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INFLUENCE OF SOME AGEING FACTORS ON INSULATION OF CROSSLINKED POLYETHYLENE

1. Introduction

In the nearest future, crosslinked polyethylene will be applied as insulation of medium voltage power cables, produced by the Polish cable industry. In this connexion, the necessity arose to choose from among the types of polyethylene, proposed by the chemical industry, the optimum one for crosslinking and to estimate the quality of the chosen type from the point of view of exploatation properties.

The results of investigations, carried out previously by the authors, showed a strict connection of changes in dielectric properties of thermoplastic polyethylene with the changes in its physical-chemical properties Therefore, scanning microscopy and infrared spectrophotometry have been widely used to evaluate the quality of insulating cable materials as well as to select optimum types of cables, proposed by the producers, as a result of necessary technological changes.

2. Method of investigations

Three types of crosslinked polyethylene, denoted by symbols A, B and C were investigated. Samples in the form of plates 0,4 mm thick underwent thermal ageing at the temperature of 135°C at free access of air. A part of the samples underwent also thermal-water ageing in an ultrathermostat at the temperature of 90°C in a tap water bath.

Unaged samples and the aged ones as well, after 168, 336 and 504 hours of action of the thermal-water agent, were subjected to infrared spectrophotometric analysis and on using the scanning electron microscope the quality of the material was estimated. The SPECORD 71 IR spectrophotome-

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ter was utilized to carry out the IR analysis and changes in the spectrum in the range from 4 600 to 650 cm^{-1} (about 2,2 - 15 μm) were analysed.

Whereas the microscopic observations were performed on suitably prepared samples in accordance with the described method in [2]. The scanning electron microscope Jeol type, was used; it allowed to obtain up to 3000 times enlargements for this type of investigated material.

Measurements of the dielectric loss factor and of the dielectric constant were also performed for unaged and aged samples. On using Schering's bridge, type 2801 manufactured by Tettex tan δ and ε_w measurements were made.

3. Results of experimental investigations

Table 1 shows changes in the value of the dielectric loss factor and the permittivity for three investigated types of polyethylene. The values presented in the table are mean values of 10 measurements.

Table 1

Changes in tan δ and ε_w for different types of crosslinked polyethylene, caused by the influence of external factors

No	Type of polyethylene	Type of ageing	Time of ageing hours	tan δ x10^4	ε_w
1	A	-	0	4,0	1,89
2	A	thermal	168	6,8	1,94
3	A	thermal	336	9,7	2,02
4	A	thermal	504	19,8	2,09
5	B	-	0	2,8	1,94
6	B	thermal	168	3,9	1,98
7	B	thermal	336	8,2	2,04
8	B	thermal	504	11,1	2,08
9	C	thermal	0	6,0	1,98
10	C	thermal	168	8,4	1,99
11	C	thermal	336	10,0	2,02
12	C	thermal	504	21,0	2,18
13	C	therm.-water	168	9,2	1,99
14	C	therm.-water	336	27,1	1,99
15	C	therm.-water	504	43,0	2,06

But figure 1, shows changes in the dielectric loss factor versus the time of thermal ageing for polyethylene A, B and C and also versus thermal-water ageing time for polyethylene C.

The fastest increase in tan δ was stated at thermal-water ageing of samples. These results are analogical to the results obtained in the case of subjecting samples or sections of power cables with thermoplastic polyethylene insulation to the action of a thermal agent [1].

