

FIELD FAILURES IN POLYETHYLENE GAS PIPES: ANALYSIS VERSUS LIFETIME PREDICTION.

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Eight excavated HDPE pipes of the 1st generation along with 3 modern PE types have been investigated. Comonomer content as a function of molecular weight of the old HDPE pipes appeared to be non-optimal with respect to the modern PE types. Further unwanted effects in the old pipes were : particles, oxydation, inhomogeneity. Premature failures were due to previously unexpected effects of external point loadings.

INTRODUCTION.

Polyethylene has been used for gas distribution in the Netherlands since the early 1970s. Recently, an increasing number of premature failures has been encountered in the so-called first generation (HDPE) materials, which have been installed before 1975. These failures always took place at positions, where local stresses (external point loadings) were present. Such point loadings lead to an indentation of the outside surface of the pipes and consequently to tensile stresses on the inside pipe surface. Often, the cracks initiated at or near the inside surface.

In those ISO requirements which were valid at the time of installation, the influence of additional external point loadings (besides the normal tangential wall stress due to the gas pressure) was not taken into account. Such tests are now considered by VEG-Gasinstituut [1] and others. Point loading tests place special emphasis on the quality of the inside surface, which in turn will be dependent on materials quality, molecular structure, extrusion quality, oxydation effects, homogeneity of the material and the presence of particles which may act as crack initiators. All these aspects will be mentioned in this contribution.

Eight HDPE pipes, denoted 1 until 8, were excavated from locations where premature failures had occurred and investigated using different techniques. For comparison, three more modern PE gas pipe materials, denoted 9, 10 and 11 were investigated as well. Material 9 is a traditional HDPE material which is still being used, 10 is a MDPE and 11 is a so-called 3rd generation HDPE material.

Description of fracture surfaces.

The fracture surfaces have been investigated using both Optical and Electron Microscopy. A typical brittle crack surface at low magnification is shown in Figure 1. At higher magnification (Figure 2), particles are visible in the fracture surface.

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Figure 1. Optical micrograph (8x) of pipe 1 failed in a brittle manner in field practice.

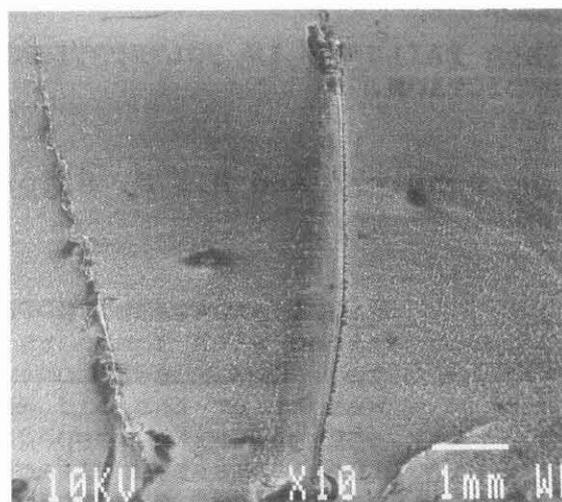


Figure 2. Enlargement of central portion of Figure 1.

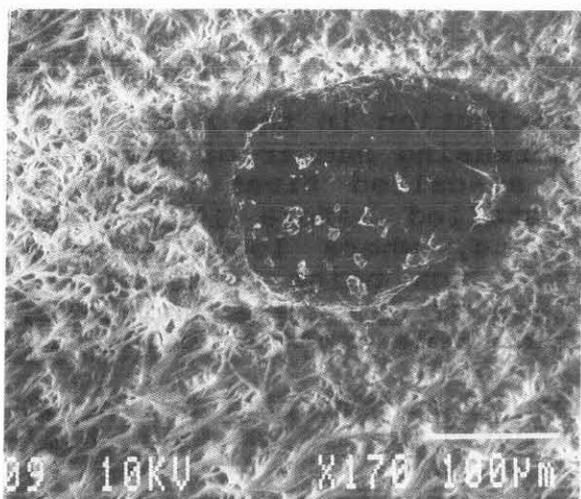


Figure 3. Enlargement of previous Figure (170 x).

