EFFECT OF CARBON BLACK DISTRIBUTION ON POST YIELD DEFORMATION PROPERTIES OF POLYETHYLENE PIPES FOR WATER TRANSPORT

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SHORT SUMMARY

Pipes with inhomogeneous CB distribution showed 80% less elongation than pipes with homogenously distributed CB. Polymer domains with less or no CB (windows) showed delamination from the polymer matrix as the material elongated, finally leading to fracture much earlier than expected.

KEYWORDS

polyethylene pipe, carbon black distribution, tensile properties, fracture surface

ABSTRACT

In this study, we investigated the effect of carbon black distribution on the degradation of mechanical properties of high-density polyethylene in the form of plastic pipes used in water distribution networks. Polyethylene pipes with similar carbon black concentrations but different carbon black distributions were produced with industrial scale compounding and extrusion equipment. Tensile specimens were directly prepared from extruded pipe samples and elongated to fracture at different strain rates. Carbon black distributions of bulk samples and fracture surfaces were investigated using stereo and scanning electron microscopy (SEM). It was found that the carbon black distributions, fracture surfaces and fracture modes were significantly different in these pipes. Although the yield properties were similar, the post-yield properties of samples were significantly different, dramatically decreasing with the carbon black distribution. Pipes with a certain level of heterogeneity in the carbon black distribution showed ductile and brittle fractures in the same fracture plane, whereas homogenous black and natural polyethylene (without carbon black) showed ductile fractures only.

Keywords: polyethylene pipe, carbon black distribution, tensile properties, fracture surface

A more detailed version of this work was previously published as "S. Deveci, N. Antony, B. Eryigit, Effect of carbon black distribution on the properties of polyethylene pipes part 1: Degradation of post yield mechanical properties and fracture surface analyses, Polym. Degrad. Stab. 148 (2018) 75–85.

This work was also published at 19th International Plastic Pipe Conference, Las Vegas, USA, 2018.

INTRODUCTION

Polyethylene has been a material of choice for many plastic pipe applications due to its optimum short- and long-term mechanical properties, easy processability and weldability. Carbon black (CB) remains the perfect and most economical solution to prevent photodegradation of polyethylene due to UV light exposure.

The efficiency of CB with respect to preventing the photodegradation of polyethylene in sunlight depends on the CB type, particle size, concentration and dispersion [1–3].

Insufficient dispersion and distribution of CB in polyethylene pipes are likely to occur if CB and natural polyethylene material are insufficiently mixed. This results in areas with lower CB contents appearing as light and dark swirls in microscopic images; these are commonly known as 'windows'. These windows are mostly a result of poor mixing of CB and natural polyethylene material in a single screw pipe extrusion line without a proper screw design and necessary mixing elements [4]. To prevent this mixing problem, the use of black pre-compounded polyethylene material for plastic pipe production is required by ISO 4427-1 [5]. It was reported that these black and white swirl patterns change direction at the butt fusion interface and camouflage lack of fusion, if present, in this region during non-destructive testing of polyethylene joints [6]. Recently, some brittle failures were observed in polyethylene pipe joints between pipes with insufficient CB distribution [7]. The role of CB dispersion in the initiation and propagation of cracks in polyethylene remains unknown [8].

It is now of great interest to clearly understand the effect of the CB distribution on the mechanical properties of polyethylene pipes and joints. Reports on this subject are very limited. In this article, we aim to show how the CB distribution affects the mechanical properties of polyethylene pipes by preparing plastic pipes with controlled CB distributions, containing low, medium and high levels of windows, and performing tensile tests on the pipes and analysing the fracture surfaces of these samples by stereo and scanning electron microscopy (SEM). The effect of inhomogeneous CB distributions on the butt fusion joint integrity and slow crack growth properties of plastic pipes will be discussed in subsequent articles.

MATERIALS AND METHODS

High-density polyethylene powder was collected from a polymerisation reactor. This was compounded with antioxidants and CB masterbatch (CBMB) containing 40% CB and 60% carrier resin to produce a black pre-compounded (ready-made) material using a counter-rotating continuous mixer (Kobe Steel, Japan). The same powder was also compounded with antioxidants but without CBMB to produce a stabilised, non-pigmented polyethylene compound (NPC).