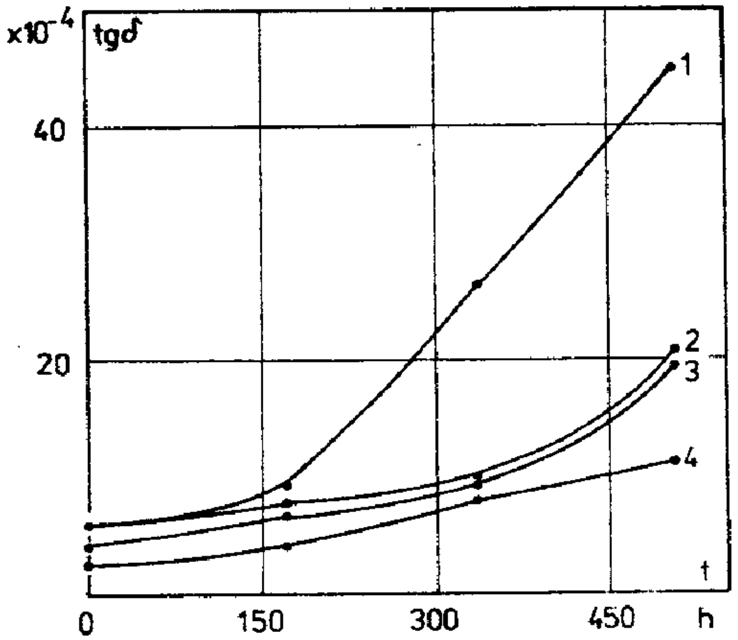


Fig.1. Changes in the dielectric loss factor versus the time of thermal ageing for polyethylene A(3), B(4), C(2) and also thermal-water ageing for polyethylene C(1)

It was also found that, for all the investigated types of polyethylene, the values of the permittivity were below 2. The strongest increase was observed at thermal ageing of the material.

When performing the spectrophotometric analysis, in order to check, if in case of recording time extension of a given infrared spectrum run an additional enrichment of the spectrum is not obtained, the time of recording one spectrogram was extended to 44 minutes. However, the obtained spectrograms did not stray in their run from the spectrograms obtained in the standard time of the IR spectrum recording.

It was found that the samples of polyethylenes B and C aged to a very differentiated degree, when influenced by the temperature of 135°C for some time (e.g. for 504 hours). It is visible when inspecting aged samples even with unaided eyes. Whereas the process of polyethylene A ageing occurs in the same way for the whole lot of investigated samples.

Ageing at the temperature of 135°C often causes deformation of the sample surface thus that it is impossible to carry on further investigations of dielectric properties.

Thermal action on polyethylene samples, in the case of ageing of a sample, placed between layers of a window-pane, makes random access of air to the surface of the sample impossible and therefore distorts the run of the material oxidation process.

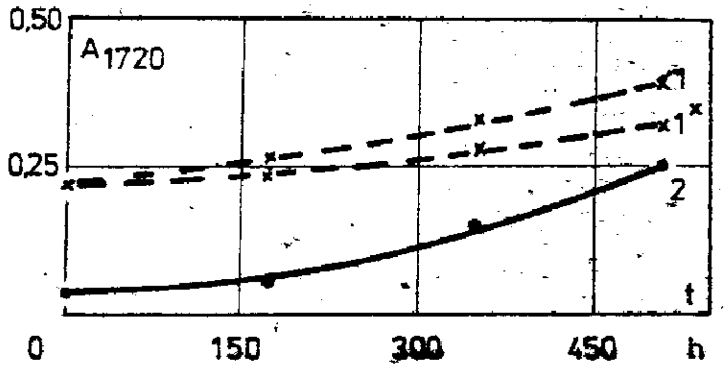


Fig.2. Changes in the intensity of 1720 cm^{-1} band versus time of ageing for polyethylene A(1) and B(2)

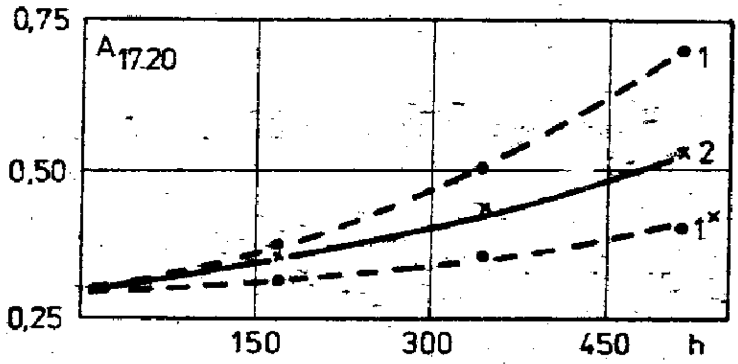


Fig.3. Changes in the intensity of 1720 cm^{-1} band for polyethylene C versus time of thermal (1) and thermal-water ageing (2)

As it has been proved in previous papers, among others in [5], the carbonyl band of minimum in the range of 1710–1730 cm^{-1} is one of the principal diagnostic bands of the occurring ageing processes, causing a change in the chemical structure of polyethylene. Figures 2 and 3 show changes in the intensity of this band versus time of ageing for polyethylenes A, B and C.

