

## Influence of thermal ageing on water sorption in EPDM rubbers

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### ABSTRACT

*There is a need in the insulation community for a deeper understanding of the effect of ageing and on insulators. The current work first focuses on EPDM derivatives. Changes of water transport properties (i.e. the coefficients of water solubility and diffusion) were correlated with chemical ageing (IRTF). The controlling parameters was found to be an oxidation criteria. These structure/property relationships will further be used to predict the penetration of water in EPDM connection accessories in service in the underground electricity network. In a longer range, this work should allow estimating their lifetime, when combined with a relevant end-of-life criterion.*

### KEYWORDS

EPDM, thermal oxidation, water sorption and diffusion, structure/property relationships.

### INTRODUCTION

An increasing number of connection accessories made of sulfur vulcanized EPDMs are used in the underground electricity network, making their durability a very important issue for electricity distributors. In addition to having good mechanical properties, these accessories must prevent water penetration within the insulating layer of the cables, because water might favor their dielectric breakdown.

In service, EPDM elastomers are submitted to several environmental constraints, in particular: oxygen, humidity, temperature and mechanical stresses. The barrier to moisture is likely to degrade long before the embrittlement resulting from the chemical/mechanical degradation. Polymer hydrophilicity essentially results from the molecular interactions between water molecules and polar groups. If the water solubility gradually increases with the concentration of polar groups, the diffusivity always varies the other way around, because of the stronger interactions. These trends have largely been observed with "highly" and "moderately" hydrophilic polymers [1]. Only scarce studies are to be found in the literature for more hydrophobic polymers.

LDPE is somewhat an exception with some sound and complete series of experiments [2]. This study concerns another type of almost apolar polymers: EPDM elastomers. The results focus on thermal oxidation campaigns. A peculiar attention will be paid to determining the appropriate physico-chemical criteria allowing both to monitor the course of oxidation and to describe polymer hydrophilicity. The measurement of water transport for various oxidation states of EPDM elastomers will also be described and analyzed in depth, to search for correlations with the established oxidation criteria.

### MATERIALS AND METHODS

Figure 1 describes the formulation of the two vulcanized EPDMs under study. Nordel IP 4520 (from Dow Chemical) was chosen for its high fraction in propylene units (42 wt%) leading to an amorphous rubber material after vulcanization. The sulphur fraction is the only parameter varied between the two formulations (4 phr in EPDM A and 1.2 phr in EPDM B).

| Function                  | Additive            | EPDM A | EPDM B |
|---------------------------|---------------------|--------|--------|
| Elastomer matrix          | EPDM Nordel IP 4520 | 100    | 100    |
| Vulcanization agent       | ZnO                 | 5      | 5      |
|                           | Stearic acid        | 1      | 1      |
| Cross-linking agent       | Sulphur             | 4      | 1,2    |
| Vulcanization accelerator | MBTS                | 4      | 4      |
|                           | TMTD                | 2      | 2      |

**Figure 1: formulation (in phr) of the two studied EPDM elastomers**

EPDM films of about 0.6-0.7 mm thick were press-molded and vulcanized at 170°C.

Accelerated thermal ageing were carried out in air-ventilated ovens at 150°C, 160°C and 170°C. EPDM samples were periodically removed from ovens and cooled in a desiccator before being characterized by several conventional laboratory techniques. Three main physico-chemical quantities were measured:

#### Carbonyls

The IR absorbance of carbonyl species C=O was determined at 1715 cm<sup>-1</sup> in an Attenuated Total Reflectance (ATR) mode with a Perkin Elmer Frontier FTIR spectrometer. Beforehand, all ATR spectra were normalized with the absorbance of the angular deformation of C-H bonds of methyl groups at 1378 cm<sup>-1</sup>. It can thus be safely assumed that the changes in carbonyl absorbance during thermal ageing quantitatively correspond to the carbonyl concentrations.

#### Density

The density ( $\rho_{\text{polymer}}$ ) was determined according to the Archimedes' principle. At room temperature, a dry sample was first weighed in air, then in ethanol (with  $\rho_{\text{ethanol}} = 0.78 \text{ g.cm}^{-3}$ ). The corresponding sample masses are noted  $w_{\text{air}}$  and  $w_{\text{ethanol}}$  respectively. Finally, the density may be expressed as: