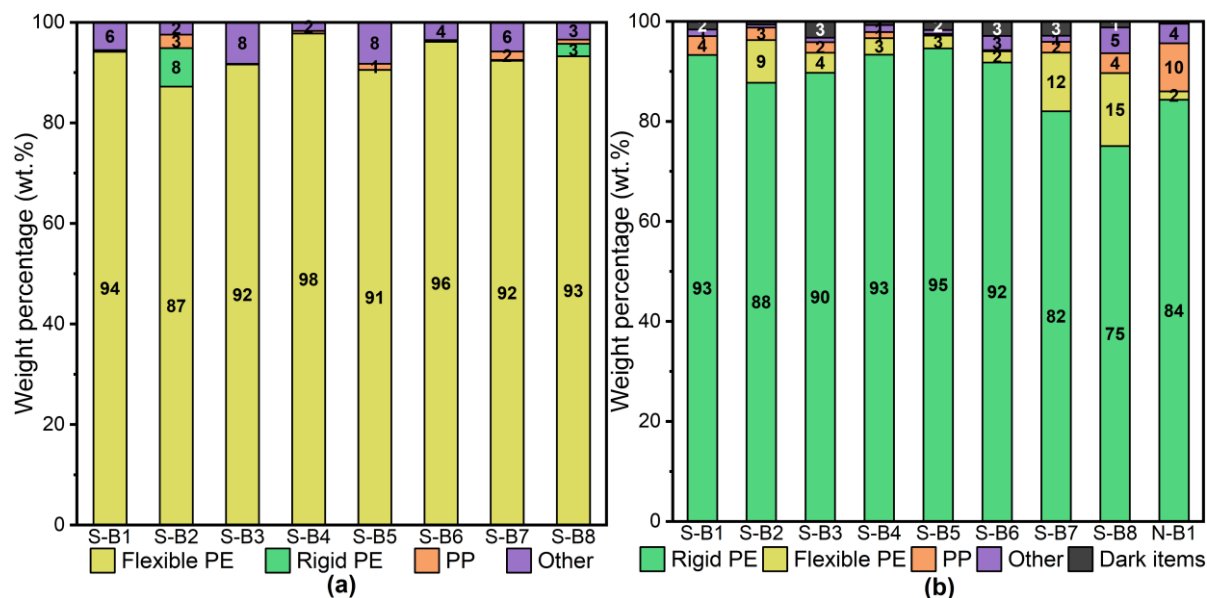


Figure 7. Feedstock distribution of polymeric and non-polymeric content given as mass percentages in each sample batch: (a) flexible PE stream, (b) rigid PE stream.

The average total PE content (flexible and rigid PE) in sorted flexible PE received from the facility in Sweden was 94 wt.% with a 95 % confidence limit of ± 2 wt.% based on eight batches. The PP was 1 ± 0.7 wt.% and other was 5 ± 2.1 wt.%. In the sorted rigid PE received from the facility in Sweden, the average total PE content (flexible and rigid PE) was 94 wt.% with a 95 % confidence limit of ± 2 wt.%. The rest was 2 ± 1.2 wt.% PP, 1.8 ± 1.3 wt.% other and 1.8 ± 0.9 wt.% dark material. The batch of sorted rigid PE received from the facility in Norway had an average PE content of 86 wt.%, the rest being 9.6 wt.% PP, 3.9 wt.% other and 0.4 wt.% dark material. Similar degrees of contamination by PP in both flexible and rigid PE streams have been also reported in other studies where it was pointed out that this was mainly due to product design, where two or more polymers were used in a single product.^{33,120} In our study, especially in the rigid PE stream, the PP originated mainly from a PP cap on a PE bottle.

5.2 INFLUENCE OF WASHING AND WASHING CONDITIONS

Washing is an important step in the recycling process, to remove contaminants that may create potential health risks and to minimize particles that may be harmful during the subsequent compounding and shaping processes. In this study, the scope of the washing was limited to understanding better the influence of the washing conditions, washing medium and temperature, on the functional properties of the recycled materials. Therefore, no analysis was made of the washing water itself although visual observations indicated that the contaminants were transferred to the washing liquid, which appeared to be coloured and opaque. This was also indicated by the weight losses during a laboratory-scale washing which were ca. 45 wt.% for the flexible PE stream and ca. 25 wt.% for the rigid PE stream. The weight loss of the rigid PE stream during washing on a pilot scale was somewhat lower at ca. 5 wt.%, but this might be a result of more efficient procedure with an automated separation technique rather than the manual collection of the float fraction during laboratory-scale washing. The surface contamination of the unwashed flakes of flexible PE material, Figure 8a, were apparently eliminated after washing and the washed flakes were much brighter as shown in Figure 8b. The colour of the compounded pellets and injection-moulded samples of the washed flakes were visually lighter and more green than the dark grey unwashed material, Figure 8. The same was evident in the case of the rigid PE samples, as batch-mixed unwashed material visually looked darker and more grey than the washed and processed material, as shown in Figure 6. This is further discussed in section 5.6.



Figure 8. From top to bottom: the flakes, pellets and IM-sample of (a) the unwashed flexible PE material, (b) the laboratory-scale-washed flexible PE material.

The first heating endotherms of the unwashed and washed recycled materials from flexible PE and rigid PE waste are shown in Figures 9a and b, respectively.

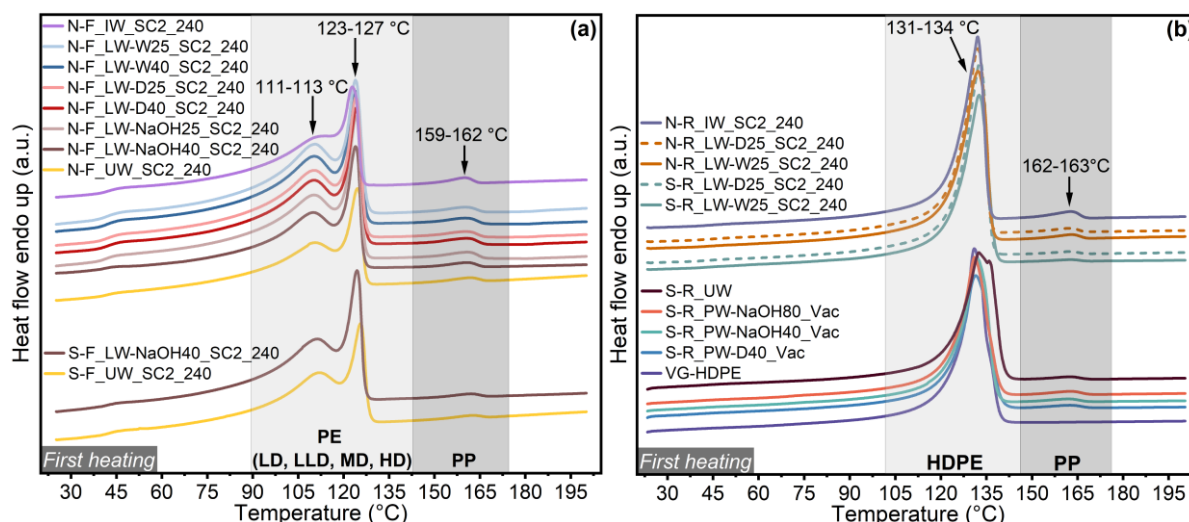


Figure 9. The first heating endotherms of selected unwashed and washed recycled pellets: (a) flexible PE samples, (b) rigid PE samples.

In general, the recycled flexible PE sample group and the recycled rigid PE sample group showed similar melting endotherms within each group. The flexible PE samples had a main peak at 123-127 °C with a shoulder at 111-113 °C and a small peak at 159-162 °C, as shown in Figure 9a. The main peak and the shoulder were associated with different grades of PE (LD, LLD, MD and HD) and the third small peak with PP.^{121,122} The rigid PE samples had a main peak at 131-134 °C which was related mainly to HDPE and a small peak at 162-163 °C associated with PP, as shown in Figure 9b.^{121,122} In none of the groups of recycled materials, did the washing conditions influence the melting temperature. The observed PP peaks had slightly higher melting enthalpies for the samples from the facility in Norway than from the facility in Sweden, in the case of both the flexible PE and the rigid PE samples, which supported the results of the feedstock characterization.

Even though the shape of the melting endotherms was not influenced by washing, the melting enthalpies (ΔH) of the flexible PE samples, Table 2, increased slightly after both laboratory-scale and industrial-scale washing, suggesting that the degree of crystallization increased and/or that the content of impurities was reduced by washing¹²³, and possibly also that some degradation occurred during washing, as a greater mobility of small polymer chains may lead a higher degree of crystallinity.⁶⁹ The laboratory-scale-washed flexible PE materials showed higher melting enthalpies than those washed on an industrial scale, but this was not observed in the rigid PE materials. The ΔH values of the rigid PE samples washed on a laboratory scale and of those washed on an industrial scale were very similar, as shown in Table 2. The pilot-scale-washed rigid PE samples showed no significant difference in ΔH values from the unwashed material, unlike the flexible PE samples. In general, neither the washing medium nor the washing temperature influenced the melting enthalpies of the recycled materials, except for the flexible PE samples washed with NaOH at 40 °C which had the lowest values among the laboratory-scale-washed samples from the facility in Norway.

Table 2. The melting enthalpy, ash content, weight-average molecular mass and polydispersity of the samples.

| Sample | ΔH (J/g) | Ash content (%) | M_w (kDa) | PD | Sample | ΔH (J/g) | Ash content (%) | M_w (kDa) | PD |
|-----------------------|---------------------|-----------------------|----------------|------|---------------------|---------------------|-----------------------|----------------|------|
| S-F_UW_SC1_200 | 76 | 5.4 | N/A | N/A | S-R_LW-W25_SC2_200 | 182 | 1.7 | 133 | 6.5 |
| S-F_UW_SC2_200 | 78 | 4.8 | N/A | N/A | S-R_LW-W25_SC2_240 | 176 | 2.3 | 121 | 6.1 |
| S-F_UW_SC1_240 | 76 | 5.2 | N/A | N/A | S-R_LW-D25_SC2_240 | 184 | 2.2 | 75 | 7.0 |
| S-F_UW_SC2_240 | 77 | 4.8 | 115 | 4.5 | S-R_LW-W25_SC2_260 | 175 | 2.4 | 92 | 5.8 |
| S-F_LW-NaOH40_SC2_200 | 86 | 3.5 | N/A | N/A | N-R_LW-W25_SC2_200 | 175 | 2.7 | 93 | 6.8 |
| S-F_LW-NaOH40_SC2_240 | 94 | 3.3 | 114 | 4.6 | N-R_LW-W25_SC2_240 | 177 | 1.8 | 132 | 7.1 |
| N-F_UW_SC1_200 | 66 | 11.5 | 123 | 4.8 | N-R_LW-D25_SC2_240 | 176 | 2.6 | 132 | 8.6 |
| N-F_UW_SC2_200 | 68 | 11.3 | 124 | 5.2 | N-R_LW-W25_SC2_260 | 174 | 2.4 | 74 | 5.6 |
| N-F_UW_SC1_240 | 67 | 10.5 | N/A | N/A | N-R_IW_SC2_200 | 174 | 2.4 | 106 | 6.7 |
| N-F_UW_SC2_240 | 69 | 11.2 | 122 | 5.0 | N-R_IW_SC2_240 | 169 | 2.3 | 123 | 6.9 |
| N-F_LW-NaOH40_SC2_200 | 83 | 7.2 | 95 | 4.8 | N-R_IW_SC2_260 | 169 | 2.1 | 84 | 7.0 |
| N-F_LW-NaOH40_SC2_240 | 85 | 7.2 | 112 | 5.8 | S-R_PW-D40_Vac | 160 | 1.8 | N/A | N/A |
| N-F_LW-NaOH25_SC2_240 | 99 | 8.0 | 95 | 3.3 | S-R_PW-NaOH40_Vac | 162 | 2.0 | N/A | N/A |
| N-F_LW-D40_SC2_240 | 98 | 7.5 | 99 | 4.5 | S-R_PW-NaOH40_NoVac | 165 | 1.9 | N/A | N/A |
| N-F_LW-D25_SC2_240 | 103 | 6.6 | 92 | 4.3 | S-R_PW-NaOH80_Vac | 158 | 1.8 | N/A | N/A |
| N-F_LW-W40_SC2_240 | 102 | 6.9 | 96 | 4.6 | S-R_PW-NaOH80_NoVac | 158 | 1.6 | N/A | N/A |
| N-F_LW-W25_SC2_240 | 105 | 7.3 | 101 | 3.6 | S-R_PW-NaOH80_W+Vac | 158 | 1.4 | N/A | N/A |
| N-F_IW_SC1_200 | 76 | 5.2 | N/A | N/A | S-R_UW | 163 | 2.2 | N/A | N/A |
| N-F_IW_SC2_200 | 76 | 5.3 | 87 | 4.1 | | | | | |
| N-F_IW_SC1_240 | 76 | 5.1 | N/A | N/A | | | | | |
| N-F_IW_SC2_240 | 76 | 5.3 | 71 | 4.4 | VG-HDPE | 181 | 1.0 | N/A | N/A |

The ash content, Table 2, was reduced after the washing, especially in the flexible PE materials but not in the rigid PE materials. The decrease in ash content after washing presumably related to a reduction in the content of impurities.^{11,14,124} The industrial-scale washing gave materials with lower ash contents than the laboratory-scale washing for the flexible PE samples, which suggests that this may be a more advanced cleaning procedure. This was not, however, the case for the rigid PE samples, where both the industrial-scale- and laboratory-scale-washed samples had similar ash contents. Neither the washing medium nor the washing temperature had a significant influence on the ash contents of the recycled flexible and rigid PE samples.

In general, both the weight-average molecular mass (M_w) and polydispersity (PD) of the flexible PE samples, Table 2, were lower after washing, suggesting that chain scission might dominate during the degradation.^{69,78,125,126} The M_w was much lower after the industrial-scale washing, probably because the more intensive washing procedure at high temperature resulted in greater degradation than the laboratory-scale washing. In the case of the industrial-scale-washed rigid PE samples, the M_w was either higher or lower than that of the material washed on a laboratory scale. The washing conditions on the laboratory scale, had no great influence, except that the flexible PE sample from the facility in Norway washed with NaOH at 40 °C had the highest M_w whereas the rigid PE sample from the facility in Sweden washed with detergent had a much lower M_w than the water-washed material, but the results for these two samples were reported as a single measurement, and this may be the cause of these differences.

The oxidation-induction-temperatures (T_{ox}) for the recycled flexible and rigid PE samples and the oxidation-induction-times (OIT) for the rigid PE samples washed and compounded on a pilot scale are given in Table 3.

Table 3. The oxidation-induction-temperatures (T_{ox}) and -times (OIT) of the samples.

| Sample | T_{ox} (°C) | Sample | T_{ox} (°C) | OIT , DSC (min) | | OIT , time-sweep (min) | |
|-----------------------|---------------|---------------------|---------------|-------------------|--------|--------------------------|--------|
| | | | | 200 °C | 240 °C | 200 °C | 240 °C |
| S-F_UW_SC1_200 | 222 | S-R_LW-W25_SC2_200 | 215 | N/A | N/A | N/A | N/A |
| S-F_UW_SC2_200 | 224 | S-R_LW-W25_SC2_240 | 209 | N/A | N/A | N/A | N/A |
| S-F_UW_SC1_240 | 216 | S-R_LW-D25_SC2_240 | 222 | N/A | N/A | N/A | N/A |
| S-F_UW_SC2_240 | 216 | S-R_LW-W25_SC2_260 | 202 | N/A | N/A | N/A | N/A |
| S-F_LW-NaOH40_SC2_200 | 211 | N-R_LW-W25_SC2_200 | 215 | N/A | N/A | N/A | N/A |
| S-F_LW-NaOH40_SC2_240 | 210 | N-R_LW-W25_SC2_240 | 213 | N/A | N/A | N/A | N/A |
| N-F_UW_SC1_200 | 224 | N-R_LW-D25_SC2_240 | 218 | N/A | N/A | N/A | N/A |
| N-F_UW_SC2_200 | 232 | N-R_LW-W25_SC2_260 | 202 | N/A | N/A | N/A | N/A |
| N-F_UW_SC1_240 | 226 | N-R_IW_SC2_200 | 229 | N/A | N/A | N/A | N/A |
| N-F_UW_SC2_240 | 231 | N-R_IW_SC2_240 | 226 | N/A | N/A | N/A | N/A |
| N-F_LW-NaOH40_SC2_200 | 232 | N-R_IW_SC2_260 | 218 | N/A | N/A | N/A | N/A |
| N-F_LW-NaOH40_SC2_240 | 231 | S-R_PW-D40_Vac | 220 | 5 | 0.7 | 15 | 7 |
| N-F_LW-NaOH25_SC2_240 | 229 | S-R_PW-NaOH40_Vac | 231 | 12 | 1.0 | 25 | 3 |
| N-F_LW-D40_SC2_240 | 227 | S-R_PW-NaOH40_NoVac | 232 | 11 | 1.0 | 26 | 3 |
| N-F_LW-D25_SC2_240 | 230 | S-R_PW-NaOH80_Vac | 233 | 13 | 1.2 | 28 | 4 |
| N-F_LW-W40_SC2_240 | 225 | S-R_PW-NaOH80_NoVac | 232 | 13 | 1.0 | 27 | 4 |
| N-F_LW-W25_SC2_240 | 227 | S-R_PW-NaOH80_W+Vac | 234 | 14 | 1.2 | 28 | 4 |
| N-F_IW_SC1_200 | 192 | S-R_UW | 236 | 18 | 1.4 | 30 | 4 |
| N-F_IW_SC2_200 | 195 | | | | | | |
| N-F_IW_SC1_240 | 180 | VG-HDPE | 250 | 210 | 3.3 | 118 | 6 |
| N-F_IW_SC2_240 | 184 | | | | | | |

In general, the washed samples had a lower T_{ox} than the unwashed samples, in the case of both the flexible and the rigid PE materials. For the flexible PE materials, the industrial-scale washing resulted in much lower T_{ox} values than laboratory-scale washing, but the opposite was observed for the rigid PE materials. Neither during the laboratory-scale washing of flexible PE materials nor during the pilot-scale washing of the rigid PE materials did the washing temperature have any great influence on the T_{ox} value, but the washing medium did have an effect. The laboratory-scale-washed flexible PE samples washed with water alone had a lower T_{ox} than both the detergent- and NaOH-washed samples. In the case of the rigid PE samples, the laboratory-scale washing with detergent resulted in a higher T_{ox} than washing with water alone, but the pilot-scale washing with detergent resulted in a lower T_{ox} than washing with a combination of detergent and NaOH. This difference in T_{ox} after washing with different washing media may perhaps be due to different levels of removal of residual chemicals, stabilizers and oxidative moieties during washing.^{29,64,127,128}

All the unwashed and laboratory-scale-washed flexible PE samples had a T_{ox} of at least 210 °C, which suggests that active stabilizers remained in the samples, but after intensive washing at a high temperature as in the case of industrial-scale washing there was a significant loss or inactivation of stabilizers, and the T_{ox} dropped to the value for unstabilized virgin PE, which

was reported to be 180 ± 5 °C.¹²⁹ With some exceptions, all the rigid PE samples had a T_{ox} above that of unstabilized PE, which was reported to be 210 °C in one study¹³⁰ and 180 ± 5 °C in another.¹²⁹

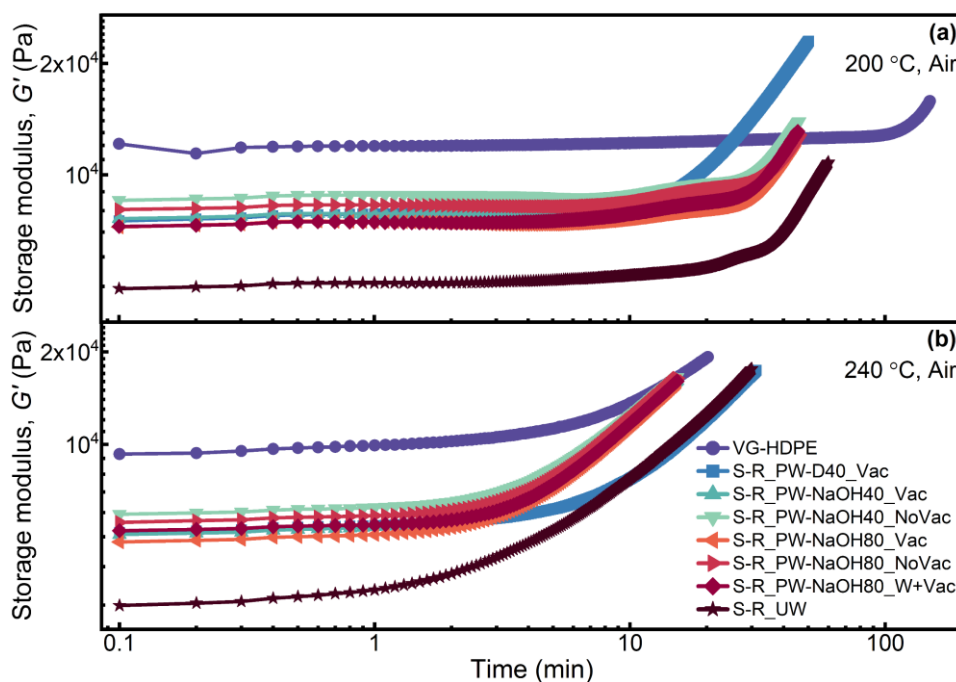


Figure 10. The storage moduli versus time in air at (a) 200 °C and (b) 240 °C for the pilot-scale washed and compounded rigid PE samples.

The thermo-oxidative stability of the rigid PE samples washed and compounded on a pilot scale was also characterized by the oxidation induction time (OIT) determined by both time-sweep rheometry and DSC-measurements. The results are shown in Table 3, where the time-sweep rheometry results were obtained from the plots of storage modulus in air versus time shown in Figure 10. The OIT -values obtained by DSC measurements were much shorter than those obtained by time-sweep rheometry, but the pattern was the same. The OIT -values followed the same pattern as the T_{ox} -temperatures. The washed materials had, in general, lower OIT -values than the unwashed sample. The washing temperature had no great influence on the OIT -values whereas the material washed without NaOH had, in general, a lower OIT -value than those washed with NaOH, but the reason for this is not clear. In all cases, the stability was poorer at 240 °C than at 200 °C. Poh et al.¹⁰² have suggested that the two-step increase in storage modulus of the recycled rigid PE samples and UW-material at 200 °C is due to chain splitting followed by chain branching, whereas Dordinejad et al.¹⁰⁰ consider the behaviour to be due to active stabilizers in the materials, but the present study does not permit conclusions to be drawn.

The melt viscosity and entrance pressure losses in capillary flow are given as a function of shear rate in Figure 11. The laboratory-scale washing had no significant influence on the viscosity of the flexible PE materials, but the industrial-scale-washed samples showed much lower viscosities than both the unwashed and the laboratory-scale-washed samples, Figure 11a.

In the case of the rigid PE materials, the industrial-scale-washed samples had a viscosity in a range similar to that of the laboratory-scale-washed material, Figure 11b, and the pilot-scale-washed samples had a viscosity similar to that of the unwashed material, Figure 11c. Neither the washing medium nor the washing temperature had any great effect on the viscosity of the recycled flexible and rigid PE materials.

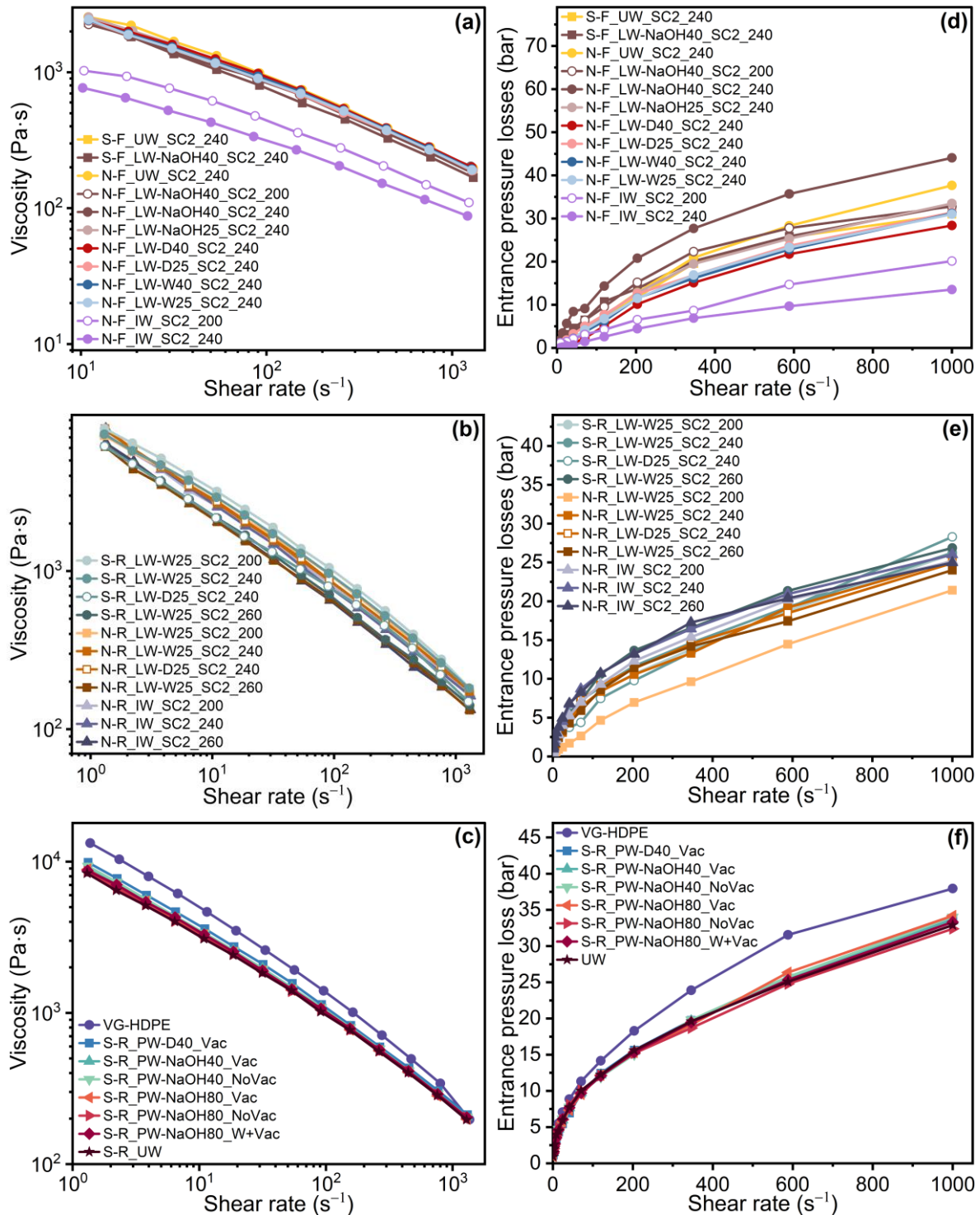


Figure 11. Viscosity versus shear rate and entrance pressure loss versus shear rate: (a), (d) flexible PE samples, (b), (e) laboratory-scale-compounded rigid PE samples, (c), (f) pilot-scale-compounded rigid PE samples.

The melt mass-flow rate (*MFR*) of the samples followed a pattern similar to that of the viscosity curves. The *MFR* of the unwashed and laboratory-scale-washed flexible PE samples varied between 0.5 and 0.8 g/10min without any major influence of the washing conditions on a laboratory scale. However, industrial-scale washing at a high temperature resulted in a significant increase in the *MFR* to between 1.9 and 4.8 g/10min. This supported the lower values of M_w , T_{ox} and viscosities observed for the industrial-scale-washed samples, but this significant difference was not observed between the *MFR* of the rigid PE materials washed on a laboratory scale (0.5-1.3 g/10min) and those washed on an industrial scale (0.4-0.7 g/10min). However, as with the flexible PE samples, the rigid PE samples washed with a detergent on a laboratory scale had almost the same *MFR* as those washed with water alone, being between 0.5 and 0.7 g/10min.

