Research on the Causes of Cracking of Pipes Derived from Recirculated Polyethylene

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ABSTRACT

The objectives of the research was to determine the causes of cracks in pipes made of high-density polyethylene (HDPE) regranulate during their storage. A visual assessment of the cracks was carried out and their fatigue nature was found. Due to the insufficient information on the composition and properties of the processed regranulate, tests were carried out on the density distribution and mass flow rate of plastic samples taken from various areas of the pipe wall. Comparative tests were performed using infrared spectrophotometry of a plastic sample taken from a pipe, commercial HDPE and commercial polypropylene (PP) by attenuated total reflection/Fourier-transform infrared spectroscopy (ATR-FTIR), thermogravimetry (TG) and differential scaning calorimetry (DSC) method. There were differences in density and flow rate depending on the distance from the outer surface of the pipe, reaching several percent. Based on the conducted spectroscopic, thermogravimetric and differential scanning calorimetry studies, none of the expected plastics contamination, mostly PP, has been found in recirculated polyethylene. Futhermore, the expected decrease in the temperature of phase transformations and the beginning of decomposition of polyethylene (PE) after recycling, compared to the original one, were visible. Possible causes of cracks were considered to be differences in the PE structure in the pipe wall and the occurrence of possible porous structures, which when storing pipes in variable load and temperature conditions could cause the propagation of microcracks.

Keywords: polyethylene recyclate, pipes, fatigue crack, phase transformations.

INTRODUCTION

Material recycling processes are used for waste polymer materials, very often includes high-density polyethylene, which contain various types of impurities in their composition, frequently in the form of ad-mixtures of other materials [1–4]. They affect the course of granulation and processing processes, consequently, the properties of the products. The course of HDPE reprocessing, mainly by the extrusion method, may be affected by the occurrence of increased pressure, temperature and flow rate pulsations in the channels of the plasticising system and the boring head [5–7]. That in the finished product, may cause heterogeneity of the structure, resulting in higher variability of the properties such as density, surface roughness, thermal and mechanical properties, and an increase in internal stresses [8–12]. That, in combination with the presence of contamination, may lead to defects in the finished products and shorten the time of their proper operation and storage. Mechanical damage, in the form of fractures on the surface of the pipes, caused by the low hardness of HDPE, may also occur during transport, storage, and subsequent installation of pipes in the ground [13–15]. According to the literature [14, 16], cracks are most often taking the form of longitudinal brittle fracture surfaces with a different rounding radius,

usually exceeding 0.02 mm. As the hardness of polyethylene decreases, its plasticity increases, and its susceptibility to the formation of surface scratches increases. Whereby, deeper fractures on the surface of the product with a smaller radius of rounding may cause the effect of the notch. The concentration of stresses in cylindrical products will be strengthened by circumferential forces, reducing the crack growth resistance, resulting in the pipe wall fracture. Simultaneously, it may be intensified by the multiple displacements of pipes, for example during long storage for instance in the pipes stored in coils, especially when the temperature changes during day-night, which may contribute to faster cracks propagation [16, 17]. The results of research on the development of cracks and deformations of HDPE pipes available in the literature regards mainly pipes for pressure installations and longitudinal cracks [14, 17–19]. The issues related to the stress state as well as the formation and propagation of a crack in pipes filled with inner pressure focus on the theoretical and experimental determination of the critical stress intensity factors [16, 18]. Viscoelastic materials, which include HDPE, subjected to long-term loads, are characterized by the variability of the critical stress intensity factor over time [20]. Within time, the value of this factor decreases with the decrease of Young's modulus, with the increase of deformation. In the case of plastic pipes, there are various types of fracture mechanisms (brittle, ductile, and transient), with HDPE pipes most often having the so-called "quasi-brittle" [16]. The Time effect of crack initiation and their propagation on the fatigue strength depends on many factors, including molecular weight, density, stress amplitude and crack depth. Similar phenomena occur during the storage of HDPE pipes under variable conditions of external load, temperature, humidity, and the possible presence of surfactants or chemically inert substances, which may cause microcracks and their propagation, leading to complete fracture of the pipe wall [5, 8, 11, 17, 19]. Such cracks are common in pipes made of recirculated HDPE during storage, and an attempt to explain the causes of this phenomenon, recognized mainly in material contamination, changes in structure, and thermal properties, is the subject of the undertaken research. Example of such cracks occur, in pipes made of recycled HDPE during storage, and the attempt to explain the causes of this phenomenon, which are mainly seen in an exceeded hoop stress amplitudes for pipes stored in coils, in several levels, as well as material contamination, changes in structure and thermal properties, are the subject of the undertaken research.

METHODOLOGY OF RESEARCH

The objectives of the research was to determine the causes of cracks in pipes made of high-density polyethylene regranulate during their storage. Particular pipes has been coils and kept in the several levels. Stored in the same conditions pure HDPE pipes has shown no cracks. In order to verify the possible occurrence of inhomogeneity in the structure of the pipe material, a visual assessment of the surface and area of cracks has been carried out. Due to the insufficient information on the composition and properties of the processed regranulate, tests were carried out on the density distribution and mass flow rate of plastic samples taken from various areas of the pipe. Comparative tests were performed using infrared spectrophotometry of a plastic sample taken from a pipe, commercial HDPE (Hostalen ACP 9255P) and commercial PP (Moplen HP456J) by infrared spectrophotometry (FTIR), thermogravimetric analysis and differential scanning calorimetry. Polypropylene was tested to confirm or rule out its addition in the regranulate from which the test pipe was obtained. The changes in the shape, fracture area and internal surface of cracks area as well as internal and external of the tested and comparative pipe, obtained in the same conditions from non-recirculated HDPE has been identify by unaided eye. Surface observations were made with an Eclipse LV100ND microscope using a 5x magnification objective. Density was determined by hydrostatic method using RADWAG AS 82/220.R2 analytical balance, distilled water, and the test temperature was 25°C. The mass flow rate tests were carried out in accordance with the recommendations of PN-EN ISO 1133-1 using a load plastomer at a temperature of 190°C and a piston load of 5 kg with the sampling time of 30 s. The density and the melt flow rate were determined for the three layers of the pipe by taking samples from the inner surface, middle layer (core) and outer surface. Spectrophotometric studies were carried out by the attenuated total internal reflection ATR-FTIR method using the TGA-IR TENSOR 27 Brüker spectrophotometer at the Department of Polymer Chemistry, Maria

Curie-Skłodowska University in Lublin. TG analysis was performed on a Netzsch STA 449 F1 Jupiter thermal analyser (Germany) in a helium atmosphere (flow 20 ml·min^{-1}), at a heating rate of 10 K·min^{-1}, temperature ranging from 30 to 500°C and sample weight of 10 mg.

RESULTS

As a result of the pipe cross-section observation, a clear symmetry disturbance was found (Fig. 1). This was most likely caused by storage with a significant external load, largely as a result of hoop stress in the colis pipes storage.

Transverse cracks were observed on the pipe surface (Fig. 2a), with predominat ductile fracture and transient mechanism. We can speak of a "quasi-brittle" fracture mechanism, typical for PEHD. However, the observation of the cross-section at the place of the crack (Fig. 2b) revealed its typical fatigue character, probably in the area of initiation and further propagation from the inner surface of the pipe. In this area of the pipe,
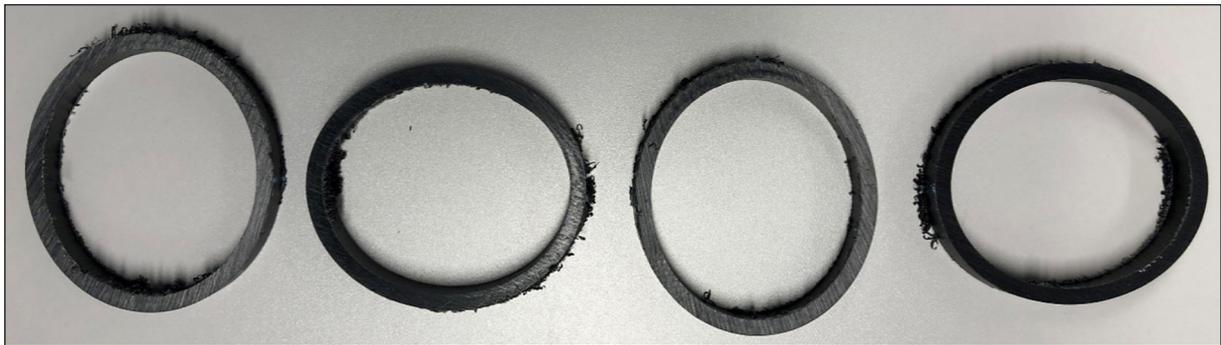