Pipes with an outer diameter of 110 mm and a wall thickness of 22 mm were produced with a single screw extruder with a screw diameter of 60 mm and a length-to-diameter ratio (L/D) of 33 (Reifenhäuser, Germany). A four-channel spiral die with precise heating control via seven different heating zones was used to shape the pipe. The black pre-compound (or ready-made compound) was used to produce reference pipes with no windows (Sample 1). A dry mixture of NPC and CBMB was prepared with a tumbler mixer. The mixture was then used to produce pipes with different CB distributions (Samples 2, 3 and 4) by changing the extruder output and keeping all other parameters constant.

Descriptions of the pipe samples are given in Tables 1. The temperature profile of the barrel was kept constant, while the screw speed was changed to obtain different residence times and thus different levels of CB distribution.

The CB contents of all the pipe samples were analysed by pyrolysis under a nitrogen stream in a tube oven at 600°C according to ASTM D1603 [9]. Melt flow rates (MFR) of pellets and pipe samples were measured according to ISO 1133 [10] with a computerised melt flow indexer (Goetfert, Germany) at 190°C and 5 kg (MFR5) and 21 kg (MFR21). The flow rate ratio (FRR) was calculated as the ratio of MFR21 to MFR5. Densities of samples were measured with a density balance according to ISO 1183-1, Method A [10].

Table 1: Description of pipe samples.

Sample No	Sample Description	Material	Note
Sample 1	Reference Sample HE3490LS	Pre-compounded	Extrusion speed: 115 kg/h, 100%
Sample 2	High level of Windows	NPC + CBMB mixture	Extrusion speed: 115 kg/h, 100%
Sample 3	Medium level of Windows	NPC + CBMB mixture	Extrusion speed: 95 kg/h, 80%
Sample 4	Low level of Windows	NPC + CBMB mixture	Extrusion speed: 70 kg/h, 60%

The CB dispersion and distribution of pipe samples were measured for 15 µm thick sections, which were microtomed perpendicular to the pipe axis (cross-flow direction using a stereo microscope in transmission light mode (Carl Zeiss, Germany). SEM analysis was carried out with an FEI Quanta 250 FEG SEM instrument.

ISO 527-2 Type 1B tensile specimens were milled directly from pipe samples with a CNC milling machine (IPT, Germany) for tensile testing at a test speed of 25 mm/min and an initial grip-to-grip distance of 115 mm according to ISO 6259-3 [10]. Another set of samples were milled to ISO 8286 Type 3 standard for high-speed tensile tests at 250 mm/min with a 40 mm initial grip-to-grip distance. High-speed tensile test specimens were used for fracture surface analyses.

Tensile tests were conducted with a universal tensile testing machine (Zwick, Germany) equipped with a 50 kN load cell with a mechanical wedge grip. Crosshead displacement with an accuracy of 1 µm was used for nominal strain measurement. Tensile tests on pipe samples were repeated at least five times. All specimens were conditioned at 23°C and 50% RH for at least 24 h after specimen preparation.

RESULTS

The CB content, MFR and density of each sample are given in Table 2. All pipes showed very similar values. However, the CB content differed slightly for Samples 2 to 4 due to the dry mixing process.

Table 2: CB content, MFR and density of materials and pipe samples.

Materials	CB Content	MFR5	MFR21	FRR 21/5	Density
	wt%	(g/10 min)	(g/10 min)		(g/cm3)
Pre-compound	2.11	0.27	8.76	32.4	0.96
NPC	Not measured	0.27	8.77	32.5	0.95
Sample 1	2.11	0.25	8.07	32.3	0.9603
Sample 2	2.26	0.27	8.09	30.0	0.9607
Sample 3	2.40	0.26	7.99	30.7	0.9606
Sample 4	2.37	0.28	8.26	29.5	0.9609

CB Dispersion and Distribution

The CB dispersion and distribution in samples prepared from cross-flow directions are given in Figures 1. The CB dispersion in all four samples was found to be good; that is, no CB agglomerates of size >30 µm were seen in any of the samples. However, the distributive mixing was seen to be very different.