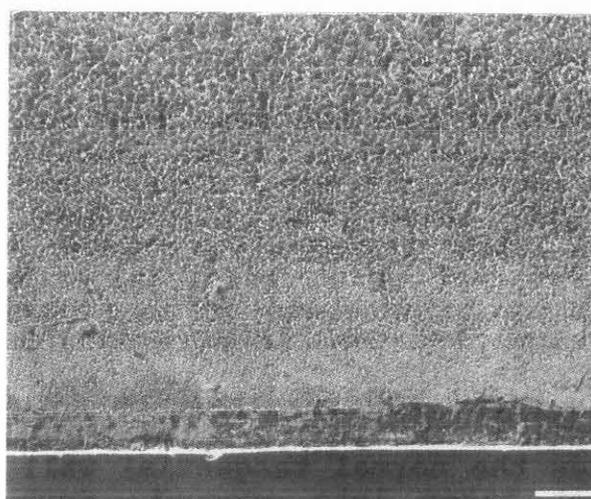


Figure 4. Inside surface of pipe 1 (75 X).

Figure 3 shows such a particle at a still higher magnification. The fact that there is poor adhesion between particle and PE matrix is typical. It is clear, that such particles may act as stress concentrators, leading to (premature) crack initiation. The crack surface images shown in Figures 1, 2 and 3 are similar to what has been described in literature [2-5].

Oxydation of pipe surfaces.

Figure 4 shows another phenomenon which is sometimes present in old HDPE pipes. On the inside of the pipe a layer with different structure is visible. This may be a layer of oxydised PE material, originating from non-optimum extrusion conditions. Probably, the PE material was extruded at a too high temperature and the inside surface of the pipe (which always stays hot longer than the outside surface) was oxydised.

Infrared Spectroscopy was also used to investigate these oxydation effects. Figure 5 shows the infrared spectra of the inner pipe

surfaces of pipes 3 and 2 respectively. Pipe 2 is slightly oxydised on the inside surface, pipe 3 is not. Pipe 2 (and only pipe 2) is also oxydised in the middle of the wall. It seems, that this pipe contains (some) recycled, oxydised material. Whether oxydation was present on the inside surfaces of the pipes is shown in Table 1.

Poor homogeneity.

Figure 6 shows an Optical Micrograph of the middle of the wall of the excavated pipe 1 after microtoming. The difference in color intensity is caused by an inhomogeneous carbon black distribution. This is an aspect of non-optimum extrusion quality as well.

In pipe 6, another unwanted effect was noted. This pipe appeared to contain local PVC impurities in the fracture surface, as was detected using an X-Ray Spectrometer connected to the Scanning Electron Microscope. Most probably, the extruder used for producing PE pipe 6 was used earlier for PVC pipe production, without proper cleaning procedures.

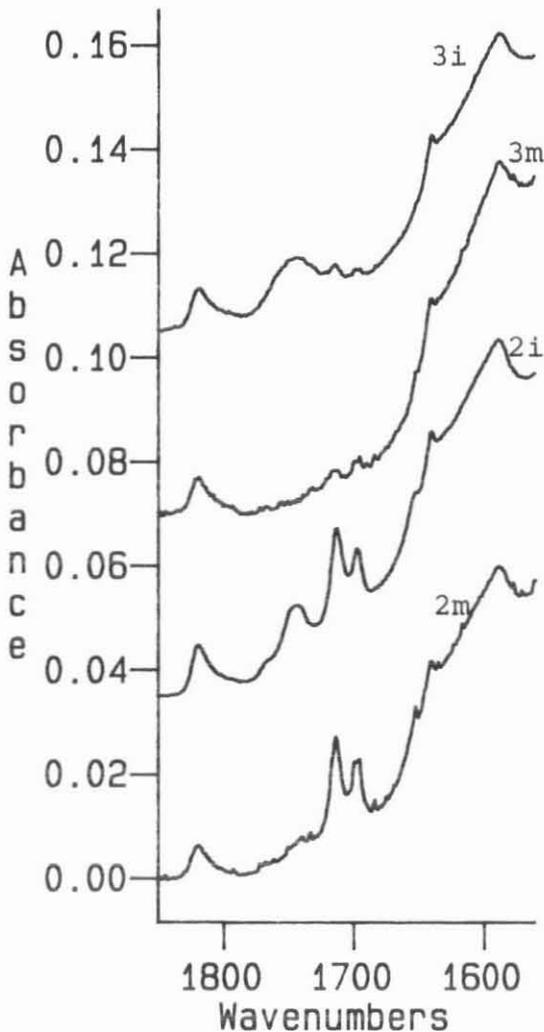


Figure 5. Infrared spectra of pipes 2 and 3; i: inside surface, m : middle of wall thickness.