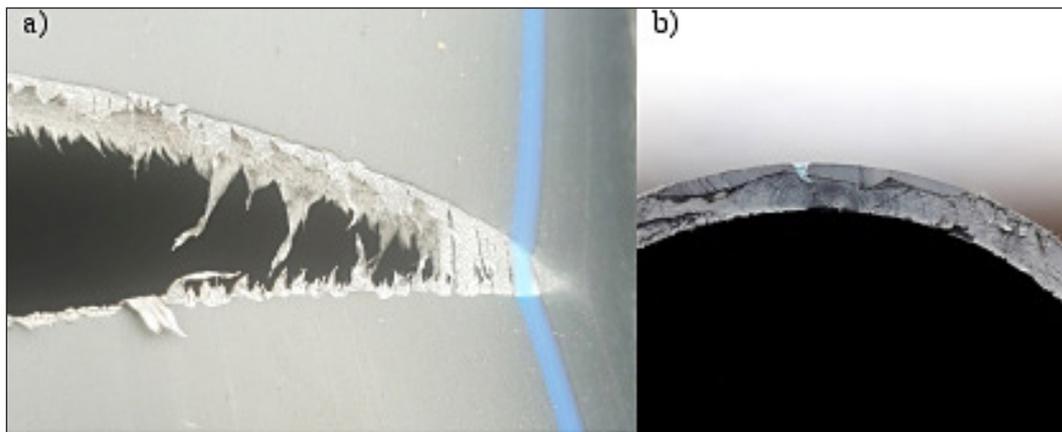**Fig. 1.** Image of the cross-sections of the pipe sections

Fig 2. View of the pipe surface in the fracture area (a) and its cross-section in the beging of the fracture's ares (b)
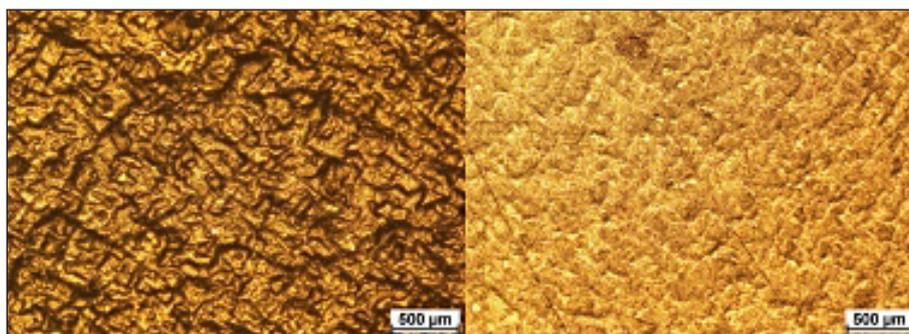

Fig. 3. Microscopic images of the recirculated (left) and fresh HDPE (right) internal pipe surface

Table 1. Results of the melt flow index tests

Sampling area	Inner surface	Core	Outer surface
	0.045	0.075	0.048
	0.046	0.075	0.048
Weight of the measuring section m, g	0.048	0.077	0.046
	0.045	0.079	0.047
	0.043	0.075	0.044
	0.044	0.076	0.045
Average weight of the measuring section m_s, g	0.045	0.076	0.046
MFR [g/10 min]	0.90	0.76	0.93

mainly compressive stresses occurred in the absence of longitudinal and transverse cracks, so the fracture was probably caused by changes in stresses due to temperature changes during storage, faulty material or surface defect.

The analysis of microscopic images of the inner surface for the tested and comparative pipe (Fig. 3). At first it indicates much greater unevenness on the surface of the pipe obtained from the recycled PE-HD, which could have contributed to cracking and to the rapid development of micro-cracks and their propagation due to the notch effect.