Sample 1 showed no windows that did not contain CB, although it exhibited a few spots that did not contain CB. According to ISO 18553 [11], the visual rating of the appearance of Sample 1 was A1/A2, which is well within the acceptable range.

Sample 2 showed the worst level of distributive mixing among the four samples. A high level of windows was seen in this sample. The width of the most prominent windows was in the range 200–300 µm. According to ISO 18553, the visual rating of its appearance was between C1 and C2.

Sample 3 also showed a high level of windows; however, the concentration of windows per unit area was slightly lower, and the windows were thinner compared to Sample 2. The width of the most prominent windows was in the range 80–140 µm. The visual rating of its appearance was closest to C1.

Sample 4 showed fewer windows than Samples 2 and 3. The width of the most prominent windows was in the range 50–100 µm. The visual rating of its appearance was between B and C1.

Tensile Tests on Pipe Samples

An overlay of engineering stress–strain curves at 25 mm/min (strain rate of 13 s^{-1}) for representative specimens selected from each sample is given in Figure 2. Samples 1, 3 and 4 showed the standard behaviour of a tensile curve for a high-density polyethylene material: (a) elastic region (linear increase in stress with strain), (b) a yield point (the first maximum stress in the stress–strain curve), (c) strain softening (an immediate decrease in stress), (d) natural drawing (constant stress versus strain), (e) strain hardening (linear increase in stress by increase in strain) and finally (f) breaking of the specimen.

Yield and post-yield properties of all samples are given in Table 3. The yield properties of the samples appear to be similar. Considering the physical properties of the samples given in Table 3, this is expected as the yield properties of polyethylene materials are exclusively related to the density of the material [12]. On the other hand, the post-yield properties of the samples showed significant differences.

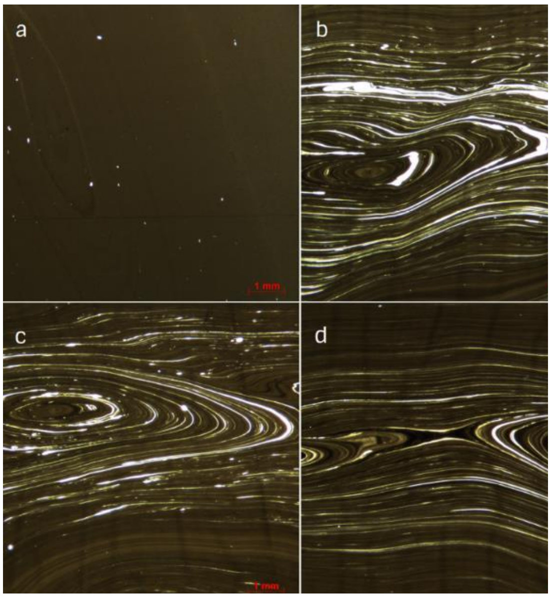

Figure **1:** Microscopy images of 15 µm slices (cross-flow) taken from pipe specimens: (a) Sample 1, (b) Sample 2, (c) Sample 3 and (d) Sample 4

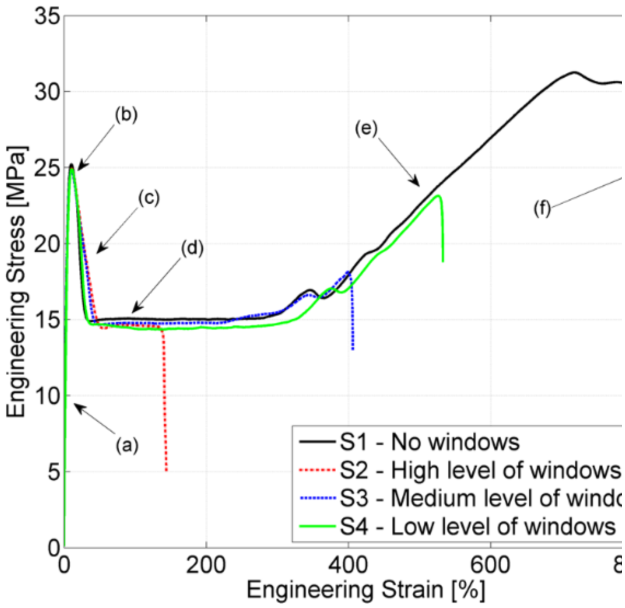

Figure 2: Engineering stress–strain curves of samples elongated to fracture at a test speed of 25 mm/min.