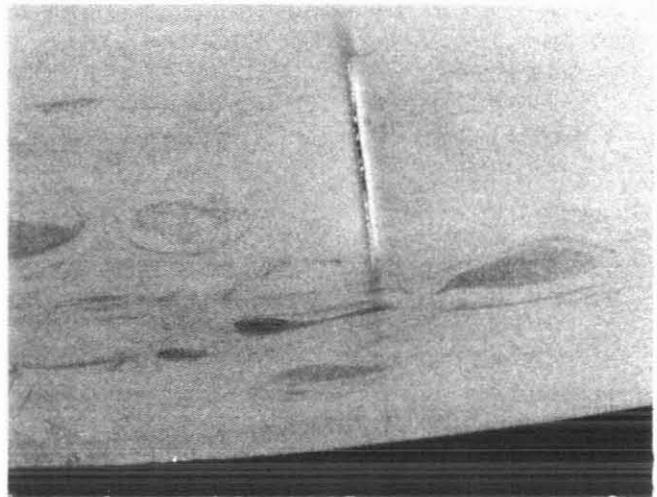


Figure 6. Optical Micrograph of pipe 1; microtomed.

Stress rupture tests on 1st, 2nd and 3rd generation materials.

The eight excavated pipes were tested using burst pressure tests at 80 °C. Pipe lengths were selected, in which no visible defects could be detected. For one material (# 7), not enough pipe was available for performing these tests. A stress level of 3 MPa was used, except for material 1, for which a much broader range of stresses were investigated, up to 15 MPa. Some results are given in Table 1, along with results on the three more modern, 2nd and 3rd generation PE materials. The failure times given for pipes 1 until 8 are averages of 3 values, the values given for pipes 9, 10 and 11 are averages of 4 values. Table 1 also shows, whether or not the inside surface of the pipes was oxydised. Degree of branching (number of ethyl or butyl groups per 1000 carbon atoms) was also measured with FTIR spectroscopy on samples from the middle of the wall.

Table 1. Stress rupture tests at 80 °C and FTIR measurements on excavated HDPE pipes and on three more modern PE types.

Material	Failure times (h) at		Oxydation of inner surface	Degree of branching (#/1000C)
	3 MPa	4.6 MPa		
1	707	90	no	2.2
2	64	-	yes	1.3
3	400	-	yes	2.2
4	354	-	no	2.2
5	199	-	yes	2.2
6	31	-	yes	2.3
7	-	-	yes	2.6
8	537	-	yes	2.2

9	-	401	no	2.7
10	-	>20,000	no	2.1
11*	-	3,805	slight	5.1

* butyl branches, all other materials : ethyl branches.

It was checked whether the old pipes meet the requirements which were valid at the time of their installation. It appears, that only materials 2 and 6 did not meet these old requirements of 170 hours at 3 MPa. For material 2, an explanation presents itself; it contains oxydised material (also in the middle of the wall) and has a low branch content [6]. Both effects contribute to poor lifetime expectancy, although the latter probably more than the former. Pipe 6 probably has poor properties because of the local PVC impurities found in the fracture surface. Because the samples for Infrared Analysis and DSC (see below) were not taken near the fracture surface, no PVC impurities were detected by these techniques in pipe 6.

There appears to be no correlation between the presence of oxydation on the inner pipe surfaces and failure times. This is understandable, because the degrees of oxydation found in pipes 1 to 8 are low and are probably too low to have a significant effect.

Stress rupture data of the manufacturer of the excavated pipes are available [15]. Failure times at 3 MPa for this type of material lie between about 200 and about 1200 hours at 80 °C. It appears,

that on average, only a small part of the lifetime expectancy has been used up after more than 25 years of service life (Table 1). This leads to the conclusion, that indeed the premature failures are due to the previously unexpected effect of external loadings.

MOLECULAR STRUCTURE.

Above, effects of poor processing (oxydation, inhomogeneity) and of impurities (particles) have been discussed. Molecular structure is another important parameter needed to understand the differences in properties between different PE generations. This has been studied by fractionation.

Fractionation.

Since a number of years it is well known, that besides Molecular Weight Distribution, Comonomer Distribution and Comonomer Function (comonomer concentration as a function of molecular weight) are important molecular parameters as well. Temperature Rising Elution Fractionation (TREF [7,8]) and Direct Extraction Fractionation [9,10] are powerful techniques to measure these parameters respectively.

Lustiger [11], Ishikawa et al [12] and Brown et al [13] have proven, that the best materials, i.e. with the best resistance against slow crack growth, are those materials which have the

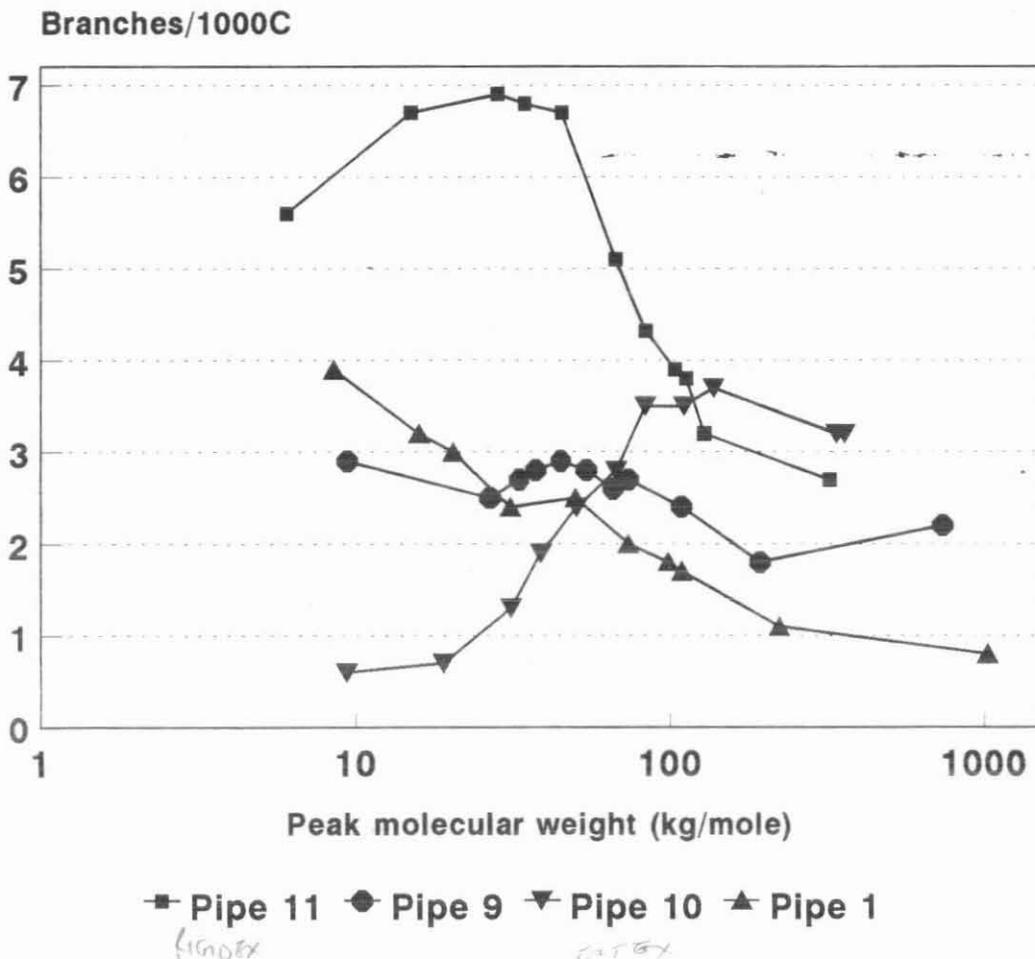


Figure 7. Comonomer Functions of 4 PE gas pipe materials.

highest comonomer concentration on the longest molecules. In this contribution the Comonomer Function of the excavated HDPE pipe 1 was determined and compared with those of more modern-type PE materials (9, 10 and 11).