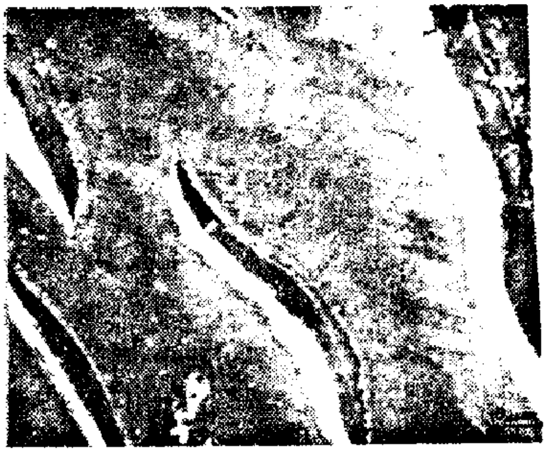
The smallest changes in intensity of the carbonyl band, caused by thermal action, were stated for polyethylene A. For polyethylene C the increase in A_{1720}, in some cases of thermal ageing, exceeds the changes in intensity of this band, which occurs at thermal-water ageing of this polyethylene.

Microscopic observations were performed on samples which were subjected initially to standard treatment [2]. This treatment included:

- etching of the sample surface in vapours of carbon tetrachloride (CCl_4) during 20–30 sec.,
- vacuum sublimation of the surface with silver.

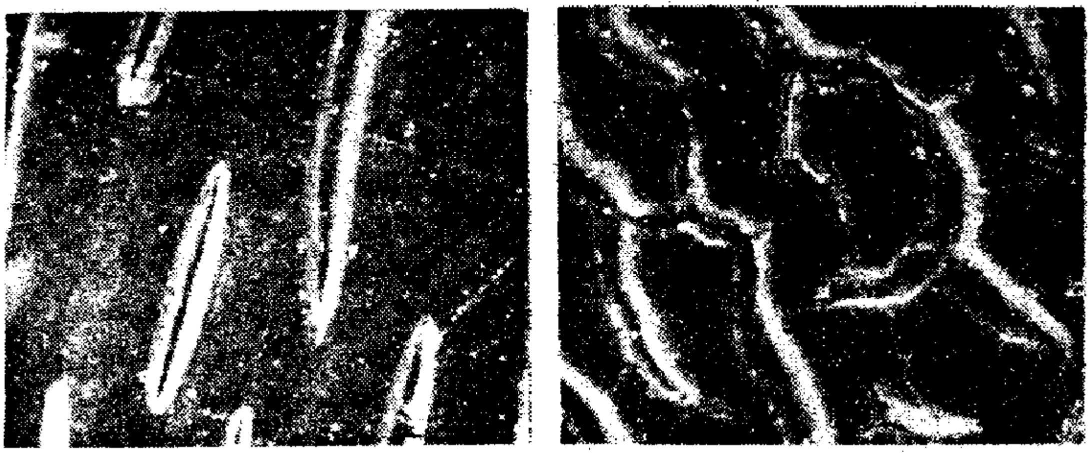
Figure 4 presents, as an example, typical surface of etched samples of polyethylenes A and B, prepared for investigations. The degree of cross-linking of these polyethylenes ranged from 65% to 75%. Whereas figure 5 is an exemplary microscopic image of the morphological structure of polyethylene C after 504 hours of ageing at the temperature of 135°C at random access of air.

a



b

Fig 4. The morphological structure of the unaged polyethylene A and B



A B

Fig.5. The morphological structure of the polyethylene C unaged and thermal aged

The degree of cross-linking of insulating polyethylene samples, was defined by means of the analysis of microscopic photographs. The echting agent, during the action on the surface of the sample, echted the cross-linked regions faster than the zones of the cross-linked polyethylene.On calculating the whole surface of the photograph, and then the surface of the sample non-cross-linked part, the degree of cross-linking of the investigated sample can be estimated.

Changes in the degree of crosslinking and also general changes in the morphological structure which occurred in the structure of the thermal and thermal-water aged polyethylene, can be observed by comparing the superficial structure of aged and unaged samples. In the aged samples of polyethylene more impurities were noticed. Micro-cracks, micro-spaces formed in the structure create a blurred image of the structure. Larger echted regions in aged samples testify the decrease in the degree of polyethylene crosslinking during thermal ageing. Changes in the degree of crosslinking of investigated types of polyethylene, as a result of thermal and thermal-water ageing, are marked diagrammaticaly in figure 6.

The investigations have proved that the best type of polyethylene from among the three investigated ones, is polyethylene A, in respect of resistance to thermal ageing. Changes in its structure, because of the influence the thermal agent, are the smallest. Changes in the dielectric loss factor also occur in the smallest range during ageing of polyethylene A, as compared with polyethylene B and C.