Table 3 Tensile test (25 mm/min) on pipe samples.

Samples		Yield Stress	Yield Strain	Stress at Break	Nominal Strain at Break
		MPa	%	MPa	%
S1	Average	25.2	10	30.1	800
	Std. Dev	0.1	0.1	0.3	70
S2	Average	25	10	15.6	270
	Std. Dev	0.5	0.1	2.5	160
S3	Average	24.9	10	19.8	430
	Std. Dev	0.3	0.1	4.2	130
S4	Average	24.9	10	25.7	670
	Std. Dev	0.3	0.1	6.3	220

Observations during the tensile experiments showed that necking in Sample 1 propagated all the way through the specimen, and material flow into the necking zone through the neck shoulder reached the gripping area. After passing through yielding, strain softening, cold drawing and strain hardening regions, once the thickness of the specimen at the gripping area decreased to a level at which the mechanical grips could no longer hold the specimen, the specimen failed at 740% nominal strain without a real break but due to slippage from the grips. This value is well above the elongation value required by ISO 4427-2 [13], which is a minimum of 350%. However, Sample 2, with the same molecular and physical properties as Sample 1, as given in Table 3, failed at significantly lower strains of 270% on average, after showing significantly reduced cold drawing with no strain hardening behaviour. Samples 3 and 4 showed post-yield properties that were better than of Sample 2 but inferior to those of Sample 1. On average, Samples 3 and 4 met the minimum elongation required by ISO 4427-

2, but one can understand from the large standard deviation that some specimens showed elongations below 350%. This behaviour is attributed to the inhomogeneous mixture of CB and the polymer matrix, especially when large windows are present, which indicate discontinuity in the physical properties of the CB polyethylene composite structure.

Apparently, these windows are the source of stress concentrations, especially at their borders. This phenomenon is observed during the tensile tests in the present study. In Figure 3a, a tensile specimen is shown before the test. The white arrows indicate windows that are visible at the surface of the specimen. Figure 3b shows the same specimen at 20% elongation after yielding. At this stage, many other windows became visible to the naked eye due to stress whitening phenomena. The borders of windows that exhibit discontinuity in physical properties become sharper as the material elongates (Figure 3c). At some point, microcracks at the sharp edges of the windows are observed due to poor interface bonding (Figure 3d) and higher stress concentration. These microcracks further developed with increasing strain and finally led to macroscopic failure as shown in Figure 3e.

Analyses of Fracture Surfaces

Figure 4 shows light microscopy images of fracture surfaces for all samples at a displacement rate of 250 mm/min. A typical fracture surface of polyethylene at high elongation was observed for Sample 1. Sample 3 and Sample 4 also showed similar fracture behaviour to Sample 1, with Sample 3 showing tiny windows visible to the naked eye (Figure 4(c), red arrows).

Fracture surfaces of samples indicated that an inherent stress concentration point initiated the fracture at the mid-section of the specimen (Figure 4(a), Zone A), followed by propagation through fibrillated wedges (Figure 4(a), Zone B) to the failure.

Sample 2 shows a complete ductile failure at the macroscopic level associated with the inhomogeneous structure of the matrix due to poor mixing of CB, which is clearly visible in Figure 4(b), in which windows are clearly visible to the naked eye (red arrows).

SEM images of black areas (Figure 5b) showed similar packed crazes tearing to failure. SEM of the fracture surface of Sample 2 in the windowed area (Figure 5a) shows brittle features associated with a highly fibrillated structure, which are similar to those observed during slow crack growth in polyethylene.

