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Processability and mechanical properties of extensively recycled high density polyethylene

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ABSTRACT

In plastics industry it is a common practice to mechanically recycle waste material arising from production. However, while plastics are mechanically recycled, their mechanical properties change. These changes may affect material processing conditions and quality of the end products; therefore they need to be quantified. In this study, mechanical recycling of high density polyethylene (HDPE) was simulated by one-hundred (100) consecutive extrusions cycles. During extrusion, processability of virgin HDPE and its recyclates was studied by recording the processing conditions, i.e. melt pressure and extrusion torque, while after preparation of the recyclates, melt flow index measurements (MFI), small amplitude oscillatory shear tests (rheological properties), and differential scanning calorimetry measurements (DSC) of thermal properties were performed. Also, mechanical properties in solid state were characterized in terms of hardness and modulus measured by nanoindentation, and finally, shear creep compliance was measured to characterize the materials' time-dependent mechanical properties and its durability in solid state. In addition, gel permeation chromatography (GPC) and solubility tests were implemented to study changes in the material structure.

The results on rheological and MFI measurements indicate significant structural changes in the material that occurred during the first 30 extrusion cycles. Those changes affect material processability which is as well supported by the recorded processing pressure and torque. On the other hand, processing did not significantly affect material thermal properties. Results on hardness and modulus show deterioration of the material mechanical properties after 10th reprocessing cycle. Similarly, shear creep compliance measurements showed an unfavourable effect of mechanical recycling on the time-dependent mechanical properties, particularly after the 30th extrusion cycle. In addition, results suggested chain branching as a dominating mechanism through first 30 extrusion cycles, domination of chain scission afterwards and also presence of cross-linking after 60th extrusion cycle.

1. Introduction

In the second half of the 20th century, plastics became one of the most universally-used and multipurpose materials in global economy. They are used in wide range of applications, for instance, packaging, household appliances, sport accessories, electronics, medicine, and as well in high performance components in automotive and aircraft industry. The plastics industry has benefited from 60 years of growth with a year expansion of 8.7% from 1950 to 2012 [1].

With continuous growth of plastic industry also amount of generated waste is increasing, not only as post-consumer waste but also as waste generated during the production process. Runners, sprues and off-specification products are common in production; they are easy to identify and are of high quality [2]. Instead of being rejected as waste they are granulated and reprocessed; this makes economic sense as it reduces both production waste and utilisation of raw materials. Reprocessing of production waste is particularly important in cases where amount of waste is high in respect to the product. As an extreme case, micro injection moulding can be exposed; in its case the volume of delivery system, i.e. sprue and runner systems, which represent the waste material, can exceed that of the injected part by a factor 100 or more [3]. Another

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example is extrusion blow-moulding technology where amount of the scrap during production can reach 40% [4].

Reprocessing of production waste is also known as in-house or primary recycling and represents a type of mechanical recycling where plastics are grinded and then processed through a physical process [5]. In these cases material can be exposed to excessive number of reprocessing that may affect its processability and quality of end products.

Properties of mechanically recycled polymers do not remain the same because of degradation from heat, mechanical stresses and oxidation during their reprocessing. Degradation affects molecule structure and consequently material mechanical properties [6,7] thus the quality is the main issue when dealing with mechanically recycled products [8]. Besides mechanical, also rheological properties are changed [9–11]. This has direct effect on the processability and needs to be considered during the following technological processes.

In line with mentioned above, our investigation is focused on study of processability and mechanical properties of two widely used polymers, namely LDPE and HDPE, which shared 29.5% of European plastics demand in 2012 [1]. Both, LDPE and HDPE, belong to the group of polyethylenes, however, their behaviour during mechanical recycling differs significantly, and one cannot be predicted from another Thus they need to be studied individually. Findings on extensively recycled LDPE have been already reported by our group [12], whereas here we summarize results on mechanical recycling of HDPE material.