The entrance pressure loss in capillary flow can be indirectly used as an indication of melt elasticity where a higher pressure loss is associated with a higher melt elasticity.¹⁰⁷ The entrance pressure losses of the flexible PE samples were lower, with one exception, after both the laboratory-scale and industrial-scale washing, the latter being more prominent, Figure 11d. The sample from the facility in Norway washed with NaOH at 40 °C exhibited the greatest entrance pressure loss, but the rest of the laboratory-scale-washed samples showed similar values to each other. The industrial-scale-washed samples had the lowest entrance pressure loss, and thus the lowest melt elasticity. For the rigid PE materials, in contrast, the industrial-scale-washed samples showed slightly higher entrance pressure loss values than the laboratory-scale-washed samples, Figure 11e, although there was no significant influence of the washing medium or temperature, Figures 11e and f. Pilot-scale-washed rigid PE samples showed entrance pressure losses similar to that of the unwashed material, Figure 11f, but there were no substantial differences as with the flexible PE material.

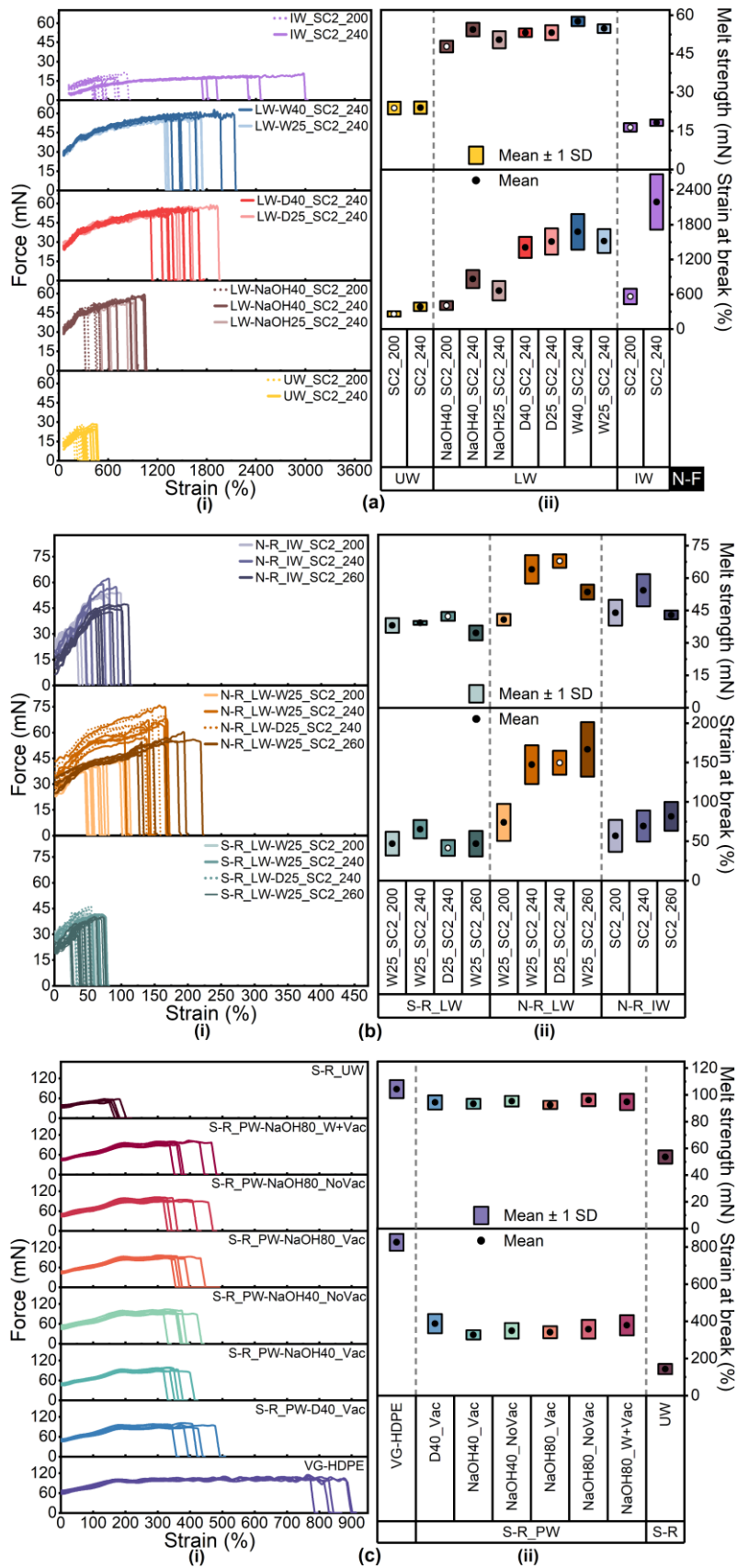


Figure 12. Melt characteristics of the (a) laboratory-scale-processed flexible PE samples, (b) laboratory-scale-processed rigid PE samples, (c) pilot-scale-processed rigid PE samples: (i) force-strain curves of the melts, (ii) melt strength and strain-at-break of the melts; where a dot in a box indicates the average value and the standard deviation is indicated by the height of the box.

The drawability and strength of the melts are shown in Figure 12. For the flexible PE samples, Figure 12a, the melt drawability increased with both laboratory-scale and industrial-scale washing, due probably to the reduced amount of impurities. The industrial-scale-washed material had a higher melt drawability but lower melt strength than the laboratory-scale-washed samples. The melt strength was similar for all the laboratory-scale-washed samples, but the sample washed with NaOH showed the lowest melt drawability, suggesting that changes occurred in the polymer properties and structure, probably due to degradation. Washing with water alone and with a detergent resulted in similar melt drawability, but the washing temperature had no significant influence on the melt characteristics. In the case of the rigid PE samples, the washing resulted in an increase in both melt strength and drawability, Figure 12c, similar to the flexible PE samples. As shown in Figure 12b, both the melt-strength and drawability were lower for the industrial-scale-washed samples than for the laboratory-scale-washed material, but neither the washing medium nor the washing temperature had any influence on the melt characteristics of the rigid PE material, Figures 12b and c.

5.3 INFLUENCE OF COMPOUNDING CONDITIONS

In general, screw configuration and compounding temperature had no significant influence on the laboratory-scale-compounded flexible and rigid PE materials, and neither the applied vacuum nor the water stripping affected the pilot-scale-compounded rigid PE materials, with regard to properties such as melting temperature, melting enthalpy and ash content, Table 2.

The M_w and PD values of the unwashed flexible samples from the facility in Norway compounded with different screw configurations and different temperatures were very similar, although the higher compounding temperature resulted in a lower M_w for the industrial-scale-washed flexible PE sample from the facility in Norway, Table 2. The pattern was not the same in the case of the washed rigid PE samples. The materials from the facility in Sweden showed a decrease in both M_w and PD with increasing compounding temperature, implying chain scission^{69,75,77,131}, but the materials from the facility in Norway washed on both a laboratory scale and on an industrial scale had the highest M_w and PD when compounded with a medium temperature profile, whereas compounding with a high temperature profile resulted in low and compounding with a low temperature profile resulted in intermediate M_w values.

In the case of the flexible PE samples from the facility in Sweden, Table 3, a higher compounding temperature gave low T_{ox} values, but the screw configuration had no influence. In the case of the flexible PE samples from the facility in Norway, Table 3, the pattern was however different. For the unwashed samples, the screw with mixing elements resulted in a higher T_{ox} value than the screw without mixing elements, and the compounding temperature had no great influence. For the industrial-scale-washed samples, an increase in the compounding temperature led to a further decrease in the T_{ox} , but the screw configuration had no influence. For both the laboratory-scale-washed and industrial-scale-washed rigid PE samples, an increase in the compounding temperature resulted in a lower T_{ox} , Table 3.

Neither the applied vacuum nor the water stripping during the pilot-scale compounding had a significant influence on either the T_{ox} of the rigid PE materials, Table 3, or on the OIT -values.

For both flexible and rigid PE materials, an increase in the compounding temperature appeared to increase the rate of degradation by lowering the T_{ox} -value, which agreed with the fact that the lowest viscosity was shown by the industrial-scale-washed flexible PE sample compounded with a higher temperature profile, Figure 11a, and by the laboratory-scale- and industrial-scale-washed rigid PE samples compounded with the highest temperature profile, Figure 11b. The pilot-scale washed and compounded rigid PE samples, Figure 11c, had similar viscosities regardless of the compounding conditions, and the T_{ox} - and OIT -values were also similar.

The MFR values were not influenced by the screw configuration or compounding temperature of the unwashed and laboratory-scale-washed flexible PE materials. They varied between 0.5 and 0.8 g/10min, but the flexible PE samples from the facility in Norway washed on an industrial scale had an MFR value of ca. 2.2 g/10min when compounded with a low temperature profile and ca. 4.4 g/10min when compounded with a high temperature profile. This important difference was also evident in the viscosity curves, Figure 11a. The MFR values for the laboratory-scale-washed rigid PE samples varied between 0.5 and 1.3 g/10min. For the rigid PE sample groups from both Sweden and Norway, the high temperature compounding resulted in the highest MFR values. The MFR value varied between 0.4 and 0.7 g/10min for the industrial-scale-washed rigid PE samples from the facility in Norway, with the medium temperature compounding giving the lowest value, which supported the M_w pattern.

The compounding conditions had a negligible effect on the entrance pressure losses for both flexible and rigid PE samples, Figures 11d, e and f, with a few exceptions.

In general, the flexible PE materials had a greater melt drawability when compounded with a higher temperature profile, especially in the case of the industrial-scale-washed samples, whereas the melt strength was not significantly affected, Figure 12a. The pattern was different for different groups of rigid PE material. The compounding temperature had no great influence on either the melt-strength or the drawability of the samples from the facility in Sweden, but both the melt-strength and drawability increased with increasing compounding temperature for the samples from the facility in Norway, Figure 12b. Especially in the case of the laboratory-scale-washed samples from the facility in Norway, the melt drawability was doubled and the melt strength was 1.5 times higher for the samples compounded with medium and high temperature profiles, but neither the applied vacuum nor the water stripping during pilot-scale compounding affected the melt-strength or drawability of the rigid PE materials, Figure 12c.

For selected samples, further rheological characterization was done by dynamic rotational rheometry and the results are given in Figure 13 as plots of (i) phase angle vs complex shear modulus, known as a van Gorp-Palmen (vGP) plot, (ii) complex viscosity vs frequency and (iii) storage and loss moduli vs frequency.

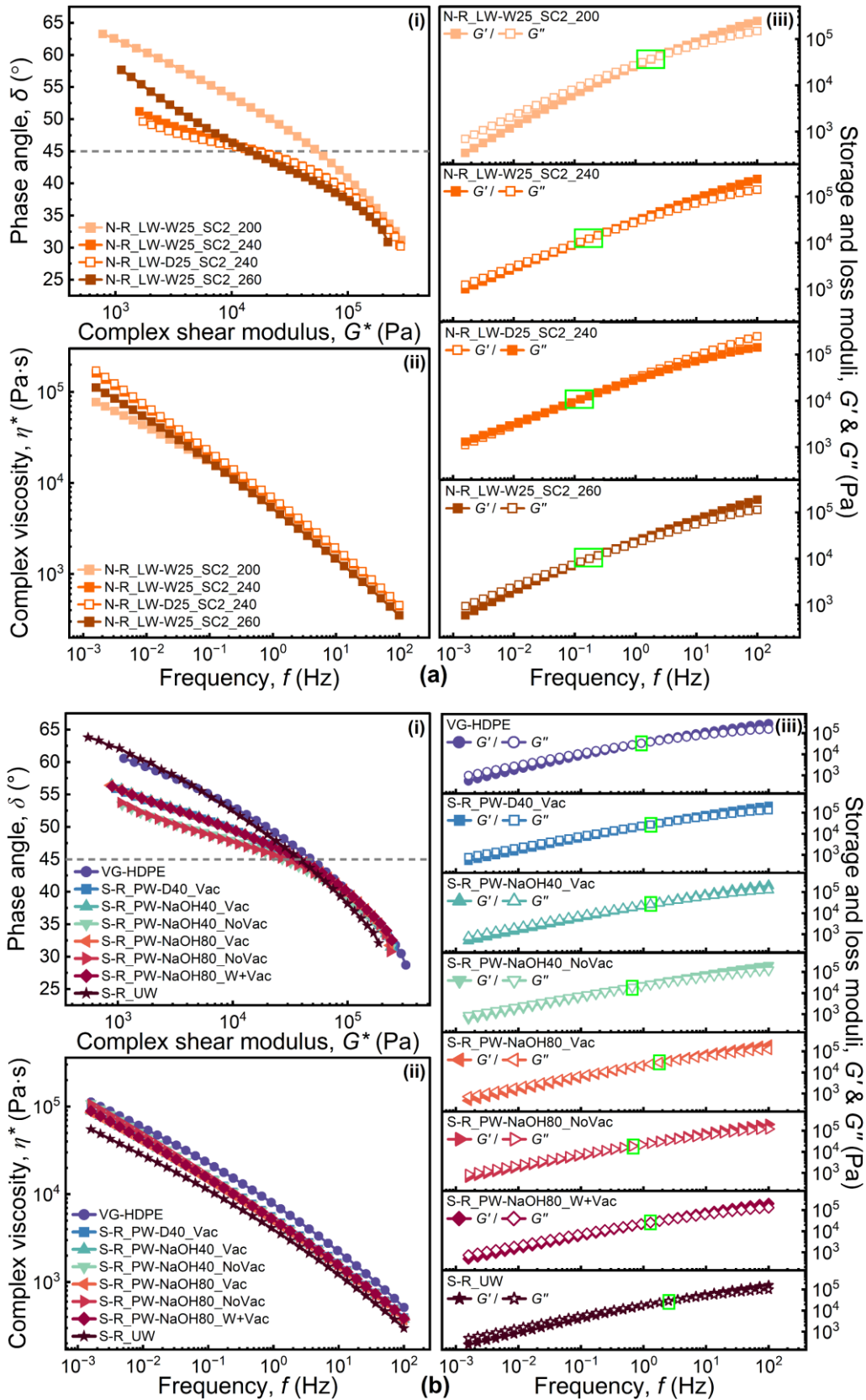


Figure 13. Dynamic rheological behaviour of the (a) laboratory-scale washed and compounded samples received from facility in Norway, (b) pilot-scale washed and compounded rigid PE samples received from facility in Sweden, in frequency sweeps: (i) van Gorp-Palmen plots showing phase angle as a function of complex modulus, (ii) complex viscosity as a function of frequency, (iii) storage and loss moduli as functions of frequency for each sample, with the crossover point shown by a green rectangle.

The VG-HDPE showed a linear-polymer-like behaviour with a monotonic decrease in the vGP-plot, as shown in Figure 13b, and the laboratory-scale-washed rigid PE sample compounded with a low temperature profile, Figure 13a, showed a similar linear-polymer-like behaviour with a monotonic decrease in the phase angle with increasing complex shear modulus, but the shape of the curve differed in the case of the samples compounded at medium and high temperatures, showing characteristics more of a branched polymer with pronounced changes in the slope with increasing G^* .^{77,85,86,97,98} The level of chain branching evidently varied in these samples, since low values of the phase angle in a vGP-plot are usually attributed to an increase in long branches or to other changes in molecular structure.^{77,85,87,89} Similar behaviour was observed in the case of the pilot-scale washed and compounded rigid PE samples, Figure 13b, where the change in the shape of the curves suggests the presence of chain branching. The lower values of phase angle shown by the samples compounded without vacuum also implied that there was a greater level of chain branching. When no vacuum was applied in the degassing zone, a higher rate of degradation and an increase in branching can be anticipated due to the greater access to oxygen.

Different compounding conditions, e.g. different temperatures and vacuum application, led to different levels of chain branching, whereas different washing conditions, e.g. temperature and medium, had no influence, in either laboratory-scale or pilot-scale washing. The batch-mixed unwashed material showed a behaviour similar to that of VG-HDPE, but this may be because the low-temperature processing in a closed batch mixer limited the access of oxygen, causing less degradation. In both laboratory-scale- and pilot-scale-processed rigid PE, the complex viscosity and storage modulus curves supported the vGP-plots, since higher values of both complex viscosity and storage modulus at lower frequencies indicate a more branched structure.^{77,98,132} Among the laboratory-scale-processed samples, the one compounded at a lower temperature was more viscous than the others, and they all showed a pronounced elastic behaviour, with regard to the reference line at the phase angle 45° in the vGP-plot.⁹⁴ In the case of the pilot-scale-processed samples, all exhibited a character that was more viscous than elastic, and this was supported by the crossover points at which the storage and the loss modulus intersect, as a low frequency value of the crossover point indicates a more elastic behaviour.⁹⁴

The results of these studies are valuable indirect indications of whether or not chain branching or molecular degradation had occurred during the washing or compounding steps. The results indicate that degradation often occurred, but it has not been possible to identify any over-riding pattern.

5.4 PARISON OBSERVATIONS DURING THE BLOW MOULDING

The parisons, i.e. the hollow tubes formed by molten plastic as shown in Figure 5, were studied during the blow-moulding of the rigid PE pellets washed and compounded on a pilot scale. As shown in Figure 14, where the fitted lines for the least severe pleating (level-1) are presented, the start of parison pleating differed between the VG-HDPE and the recycled materials. For all the materials, pleating started when the die-opening was less than 4 mm, but with a pressure greater than 50 bar in the case of the VG-HDPE and greater than 30 bar in the case of the recycled materials.

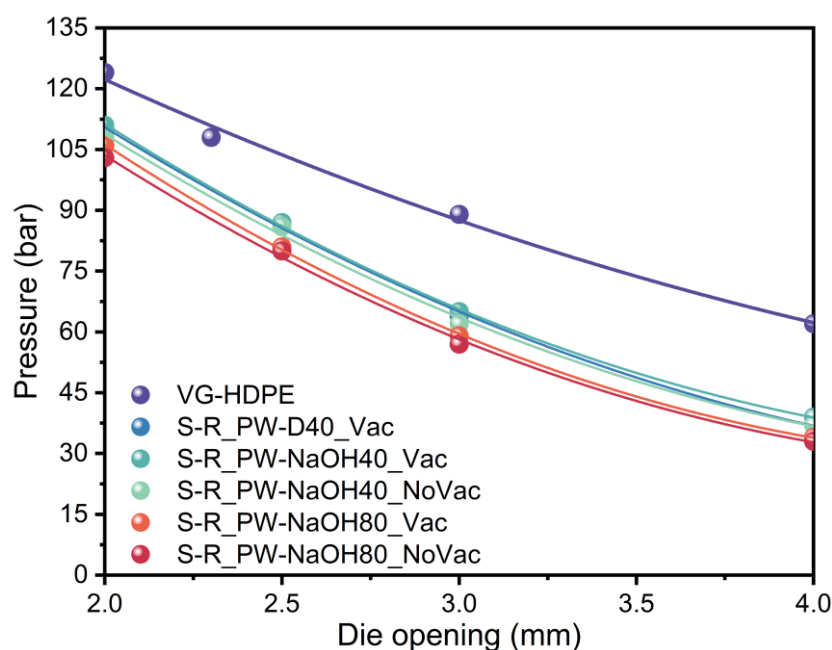


Figure 14. Pressure versus die-opening (points and fitted lines) representing the pleating level-1 of the parisons of the tested materials.