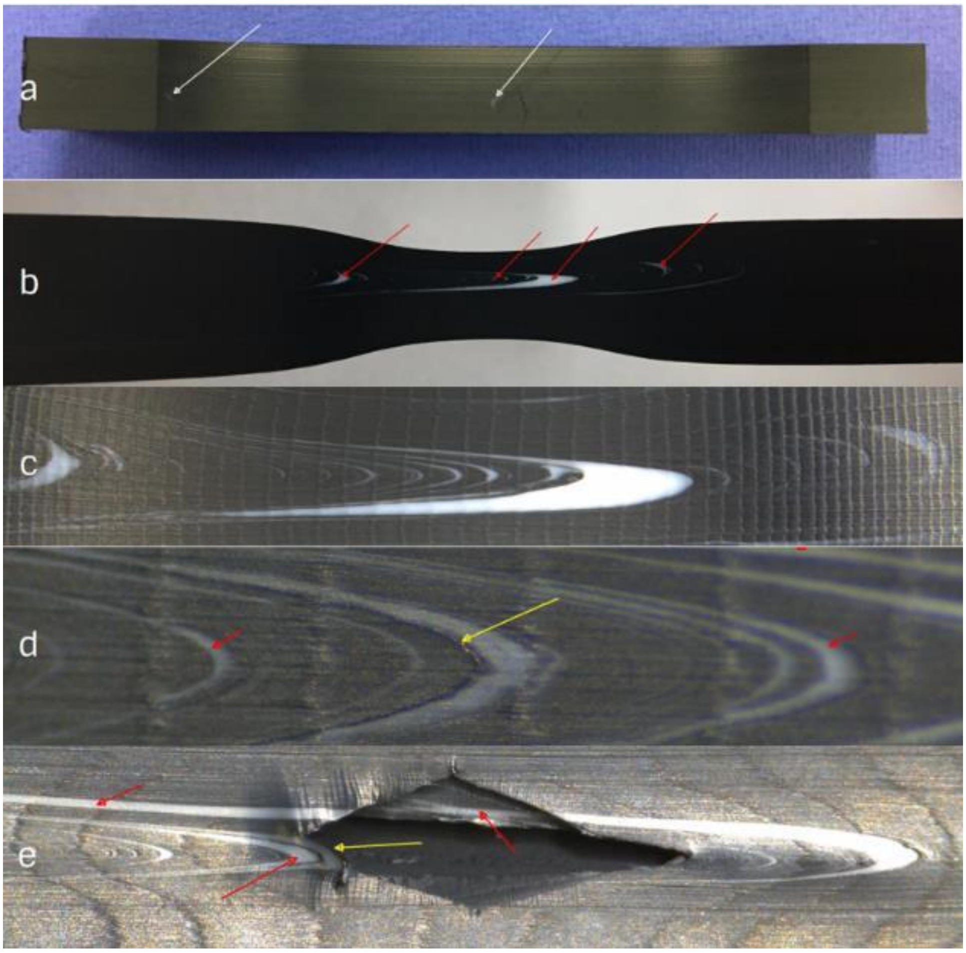

Figure 3: Propagation of windows to fracture: (a) tensile specimen before testing—white arrows indicate 'windows' visible to the naked eye; (b) tensile specimen after yield (20% elongated)—red arrows show windows curled towards the tensile direction; (c) tensile specimen (40% elongated); (d) interface separation at the edge of windows marked with a yellow arrow; (e) fracture occurrence at the interface separation (yellow arrow).

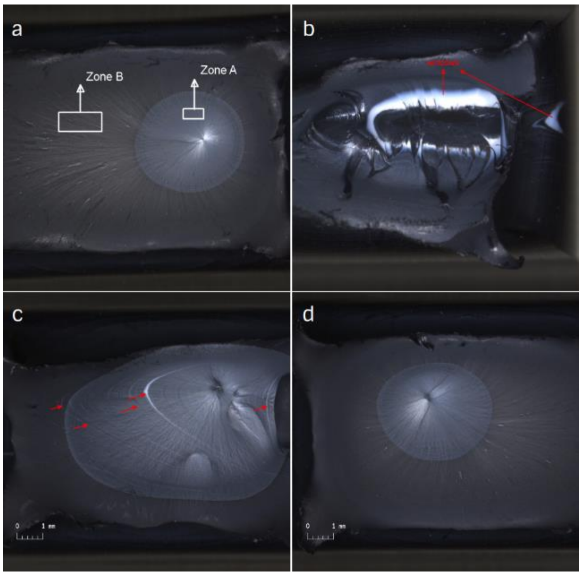

Figure 4: Optical images of fracture surfaces: (a) Sample 1, (b) Sample 2, (c) Sample 3 and (d) Sample 4.