The Comonomer Functions were determined after direct extraction fractionation according to Holtrup [9]. Ten or eleven fractions were obtained, fractionated according to differences in molecular weight. Of each fraction, molecular weights (Weight Average and Number Average Molecular Weights and Peak Molecular Weights) were measured by Dr. Godard of the Polymer Department of Louvain la Neuve University, Belgium, using GPC. ¹³C-NMR Spectroscopy was carried out by Dr. Frank Heatly of Manchester University, UK, to measure comonomer type and content of all fractions.

Figure 7 shows the Comonomer Functions of the excavated HDPE pipe, of a traditional HDPE, a traditional MDPE and of a so-called 3rd generation HDPE. The differences are very clear. It appears, that there is a straightforward correlation between the comonomer content of the molecules with a molecular weight above around 100 kg/mole and the burst pressure test results shown in Table 1. This is totally in accordance with expectations [11-13]. It appears, that Figure 7 combined with Table 1 shows an overall picture of the history of the development of PE pipe quality in the last 30 years. The improvements are impressive and Figure 7 shows the main reason for these improvements.

DSC analysis.

The Comonomer Functions of the 3 modern materials have been confirmed by the respective manufacturers. As far as the excavated pipes are concerned, the Comonomer Function is only known for material 1. Comments by the manufacturer for this material are not available yet. It has to be ascertained, whether this material is representative for all 8 excavated HDPE pipes. The stress rupture failure times are highest among the 8 excavated pipes (Table 1), although not significantly. A short term method was sought and found in Differential Scanning Calorimetry (DSC) analysis.

DSC measurements using normal procedures (10 °C/min cooling and heating) were carried out on all 11 PE materials in Table 1. To all samples, a thermal pretreatment was given which consisted of heating with 10 °C/minute to 170 °C and subsequent cooling with 10 °C/minute to room temperature. This ensured that the thermal history of all samples was destroyed.

The samples were taken from the middle of the wall. In previous investigations, it was noted that samples that contain a part of the inside or outside surface show different DSC curves, even after thermal pretreatments. This is probably due to oxydation or impurities. Even new pipes were found to contain impurities on the inside surface, which are capable of influencing the DSC results. To avoid these effects, the outside and inside surfaces of the pipes were carefully removed before the samples for DSC analysis were taken.

The differences in the results are much more clear in the cooling (crystallisation) curves than in the heating curves (Figure 8). It appears, that the 8 old HDPEs are very similar (except again material 2). There are no indications, that material 1 should not be representative for all 8 excavated pipes (except material 2). The excavated pipes are clearly different with respect to the 3 more modern materials. The last result supports the results of the fractionations (Figure 7).