In general, the recycled rigid PE materials showed slightly lower melting enthalpies and a higher ash content, Table 2, a lower thermo-oxidative stability, Table 3, a lower viscosity and lower entrance pressure losses, Figures 11c and f, and a lower melt strength and drawability, Figure 12c, than the virgin grade high density polyethylene used as the reference blow-moulding grade. The operating window was therefore shifted towards lower pressure values with a given die-opening for the recycled materials compared to the VG-HDPE, and the melts of the recycled materials had a lower flow resistance. The recycled materials washed at a high temperature showed slightly lower pressure values under the same process conditions than the material washed at a low temperature. The small shift in processing characteristic allowed a successful blow moulding into 4L-containers with the desired bottle mass of 145 ± 5 g.

5.5 MECHANICAL PROPERTIES

Figure 15 shows the mechanical properties of the injection-moulded flexible PE samples in the different regions of the IM frame.

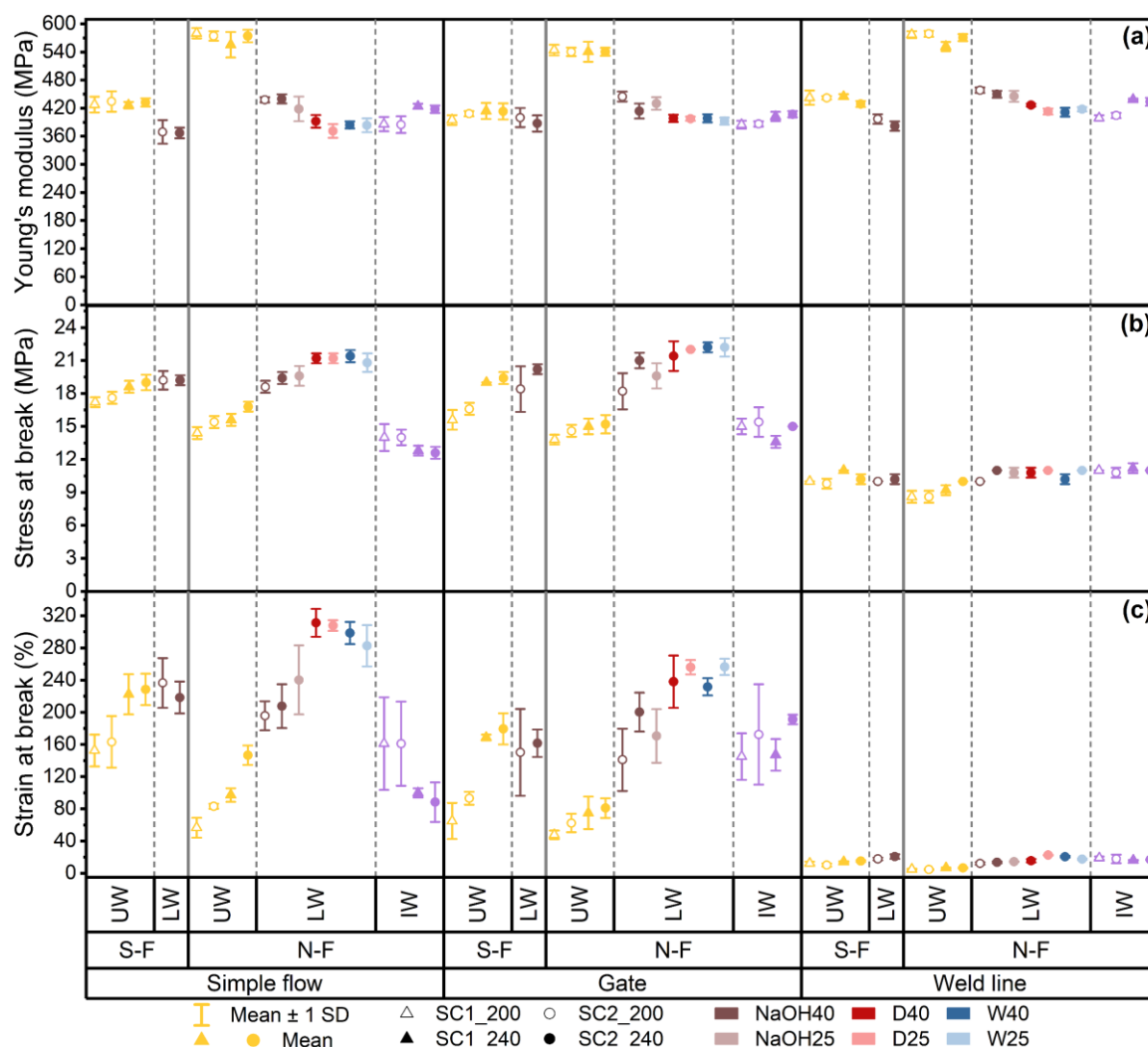


Figure 15. Mechanical properties of the flexible PE samples in the different regions of the IM frame: (a) Young's modulus, (b) stress-at-break, and (c) strain-at-break.

The flexible PE samples, washed on both a laboratory scale and an industrial scale, had lower values of modulus than the unwashed samples, but the washing conditions, i.e. the washing medium and washing temperature, had no great influence. In general, the samples washed on a laboratory scale with NaOH showed a slightly higher Young's modulus than the samples washed with detergent or with water alone on a laboratory scale and also than the samples washed on an industrial scale. In the case of the flexible PE samples, from both Sweden and Norway, neither the compounding temperature nor the screw configuration had any major influence on the Young's modulus, although the industrial-scale-washed material from the facility in Norway had slightly higher values when compounded at a high than at a low temperature. Both the Young's modulus values and the pattern were similar in all regions of the IM-samples, Figure 15a.

The laboratory-scale-washed samples had a higher stress-at-break (tensile strength) than the unwashed samples in both the simple flow and gate regions, but the industrial-scale-washed samples had a lower tensile strength than the unwashed samples in the simple flow region and a tensile strength similar to that of the unwashed samples in the gate region, Figure 15b. Within the laboratory-scale-washed samples from the facility in Norway, the samples washed with NaOH showed, in general, a lower tensile strength than the samples washed with detergent and with water alone but the washing temperature had no significant influence. The difference in tensile strength of the laboratory-scale-washed and industrial-scale-washed samples, in both regions, was quite significant, the latter being much lower, like the melt strength. The tensile strength increased slightly with increasing compounding temperature in both the simple flow and gate regions for both unwashed and laboratory-scale-washed samples, although the industrial-scale-washed samples from the facility in Norway showed the opposite. The screw configuration had no great effect. In the weld line region, all samples showed a similar tensile strength, ca. 10 MPa, which was, as expected, lower than in the other regions but was unexpectedly high and close to the reported value (13 MPa) for virgin PE-LLD.¹³³ Overall, the industrial-scale washing was more severe and reduced the tensile strength more than laboratory-scale washing.

In general, the laboratory-scale-washed samples had a greater strain-at-break in the simple flow and gate regions than either the unwashed or the industrial-scale-washed samples, Figure 15c. The industrial-scale-washed samples had a greater strain-at-break than the unwashed samples in the gate region and in the simple flow region when compounded at a low temperature, although they had a strain-at-break similar or less than that of unwashed samples in the simple flow region when compounded at a high temperature, which may imply that the effect of reduced contaminants dominates at lower process temperatures but that the effect of increased degradation dominated at higher temperatures. The samples washed with NaOH exhibited lower strain-at-break than the samples washed with detergent or with water alone and the washing temperature had no significant influence in either the simple flow or gate regions. In the weld line region, the strain-at-break values were as expected the lowest for all the samples although the washed samples exhibiting slightly greater strains-at-break than the unwashed samples. The strain-at-break increased, in general, with increasing compounding temperature in both the simple flow and gate regions for the unwashed and laboratory-scale-washed samples of both materials received from Sweden and Norway, whereas the industrial-scale-washed samples from the facility in Norway showed the opposite in the simple flow region. In the case of the unwashed samples, this may be due to a better melting of contaminants during high temperature compounding.^{14,124} However, the opposite effect was observed for the industrial-scale-washed samples, which may be due to a reduction in the amount of impurities with washing and also to an increased degradation with increasing process temperature.¹³⁴ The mixing screw (SC2) resulted in higher elongation values due perhaps to a better mixing during compounding but the influence of the screw configuration was lower than that of the compounding temperature.

Overall, in the simple flow region, the recycled flexible PE samples had average Young's modulus, stress-at-break and strain-at-break values that varied between 390-570 MPa, 13-20 MPa and 95-260 % which, together with the MFR values, implied that it might be technically possible to produce end products such as packaging film, flexible bottles or tubes using conventional film extrusion and extrusion blow moulding.¹³⁵

Figure 16 shows the mechanical properties of the laboratory-scale injection-moulded rigid PE samples in the different regions of the IM frame.

