RADIOCHEMICAL AGING OF ATH FILLED EPDM

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EPDM elastomer, representative of current formulations of cable insulators used in reactor buildings of power plants, was elaborated by press molding into 2-3 mm thick plates at 170°C under 200 MPa during 15 minutes. EPDM elastomer was filled by 0, 33 and 100 phr of alumina trihydrate particles (ATH), surface untreated (U-ATH) or treated by a vinylsilane coupling agent (T-ATH), and crosslinked with 3phr of dicumyl peroxide. The plates were exposed to ionizing γ irradiation under different dose rates (0.1, 1 & 10kGy/h) in air at room temperature. The resulting changes in the tridimensional crosslinked structure of this elastomer were determined by different complementary analytical technics at molecular, macromolecular and macroscopic scales.

For instance, the distribution of oxidation products concentration in the sample thickness and its change versus exposure time were determined by FTIR spectrophotometry (Fig.1). The results are in accordance with our expectations: as the dose rate increases, the thickness of oxidized layer decreases while the concentration of oxidation products increases.

As another example, the changes in crosslinking density (or in concentration of elastically active chains) were determined by swelling measurements in cyclohexane at 25°C using the Flory-Rehner equation, modified by Krauss for filled samples, and were verified by mechanical spectrometry. The results indicate that a post-crosslinking process occurs in the early periods of exposure to irradiation, which is expressed by a raise in crosslinking density. Then, a chain scission process predominates over crosslinking once the crosslinking density reaches a maximum value (Fig.2). It is deduced that post-crosslinking results from addition of peroxy radicals to residual insaturations of norbornene monomer not consumed during vulcanization step.

The oxidation kinetics is practically unaffected by the presence of ATH. Swelling results only show a higher initial crosslinking density in the case

of surface treated ATH, which may be attributed to chemical crosslinks at the filler/polymer interface.

The Young's modulus follows the same pattern as the crosslinking density. The elongation at break similarly shows an increase followed by a slight decline to demonstrate an acceptable correlation between different scales. In case of filled elastomers, the tensile tests show a more significant fall when the filler content increases. This last result may suggest more significant degradation at the filler/polymer interface.

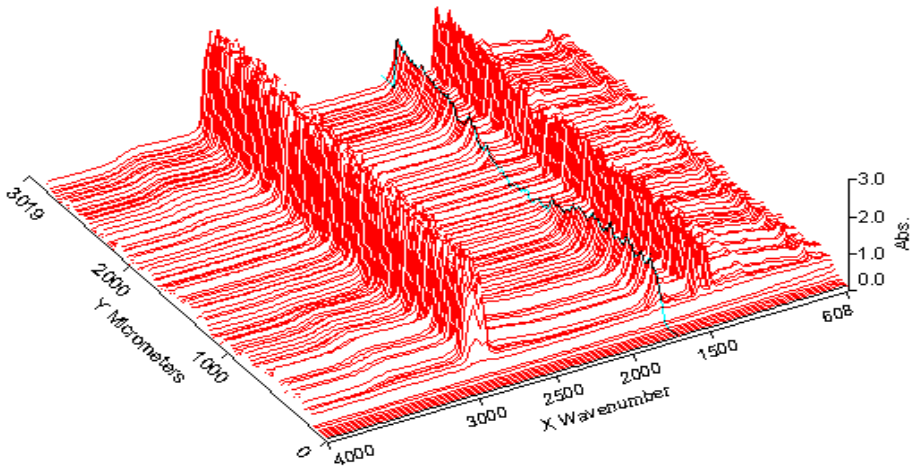

Figure.1: FTIR spectra presenting oxidation products profile for unfilled EPDM γ-irradiated for 800 hours in air under 1kGy/h dose rate

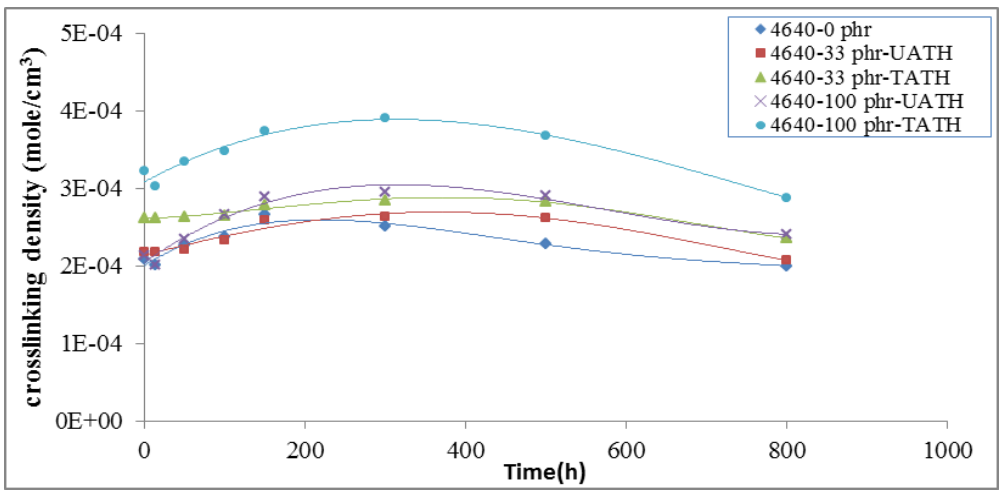

Figure.2: Evolution of crosslinking density of γ-irradiated EPDM samples under 1 kGy/h dose rate determined by swelling measurements