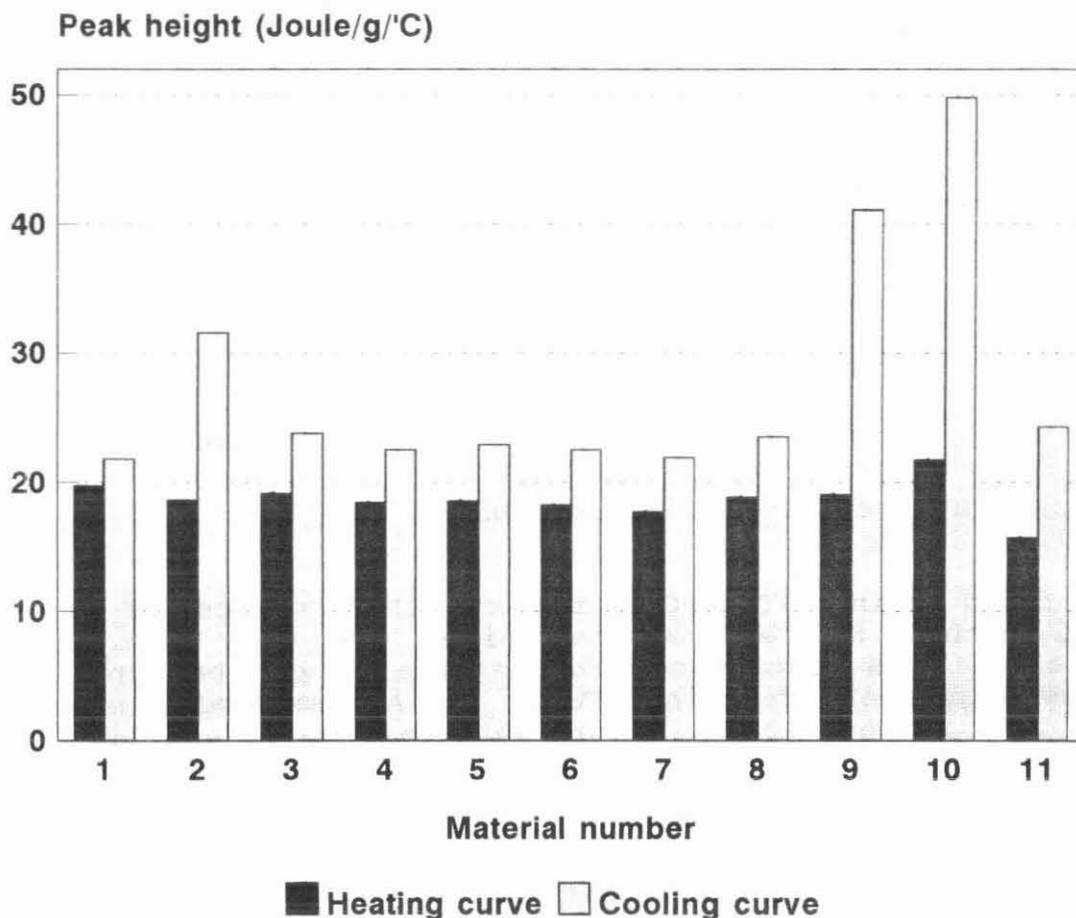


Figure 8. Peak heights of DSC curves (heating and cooling with 10 °C/min) of all 11 pipes.

SIS (DSC) analysis.

Above, it was concluded, that cooling (crystallisation) curves show much larger differences between materials than heating curves. This led us to place more emphasis on crystallisations during DSC measurements. A new DSC method can be used for this, the so-called Stepwise Isothermal Segregation (SIS) method, published by Kamiya and Ishikawa et al [14].

The SIS method is in fact a Stepwise Isothermal Crystallisation method, by which the sample is crystallised isothermally at different temperatures (Figure 9). After the SIS treatment, which is a thermal pretreatment, a split up melting (heating) curve is obtained (the so-called DSC-S curve), in which the different peaks reflect the different crystallisation steps during SIS. Figure 9 shows the temperature programme for the SIS method which uses 3 crystallisation steps; 2 isothermal and one dynamic.

The DSC-S curves of materials 1 and 3 until 8 are all the same. Figure 10 shows an example, of material 8. Material 2 is again clearly different. Materials 9, 10 and 11 deviate strongly from the excavated pipes.