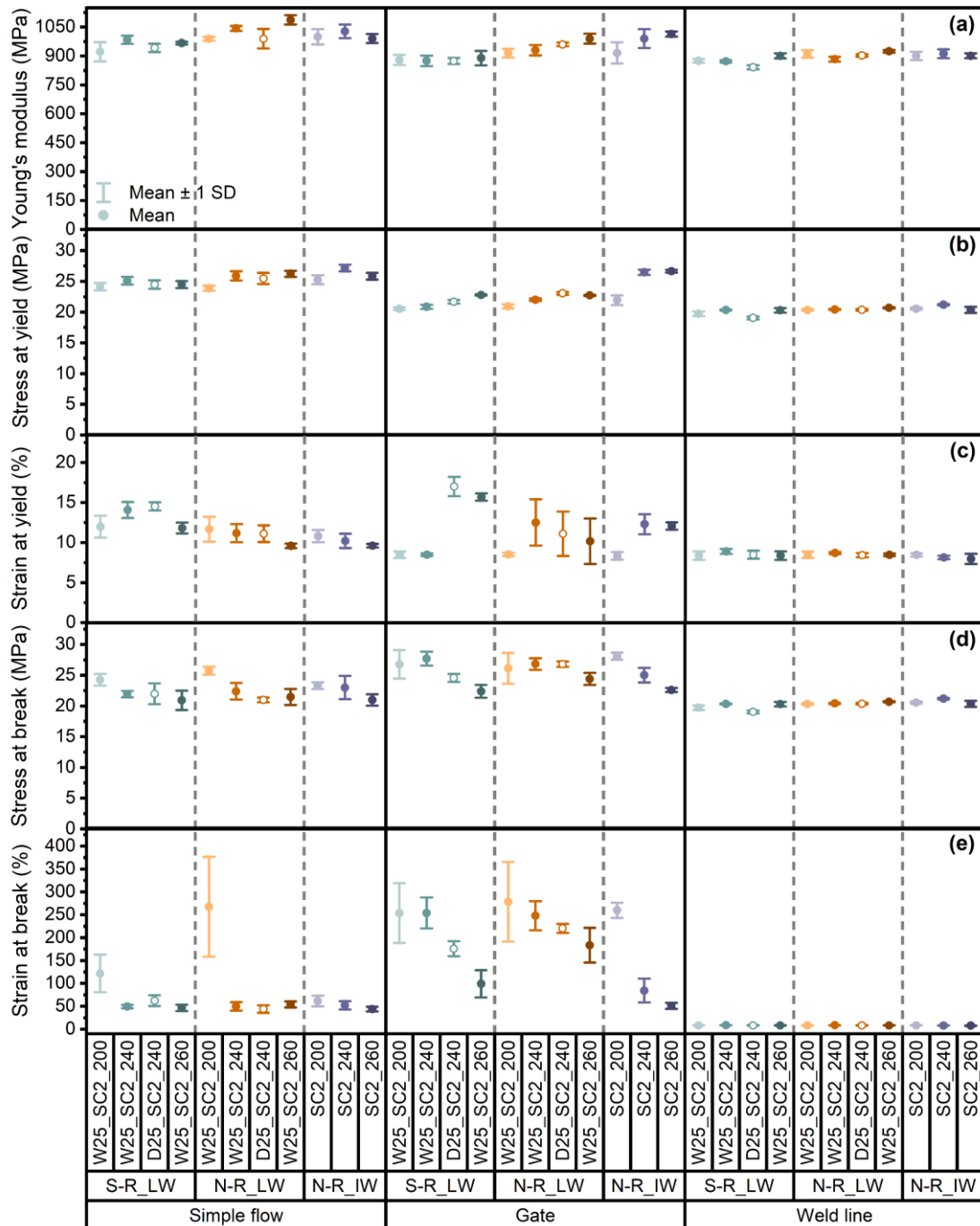


Figure 16. Mechanical properties of the laboratory-scale-processed rigid PE samples in the different regions of the IM frame: (a) Young's modulus, (b) stress-at-yield, (c) strain-at-yield, (d) stress-at-break, and (e) strain-at-break.

Industrial-scale-washed samples from the facility in Norway had values of Young's modulus similar to those washed on a laboratory scale. As with the flexible PE samples, neither the washing medium nor the compounding temperature had a great influence on the Young's modulus of the laboratory-scale injection-moulded rigid PE samples from either Sweden or Norway. The Young's modulus of all the samples were in a similar range in all regions of the IM-frame, Figure 16a.

All groups of samples exhibited similar stress-at-yield values, the highest stresses being in the simple flow region and the lowest in the weld line region, Figure 16b. There were only slight differences due to the process conditions. Similarly, the strain-at-yield values were in a similar range within the same sample groups and between different sample groups without any certain pattern and this was the same in all regions, Figure 16c.

The stress-at-break values were in a similar range for all groups of samples, Figure 16d. The industrial-scale-washed samples exhibited values similar to those of the samples washed on a laboratory scale and the samples washed on a laboratory scale with a detergent showed values similar to those washed with water. There was however a tendency for the stress-at-break to decrease with increasing compounding temperature in both the simple flow and gate regions. The stress-at-break values were slightly lower in the weld line regions, but they were still at ca. 20 MPa.

The industrial-scale-washed samples from the facility in Norway showed lower strain-at-break values than the laboratory-scale-washed samples except when compounded with a low temperature profile, Figure 16e. The samples washed on a laboratory scale with a detergent showed slightly lower strain-at-break values than the water-washed samples. The low-temperature compounded laboratory-scale-washed samples from both Sweden and Norway had the highest strain-at-break values in the simple flow region and the rest of the samples showed similar strain-at-break values. In the gate region, the strain-at-break values decreased in general with increasing compounding temperature.

Overall, the recycled rigid PE samples injection-moulded on a laboratory scale had average Young's modulus, stress-at-yield, strain-at-yield, stress-at-break and strain-at-break values in the simple flow region between 955-1025 MPa, 24-26 MPa, 10-13 %, 22-23 MPa and 50-265 %, which suggests that it might be technically possible to produce end products such as heavy duty films, milk bottles, caps and closures by conventional film extrusion, pipe extrusion, blow moulding and injection moulding.¹³⁵

Figure 17 shows the mechanical properties of the industrial-scale blow-moulded rigid PE samples and VG-HDPE in the different regions of the 4L-container and of the laboratory-scale injection-moulded unwashed material.

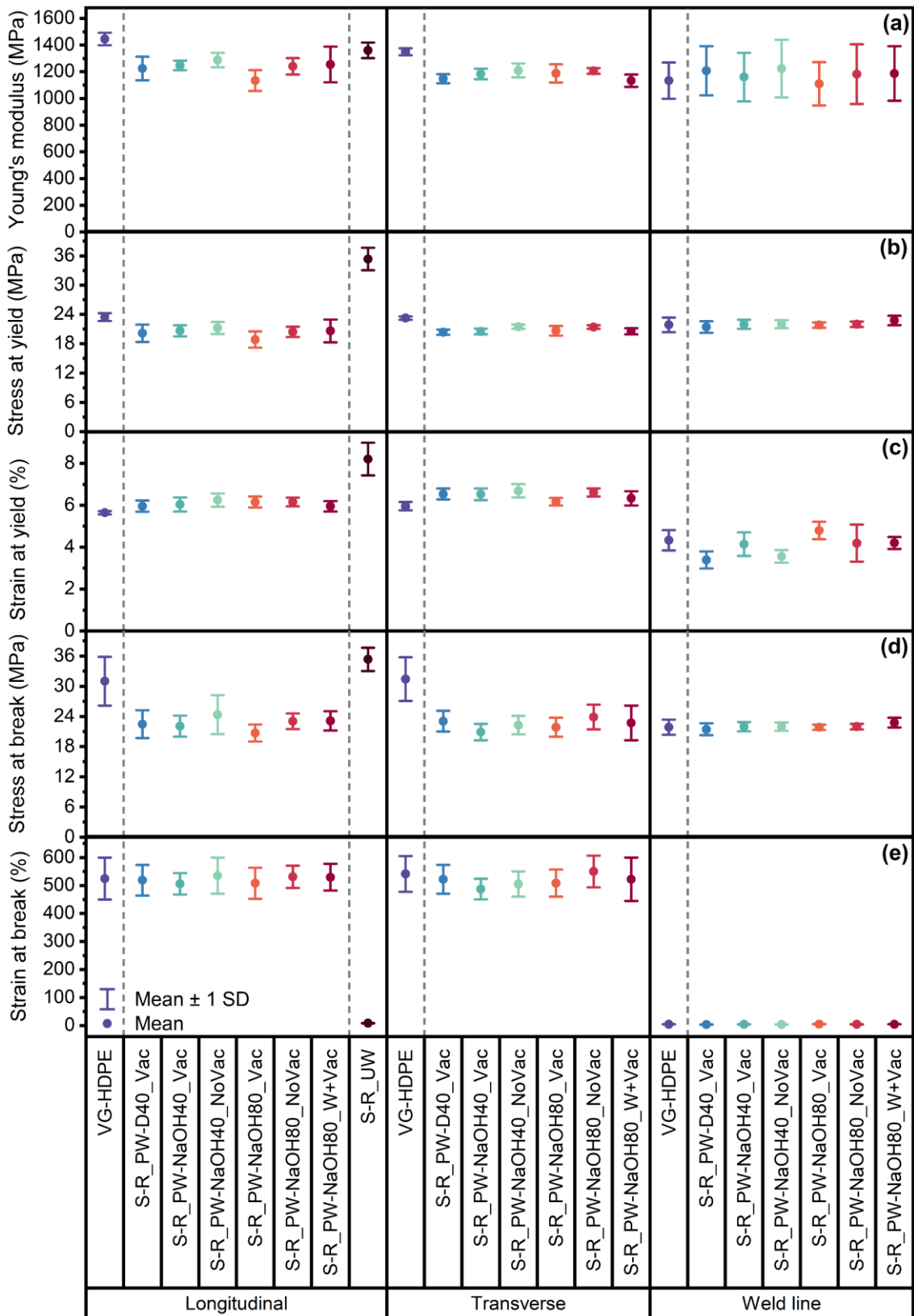


Figure 17. Mechanical properties of the pilot-scale-processed rigid PE samples in the different regions of the 4L-container: (a) Young's modulus, (b) stress-at-yield, (c) strain-at-yield, (d) stress-at-break, and (e) strain-at-break. *Injection-moulded tensile bar of UW material considered as being in the longitudinal direction.