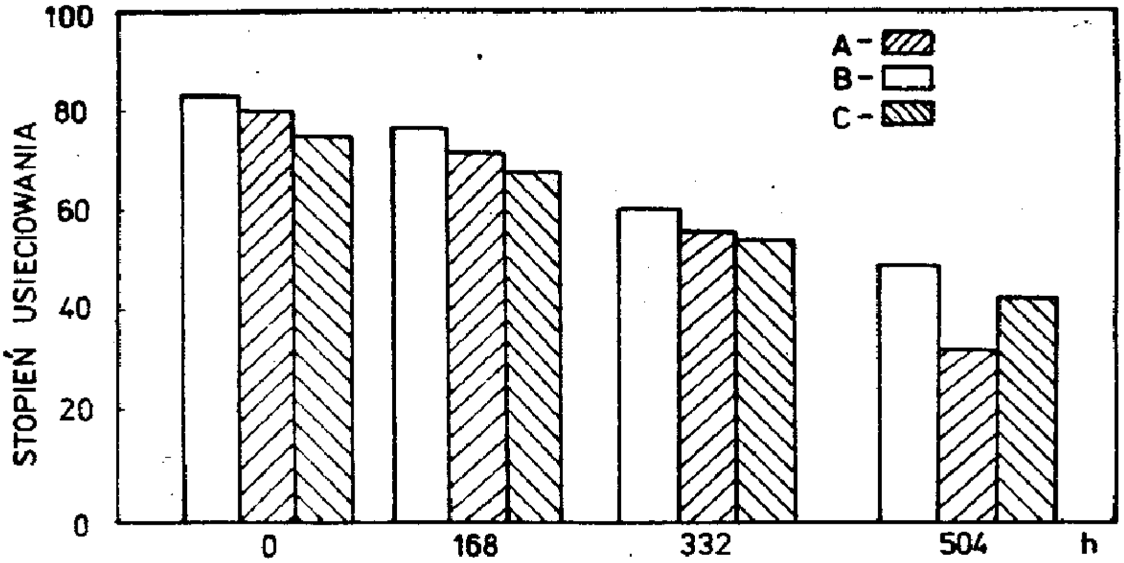


Fig 6. Changes in the degree of crosslinking of polyethylene

4. Summary

On the basis of carried investigations and observations, it was found that:

- scanning microscopy and infrared spectrophotometry are methods of investigations which allow to obtain many informations on the subject of the analysed insulating material,

- thermal ageing at the temperature 135°C seems to be a very strong criterion to estimate the properties of the crosslinking polyethylene, because it causes durable destruction of the morphological structure: during a relatively short time of thermal ageing (equal to 504 h.), the degree of crosslinking decreased in 50%,

- if carrying accelerated ageing at the temperature of 135°C to estimate the quality of the crosslinked polyethylene will still be required then ageing will require the elaboration of a method which maintaining random accees of air to the samples- will make distortion of their surfaces impossible,

- ageing at the temperature of 135°C influences unfavourably the morphological structure of the crosslinked polyethylene (reduction in the degree of crosslinking, increase in the number of micro-cracks),

- thermal ageing causes also an increase in intensity of the carbonyl band, connected with the oxidation of the material,

- the dielectric loss factor of aged polyethylene increases; this denotes the deterioration of the dielectric properties of the insulation,

- polyethylene A proved to be the best type of polyethylene in respect of the resistance to thermal ageing.

References

1. Grzybowski S., Rakowska A., Thompson J.E.: Examination of ageing process in polyethylene cable insulation. IEEE International Symposium on Electrical Insulation, Montreal, Canada 1984.
2. Grzybowski S., Zubielik P., Kuffel E.:"Spherulite structure in polyethylene high voltage cable insulation" IEEE Int. Symp. on Electrical Insulation, Boston 1980.
3. Grzybowski S., Zubielik P., Kuffel E.: Structure changes in PE cable insulation. Waszyngton 1986 (printed).
4. Rakowska A.: Relation between structure and electrical properties of polymer dielectric under various environmental conditions. Thesis,Poznań 1982.
5. Rakowska A.: Relationship between structural changes and dielectric properties of polyethylene "Przegląd Elektrotechniczny" Vol.1983, No 11/12.
6. Zubielik P.: An influence of short thermal overheats on PE insulation of polimer cables. Thesis, Poznań 1984.