As a result of the conducted measurements, slight differences in the density of the pipe material were obtained depending on the location of the sampling area. The material collected from the internal surface was characterised by an average density value of 904 kg/m³, 963 kg/m³ from the core, and 948 kg/m³ from the external surface. Changes in the density of the material in the cross-section of the pipe wall could have caused internal stresses. The lower density in the

area closer to the inner surface may indicate the occurring discontinuities of the structure, constituting internal notches and which may contribute to the formation of micro-cracks. The results of the melt flow index tests (Table 1) of samples taken from the pipe indirectly confirm the differences in the structure and structure of polyethylene in the pipe wall.

The infrared spectra obtained by the attenuated total internal reflection (ATR) method of the tested samples are presented in the following Figures 4–6. The absorption bands in the spectra (Fig. 4, 5) in the region of 2915 cm⁻¹ and 2847 cm⁻¹ are bands related to asymmetrical, symmetrical and C-H stretching vibrations in the -CH₂ group. The band at the length of 1455 cm⁻¹ is characterised by asymmetric deformation C-H vibrations. A band in the range of 722 cm⁻¹ indicates the presence of aliphatic -C-C-C- backbone bonds. The spectra recorded for the polyethylene sample taken from the tested pipe did not differ significantly from the spectra recorded for the porcine polyethylene sample. Figure 6 shows the spectrum for the tested polypropylene,
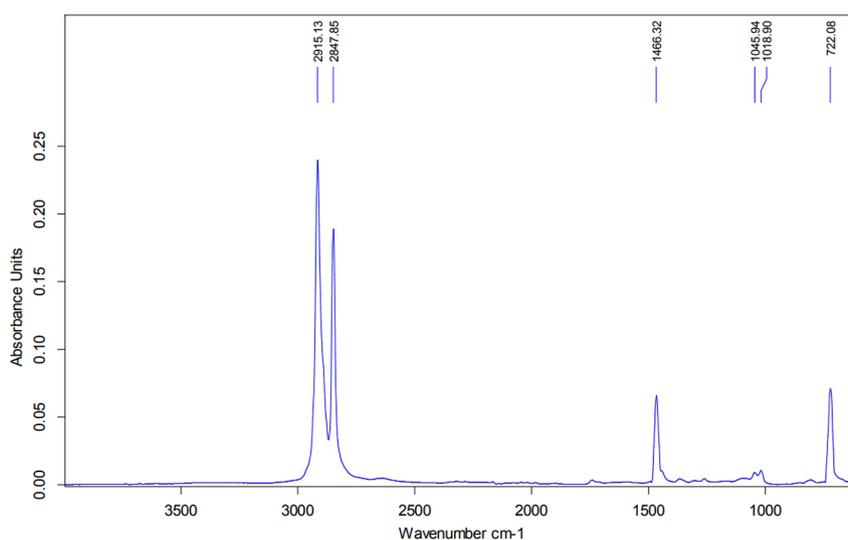

Fig. 4. Spectrum of ATR-FTIR HDPE Hostalen ACP 9255P

where the 2952 cm⁻¹ and 1375 cm⁻¹ bands are noticeable, which are responsible for the vibrations of the CH$_3$ group characteristic for polypropylene. The absence of these bands in the sample of polyethylene constituting the pipe material indicates no contamination with polypropylene.

Figure 7 shows the DSC curve of the tested polyethylene regranulate used in the process of extrusion of the tested pipe in the range from 25 to 500°C. When examining the course of the DSC curve, its linear course can be noticed in the temperature range 25–120°C. Then there is a large endothermic signal at 138.3°C, which is responsible for the softening of the polyethylene due to disappearance of crystallites. Another recorded endothermic signal starts at about 440°C with a maximum at 486.9°C and is related to the thermal

decomposition of the material. There are no additional signals, which proves the absence of significant quantitative impurities, for instant PP.

The results of thermogravimetric tests of pipe material samples and comparative samples are presented in Figure 8.

The analysis of the obtained TG/DSC curves makes it possible to state that the tested material taken from the pipe has the highest level of the transformation peak associated with the disappearance of the crystal structure at the temperature of 131°C, which is close to the transformation temperature of non-recirculated HDPE (133°C). At the same time, the transformation peak occurs at a slightly (about 7°C) lower temperature than in the regranulate sample, which may indicate some mechanical and thermal degradation of
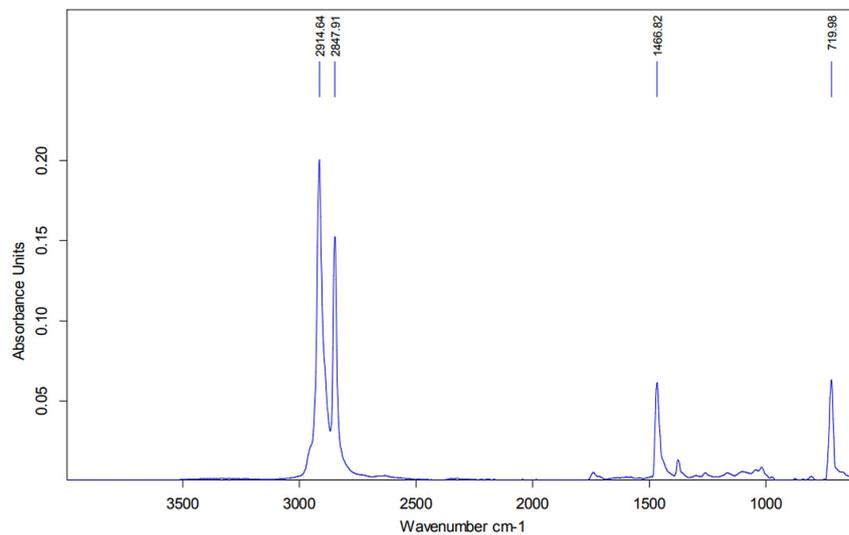

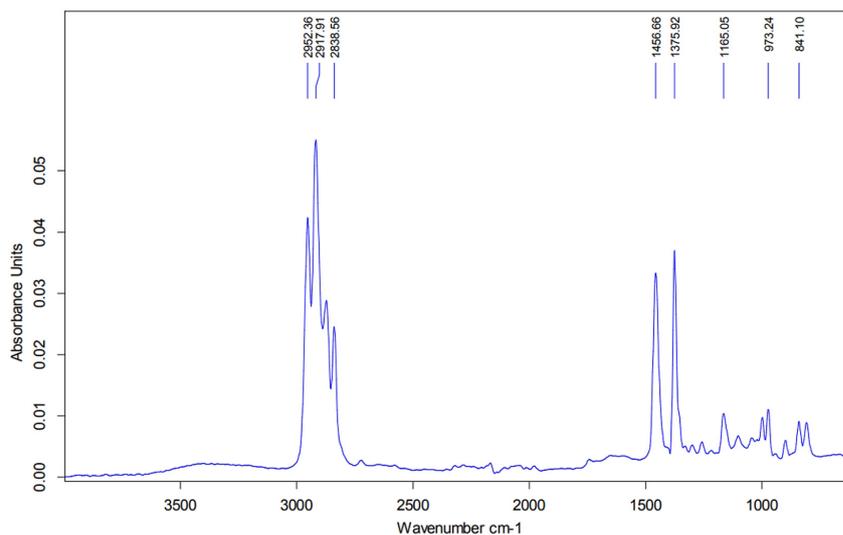**Fig. 5.** ATR-FTIR HDPE spectrum of the pipe