2. Material and methods

2.1. Material

For the purposes of the investigation, high density polyethylene (HDPE) LANUFENE®HDI-6507UV, produced by Ras Lanuf Oil & Gas Processing Co. was used. The material has density 0.965 g/cm^3 (23 °C) and melt flow index 7.5 g/10 min (190 °C and 2.16 kg) [13]. Furthermore, producer provides information on suitability of material for the manufacturing of crates, containers, trays and other similar injection moulded objectives where good weather-ability and mechanical properties are needed. In addition, producer gives information that material contains UV light stabilizers and recommends its processing temperature to be 220–270 °C [13].

2.2. Simulation of mechanical recycling

The process of mechanical recycling was simulated by extensive extrusion. For this purpose, twin-screw extruder, PolyLab PTW 16/40 OS, produced by Thermo Scientific (Germany) was used. Extruder had installed horizontal rod die (type 557–3235) and a nozzle with diameter of 4 mm. The arrangement of the co-rotating extrusion screws used for the process of simulated recycling is presented in Fig. 1.

Material was extruded at screw rotation of 150 min^{-1}, and processing temperature of 240 °C with a throughput between 1200 and 1300 g/h. After material was extruded, it was pelletized using a Thermo Haake pelletizer (type 557-2685) and a portion was removed for the purpose of afterwards characterization. The rest of material was submitted to a new extrusion cycle.

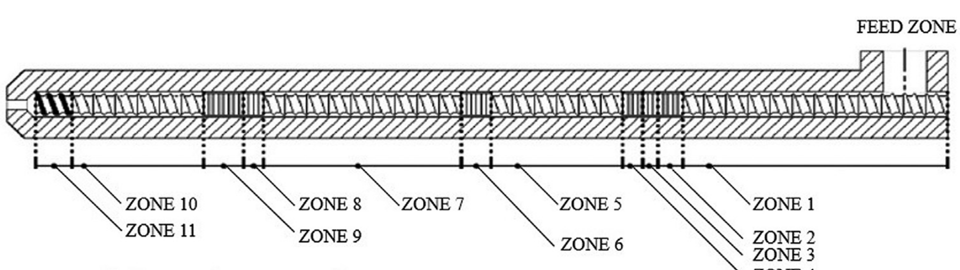
2.3. Sample preparation

In order to perform characterization of recycled materials, samples of cylindrical shape with a diameter of 6 mm and length of 150 mm were prepared. For the preparation, a non-commercial apparatus for the gravimetric casting was used [14]. Gravimetric casting proved to be the most suitable method for assuring isotropy of samples. A glass tube Ø9 × Ø6 × 200 mm was placed between upper and bottom fixation elements (Fig. 2). A heater at 200 °C in the shape of a hollow cylinder was positioned around the glass tube and travelled with 4 mm/min from the bottom to the top of the tube. The glass tube was filled with a polymer granulate, between 20 and 30 mm above the upper surface of the heater. Compression force was assured with the piston having a loading mass of 1100 g on top of it (Fig. 2).

2.4. Characterization

2.4.1. Measured processing parameters

PolyLab PTW extruder used for mechanical recycling simulation allows monitoring and recording the screw torque, $M(t)$, as well pressure of the melt, $p(t)$, in the die channel. Screw torque was



ZONE 1: feed screw elements – 10 pieces
ZONE 2: mixing 0° offset elements – 3 pieces, mixing 90° offset elements – 2 pieces, 30° between elements
ZONE 3: mixing 0° offset elements – 3 pieces, 60° between elements
ZONE 4: mixing 0° offset elements – 2 pieces, mixing 90° offset elements – 2 pieces, 90° between elements
ZONE 5: feed screw elements – 6 pieces
ZONE 6: mixing 0° offset elements – 6 pieces, 60° between elements
ZONE 7: feed screw elements – 9 pieces
ZONE 8: mixing 0° offset elements – 4 pieces, 60° between elements
ZONE 9: mixing 0° offset elements – 4 pieces, mixing 90° offset elements – 4 pieces, 90° between elements
ZONE 10: feed screw elements – 6 pieces
ZONE 11: extrusion screw element – 1 piece

Fig. 1. Used configuration of extruder screws.