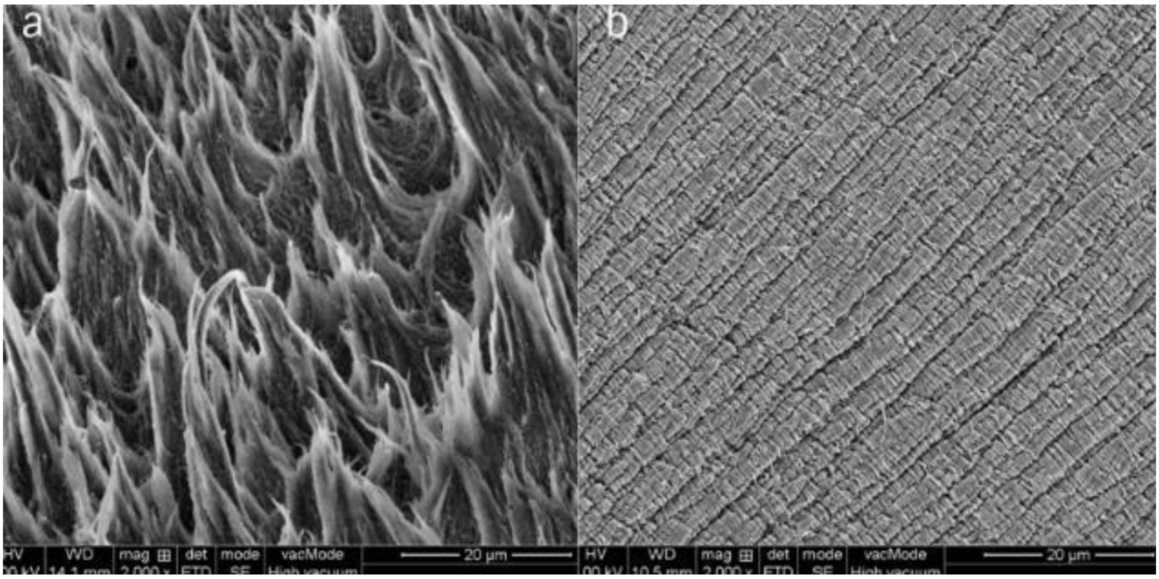

Figure 5: SEM images of fracture surfaces of (a) windowed area of Sample 2 and (b) black area of Sample 2.

CONCLUSIONS

In this study, four plastic pipes made of the same polyethylene material with different CB distributions were produced by single screw extrusion. These pipes were elongated to fracture by means of tensile testing. Fracture surfaces of pipes were investigated with light microscopy and SEM.

It is observed that the CB distribution in in-line compounded pipes was inadequate. Increasing the residence time by reducing the production speed helped to improve the CB distribution; however, even with a 40% decrease in throughput, the CB distribution in in-line compounded pipes did not match that in pre-compounded pipes.

A significant decrease was observed in the post-yield properties of polyethylene pipes with an insufficient CB distribution, whereas the yield properties were not affected. In-line compounded pipes showed 80% less elongation than pre-compounded pipes. Polymer domains with less or no CB (windows) showed delamination from the polymer matrix as the material elongated, finally leading to fracture much earlier than expected. A highly heterogeneous CB distribution was observed on fractured surfaces of pipes produced with the in-line production method. Interestingly, polymer domains with no CB within the polymer matrix showed brittle failure once elongated to fracture, although the corresponding natural material shows completely ductile failure when it is strained as a single component.

Finally, we have reported the significance of CB homogeneity with respect to the mechanical properties of plastic pipes. However, these are connected with proper fittings. It is also of great interest to design engineers to understand the integrity of plastic pipe welds. Therefore, in a subsequent article, the effect of the CB distribution on the mechanical integrity of polyethylene butt fusion joints will be reported.

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