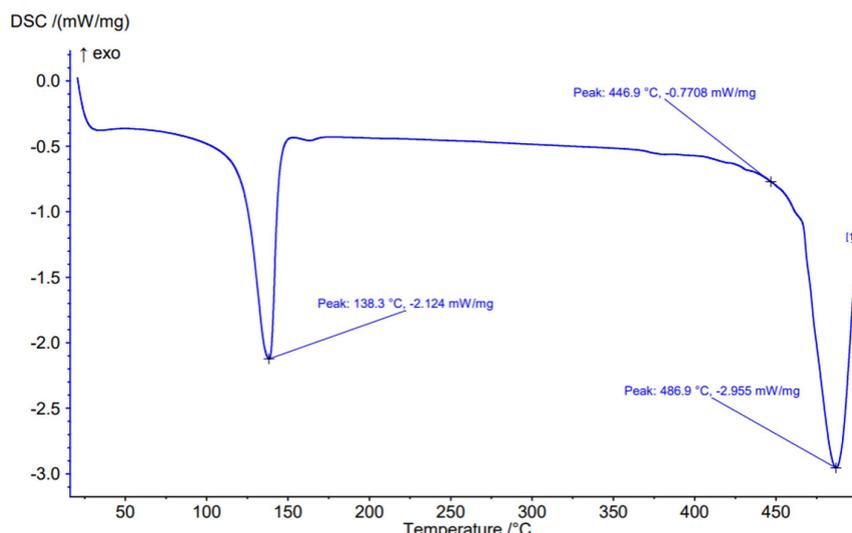**Fig. 6.** ATR-FTIR spectrum of PP Moplen HP456J
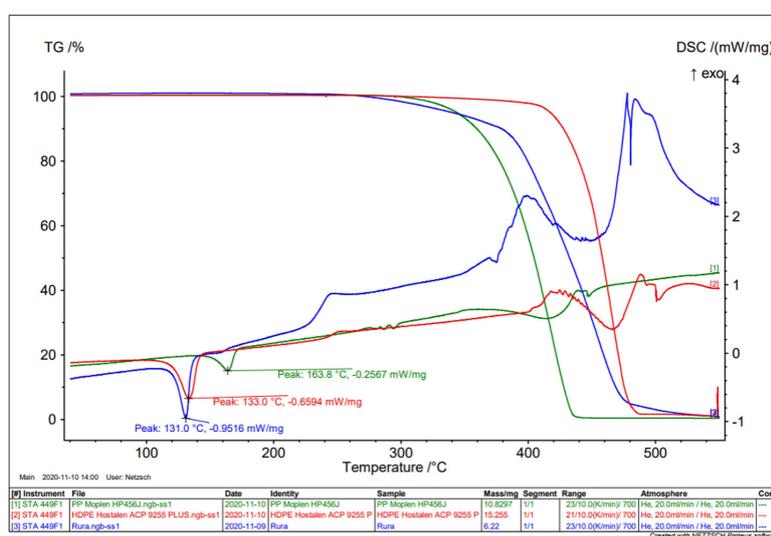
23

Fig. 7. DSC curve of HDPE regranulate

Fig. 8. TG curves of the tested samples

polyethylene during the extrusion process. The phase change temperature of polypropylene is 163.8°C and is not visible on the TG curves in both tested polyethylene samples. The only dissimilarity that can be read from the TG curve are significant differences in the decomposition temperature of the tested materials. For recirculated polyethylene coming from the pipe it is about 280°C (maximum 400°C), while for non-recirculated PE it is about 420°C (maximum 460°C). The corresponding value for PP is 320°C (max. 430°C). Quiet slow mass reduction in the sample of recycled HDPE in the temperature range of 280–400°C, may indicate the presence of organic contaminant in the regranulate. In comparision to the unprocessed HDPE the initiate temperature of the dynamic mass reduction is lower, which

might be caused by passed thermo-mechanical poliethylen's degradation during preminal use and futher processing.

CONCLUSION

As a result of the research, possible causes of cracks in stored pipes made of recirculated HDPE has been determined. The analysis of cracks surface revealed their fatigue character in the area of cracks initiation area. Once the increase in distance from the initiation area take place, we can consider the "quasi-brittle" phenomenon present. It has been found that the differences in the density and the melt flow index depending on the distance from the outer surface of the pipe, reaches several percent. Based on the conducted

spectroscopic, thermogravimetric and differential scanning calorimetry studies, none of the expected plastics contamination, mostly PP, has been found in recirculated polyethylene. Futhermore, the expected decrease in the temperature of phase transformations and the beginning of decomposition of polyethylene after recycling, compared to the original one, were visible. In the undertaken study, the fatigue nature of pipe cracks, in the absence of significant quantitative contamination with other materials, may be the result of structural differences in the pipe wall proves increased roughness of inner surface of the pipe and the presence of any porous structures, which could cause micro-cracks to propagate when stored under variable load and temperature conditions.

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