The mechanical properties of the recycled rigid PE samples were in a similar range, with only slight differences. No significant influence of washing or compounding conditions was observed. The samples washed at 40 °C had a slightly higher Young's modulus in the longitudinal direction than those washed at 80 °C, but the values were very similar in both the transverse direction and the weld line region, Figure 17a. The Young's modulus of the recycled materials varied between 1000 and 1300 MPa. In the case of the specimens cut from the bottom of the 4L-container, the yield generally started at the weak weld line and the stress- and strain-at-yield were therefore equal to the stress- and strain-at-break for those specimens. Nevertheless, both the stress-at-yield and -break values were in a similar range for all the recycled materials where the stress-at-yield varied between 19 and 21 MPa, Figure 17b, and the stress-at-break varied between 21 and 24 MPa, Figure 17d. The average weld line strength was 22 MPa. The strain-at-yield and strain-at-break values were also similar for all the recycled materials in both the longitudinal and transverse directions and, as expected, were lower for the specimens with a weld line. The strain-at-yield varied between 6 and 7 %, Figure 17c, and strain-at-break varied between 480 and 560 % in the longitudinal and transverse directions, Figure 17e, whereas both strain-at-yield and -break varied between 3 and 5 % in the weld line regions.

There were no great differences between the mechanical properties of the 4L-containers made from recycled rigid PE and from VG-HDPE, where the latter showed slightly higher value of Young's modulus in the longitudinal and transverse directions at ca. 1400 MPa. Similarly, this material had slightly higher values of stress-at-yield (ca. 23 MPa) and stress-at-break (ca. 31 MPa) than the recycled rigid PE samples in longitudinal and transverse directions.

Since the unwashed material was not blow-moulded, no direct comparison with the containers is possible. In the injection-moulded bars there was no yield but a direct ductile fracture. The average Young's modulus was 1360 MPa and the average stress- and strain-at-break were 35 MPa and 8 %, respectively.

5.6 COLOUR, GLOSS AND APPEARANCE

The colour and gloss were determined only for the 4L-containers shown in Figure 18, which were lighter when NaOH was used in the washing. Fisheye formation was observed on the 4L-containers washed without NaOH, but the reason was unclear. Table 4 shows the average L^* , a^* , b^* and gloss values of the 4L-containers and batch-mixed UW sample.



Figure 18. Examples of blow-moulded 4L-containers.

Table 4. The L^* , a^* , b^* and gloss of the 4L-containers and batch-mixed UW material.

| Sample | SCI | | | SCE | | | Gloss (GU) |
|---------------------|-------|-------|-------|-------|-------|-------|------------|
| | L^* | a^* | b^* | L^* | a^* | b^* | |
| VG-HDPE (white) | 83.6 | -0.6 | -4.9 | 83.4 | -0.7 | -4.8 | 7.5 |
| VG-HDPE (black) | 59.5 | -2.0 | -8.6 | 59.2 | -2.1 | -8.5 | 6.9 |
| S-R_PW-D40_Vac | 55.7 | -3.7 | 3.5 | 55.0 | -3.8 | 3.7 | 11.0 |
| S-R_PW-NaOH40_Vac | 60.0 | -5.6 | 2.5 | 59.6 | -5.7 | 2.6 | 9.0 |
| S-R_PW-NaOH40_NoVac | 61.0 | -5.9 | 2.5 | 60.6 | -6.0 | 2.6 | 7.3 |
| S-R_PW-NaOH80_Vac | 61.7 | -5.9 | 2.8 | 61.1 | -6.0 | 2.9 | 10.7 |
| S-R_PW-NaOH80_NoVac | 61.8 | -5.8 | 2.4 | 61.2 | -5.9 | 2.5 | 10.9 |
| S-R_PW-NaOH80_W+Vac | 61.9 | -6.0 | 2.7 | 61.4 | -6.1 | 2.8 | 9.5 |
| S-R_UW | 50.0 | -1.4 | 4.9 | 49.9 | -1.5 | 5.0 | 3.3 |

The visual observation was confirmed by colour measurement. It was, unfortunately impossible to obtain meaningful data for the translucent VG-HDPE sample. For the opaque washed 4L-containers, the values for the samples measured with the specular component included (SCI) were very similar to the values with the specular component excluded (SCE), in agreement with the low gloss values. These samples were essentially grey, but after washing with NaOH, they were slightly more green and slightly less yellow. The lightness L^* -value was significantly higher when NaOH was present in the washing medium. The reason for the colour differences observed between the recycled samples, may be either better removal of contaminants or greater degradation.^{65,136,137} The gloss of the 4L-containers was very low.

6 CONCLUSIONS

The feedstock in both the flexible and rigid PE streams had a PE content of ca. 94 wt.%, the main contamination in both streams being by PP. The properties of recycled materials recovered from separated household plastic packaging waste in Sweden and from mixed municipal residual waste in Norway were similar to each other, with a few exceptions, in both the flexible and rigid PE streams.

Washing was required to obtain plastic waste flakes free from surface contaminants due to potential health risks and to facilitate the subsequent compounding and shaping processes. Washing, in general, improved the properties of both the recycled flexible and rigid PE materials, but some degradation was evident, especially in the industrial-scale-washed flexible PE samples. The laboratory-scale-washed flexible PE samples were however less degraded, and showed in general better properties than the industrial-scale-washed samples. The laboratory-scale-washed rigid PE samples had properties similar to those of the industrial-scale-washed samples.

The influence of the washing medium was greater than that of the washing temperature, which had a negligible effect. Washing with water alone or with added detergent gave properties close to each other, in the case of both laboratory-scale-washed flexible and rigid PE samples. For the flexible PE material, however, washing with NaOH on a laboratory scale evidently led to degradation and different properties. For the rigid PE material, a pilot-scale detergent washing, on the contrary, resulted in a lower thermo-oxidative stability than washing with a combination of detergent and NaOH.

In general, neither the screw configuration nor the compounding temperature had a significant influence on the properties of the unwashed materials. The industrial-scale-washed flexible PE samples were more prone to further degradation when compounded at a high temperature, unlike the unwashed materials, but the influence of screw configuration was insignificant. An increase in compounding temperature also led to greater degradation of the laboratory-scale-washed rigid PE materials. Different compounding temperatures during the laboratory-scale compounding and the application of vacuum during pilot-scale compounding led to different levels of chain branching and changed the molecular structure, but the water stripping had no significant effect.

These observations for both flexible and rigid PE material indicate that the process conditions influence especially the thermo-oxidative stability of the material and should be considered carefully with regard to the long-term durability of the recycled materials. Washing on a large scale requires optimization for each stream and NaOH should be used with caution in the case of the flexible PE stream. Rheological characterizations were found to be useful and sensitive to provide information on degradation-related structural changes of the relatively difficult and heterogeneous samples of post-consumer waste.

Based on the results of this study, a recommendation of a good recycling route in the case of the flexible PE waste would be washing with low to moderate temperature using detergent or water alone followed by a low to moderate temperature compounding. In the case of the rigid PE waste, washing at a low to high temperature using detergent and NaOH is recommended followed by low to moderate temperature compounding with perhaps an applied vacuum during compounding.

Nevertheless, the properties of all the recycled materials studied here were useful in many applications. The successful production of containers on an industrial scale also supported the recyclability and applicability of post-consumer rigid polyethylene. In the case of highly degraded samples, the addition of upgrading components such as thermo-oxidative stabilizers and virgin grade polyethylene might be considered.

7 FUTURE WORK

Future studies might investigate the influence of washing conditions on an upscaled processing of the flexible PE stream. The milder conditions of a laboratory-scale washing caused less degradation of the flexible PE material, whereas high-temperature washing with harsh chemicals on an industrial scale led to considerable deterioration. Therefore, it is of interest to explore different washing temperatures and washing media on a large scale. The industrial-scale shaping of the rigid PE stream into 4L-containers worked well, and an industrial-scale manufacture of flexible end products using the recycled flexible PE stream might be further explored.

The durability of the recycled materials should be further studied to better understand both the short-term and long-term behaviour of the materials, depending on the intended application. This might involve ageing studies, due to the environment, chemicals and stress.

The rheological characterization suggests that chain branching and structural changes occur in the recycled materials, especially after certain process conditions. This should be further studied to better understand the underlying degradation mechanisms, and other characterization methods should be implemented, such as nuclear magnetic resonance spectroscopy, intrinsic viscosity measurements, multi-angle light scattering coupled with size-exclusion chromatography or asymmetric flow field-flow fractionation and other methods.

A common problem with recycled materials is their odour, and this might be problematic both during manufacture and in the end product. Further studies to minimize or eliminate the odorous components during the recycling process with the help of washing and compounding would be essential.

The cleaning efficiency during washing is of interest considering the further use of the recycled materials as an end product on the market. The washed samples, in general, showed colour differences but the reason was unclear and this might be of interest to study further.

The second most frequent polymeric material in use, polypropylene, should be similarly studied. The influence of washing would be of great interest, because the findings of the present work suggest that there is no common washing process that suits both flexible and rigid PE streams.

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