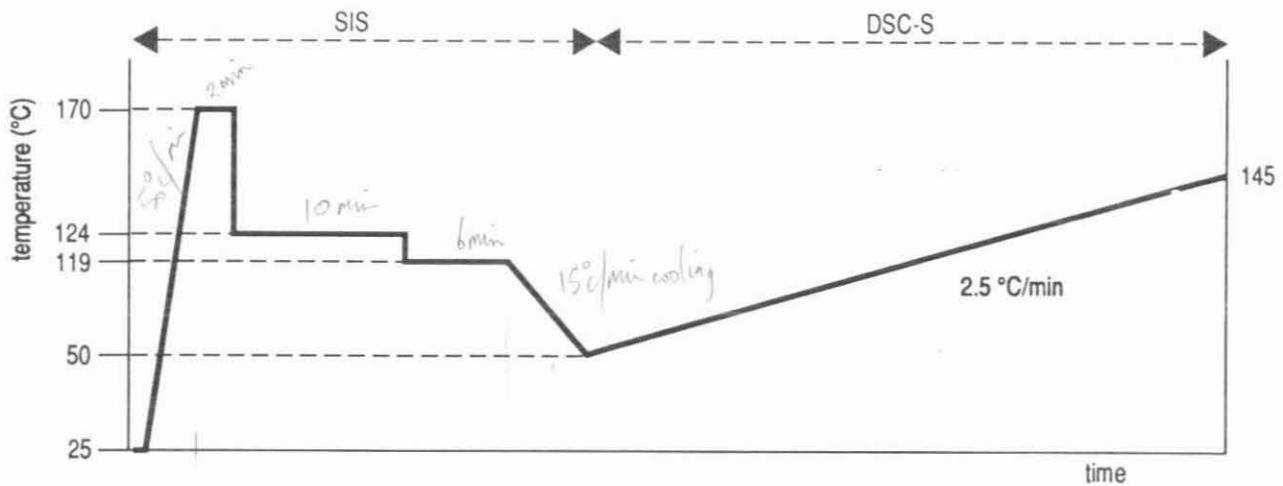


Figure 9. Temperature profile for SIS and DSC-S.

The results found using SIS/DSC-S support the results of the traditional DSC method : the excavated pipes are very similar, except material 2. This discrepancy for material 2 can be explained by its low degree of branching (Table 1). All excavated pipes are different with respect to the modern materials 9, 10 and 11.

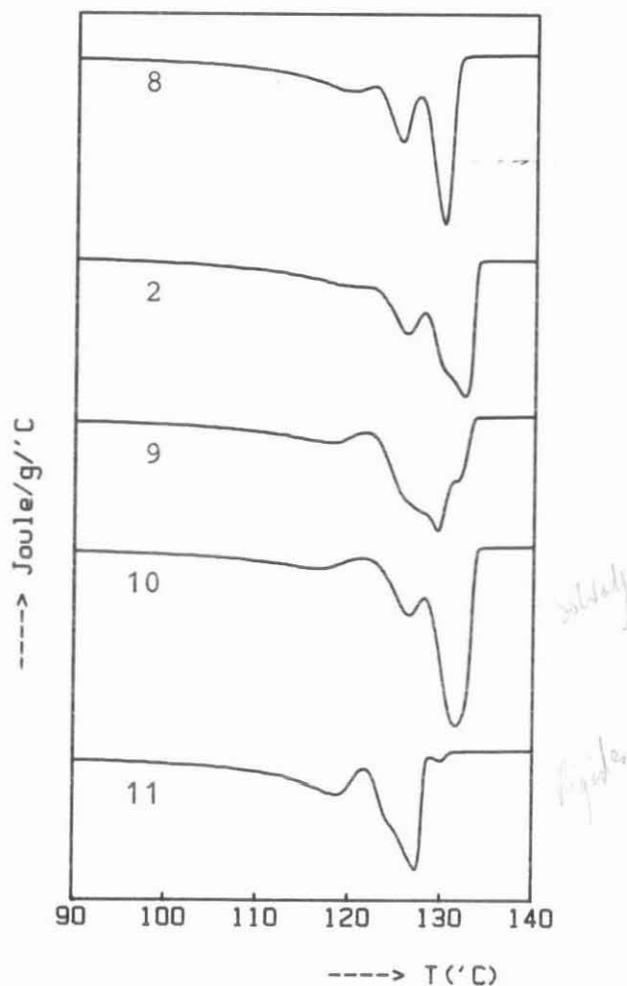


Figure 10. DSC-S curves at 2.5 °C/min. for different materials.

However, the SIS method is capable of finding much clearer differences between similar PE materials or between different batches of the same material than the normal DSC method. This is especially true with respect to the heating curves (Figure 8). SIS may be used as a quality control method. However it is not yet known, what the physical meaning of the different peaks in the DSC-S curves is, although it is save to say, that the fractions that make up these 3 peaks differ in crystallisability. This parameter in turn is dependent in a complicated way on molecular weight, comonomer content and Comonomer Function.

CONCLUSIONS.

- For lifetime prediction of pressure pipelines not only internal pressure should be taken into account, but external loadings as well.
- Molecular structure of 1st generation PE materials was not optimal with respect to the long-term performance. Since then large improvements in PE gas pipe grades have taken place, resulting in materials with excellent lifetime expectancy.
- Besides strong improvements in molecular structure, processing of pipes has improved as well. This resulted in a better homogeneity and the avoidance of oxydation.
- There is every reason for great confidence in the present PE types for gas distribution.

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