

CHARACTERIZATION OF MECHANICAL PROPERTIES OF IRRADIATED EPDM COPOLYMER COMPOSITES LOADED WITH CARBON BLACK AND ALUMINUM TRI-HYDROXIDE

Scagliusi, S. R. ¹*, Cardoso, E. C. L.¹,Lugão, A. B.¹

¹IPEN, Center of Chemistry and Environment, Sao Paulo, Brazil

*Corresponding author: srscagliusi@ipen.br

Abstract: Ethylene propylene diene rubber (EPDM) is known by its heat resistance and excellent electrical properties besides an effective radiation resistance. EPDM compositions containing carbon black and aluminum tri-hydroxide (ATH) were prepared and further irradiated aiming to evaluate mechanical performance as well the influence of these agents on compounds degradation. There were developed vulcanizable formulations with EPDM elastomer; besides addition of conventional additives for elastomers, inorganic loads, N-330 carbon black and aluminum tri-hydroxide were added too. Composites were subjected to gamma-radiation at 25 kGy , 50 kGy, 100 kGy and 200 kGy doses. Irradiated and non-irradiated composites were assessed as per mechanical-physical properties such as hardness, tensile, elongation at break, among others. It was verified that mixtures containing carbon black showed a better performance of mechanical properties for both non-irradiated and irradiated compositions; it was observed too that for irradiated composites at 200 kGy occurred the predominance of chain-scission.

Keywords: EPDM, mechanical properties, carbon black, gamma-radiation.

1. INTRODUCTION

Nowadays elastomeric composites thermally stable EPDM rubber-based loaded with inorganic as Carbon Black and Aluminum tri-Hydroxide have been very used in manufacturing of high voltage insulators, especially in polluted environments and in regions with high risk of vandalism. Mechanical properties of these compositions are very well considered for given applications. From the viewpoint that for any real work condition properties such as tensile resistance, modulus, and elongation at break and impact resistance are required for selection of polymeric material [1-3].

EPDM choice is justified due to its great versatility in processing and use in artifacts, including in various engineering areas, due to its excellent electrical property and oxidation resistance [4]. When particles are incorporated in polymeric matrix to produce composites, particles and matrix (interface) interaction is a relevant factor that affects composite properties [5].

Carbon black is traditionally employed as reinforcing agent in rubber industries [6]. A considerable amount of this material is applied in production of polymeric composites where electrical conductivity is required and used in various technological applications, as in electrostatic charges dispersion [7], touch power switches and protection against electromagnetic interference [6,8]. These materials aim to combine good electrical properties and excellent mechanical properties.

Addition of aluminum tri-hydroxide allows too improving mechanical properties of EPDM composites, besides promoting resistance to flames, smoke suppression, resistance to surface electrical discharges and inhibition of electrical treeing [9].

EPDM is usually used as insulating for electrical transmission cables, particularly in nuclear plants, due to its radiation resistance. Nevertheless, they can be degraded and fail in its electrical insulation. Degradation of mechanical properties showed to be the precursor of changing in electrical properties. So, the estimate of time life of a EPDM compound is based in elongation evolution.

The major effect of interactions between gamma-rays and polymers is based in build-up of free radicals; this evolution can provoke chain-scission, chain-branching and/or cross-linking. In general, all these phenomena coexist and the prevalence of each one depends on various factors. Among them, irradiation parameters, such as absorbed doses and irradiation dose rate because they affect the concentration of build-up of free radicals [10].

Ionizing radiation induces chemical reactions in polymers capable to result in changes either in molecular structure or macroscopic properties. Radiation energy transfer for the polymer does not occur selectively in relation to mixture components. The probability of generation of free radicals depends on strength of inter-atomic connections. Lower the power, easier the scission. In presence of oxygen, changes radiation induced are different in comparison with irradiation at inert environment [11].

This study aims to establish a correlation between mechanical properties of EPDM compounds loaded with carbon black and aluminum tri-hydroxide and how they interact on degradation, besides verifying the influence of loading agents on radio-chemical degradation process.

2. MATERIALS AND METHODS

2.1 Materials

Following materials were used in this study:

EPDM (ethylene-propylene terpolymer) provided by Flexys as Keltan 5470; Zinc oxide (ZnO) (Uniroyal do Brasil S.A.); Sulfur (S) (Basile Química LTDA); stearic acid (Basile Química LTDA.); 2-Mercaptobenzothiazol:(MBT) (EnroQuímicaFina LTDA.); BisulphideTetramethylthiuram (TMTD) (Enro QuímicaFina LTDA.); carbon black (NF) (National Bureau of Standard);aluminum trihydroxide (ATH) (Alcoa Aluminio S.A.). Materials were used as such with no previous purification.

Chosen formulation and mixtures processing were based in ASTM norms. Different developed compositions are shown in Table 1 (all amounts in phr) and were processed in Copé roll mill, 3 kg capacity.

Ingredients	Quantities	
	1	2
Keltan 5470	100	100
Carbon Black	120	-
ATH	-	120
Zinc oxide	5	5
Stearine	1	1
ParafinicOil	25	25
Sulfur	1,5	1,5
TMTD	1,0	0,5
MBT	0,5	1,0

Table 1. Developed Formulation



After mixture, it was accomplished rheometric analysis by using Oscillatory Disk rheometer(MDR) Monsanto (model R-100), in following conditions:180 °C, for 20 minutes and 3 degrees arch. Based in vulcanizing times (t90) provided by rheometric analysis, as compositions were compressed vulcanized in hydraulic press HIDRAUL-MAQ, 5 Mpa pressure, for specimens indicated for each type of essay.

2.2 Metodos

2.2.1 Irradiation

Gamma rays exposure was accomplished in air, at room temperature inside machinery provided with 60 Co. Dose rate was 5kGy h⁻¹, at 25, 50, 75, 100 and 200 kGy doses range. The dose accumulation procedure was preferred, because it keeps the identity of sample characteristics.

2.2.2 Tensile and Elongation at Break

Tensile and elongation at break values were determined according to ASTM D-412-08 [12], by using model C specimen, in essay universal machine, (EMIC), DL 300 model, 300 kN maximum capacity and 500 mm/min grips speed, at room temperature.

2.2.2. Hardness

Hadness values were determined according to ASTM D- 2240 norm [13]. The apparatus used was a Shore A durometer, Instrutemp, portable digital model Dp-100. This property is closely related to crosslink density.

3. RESULTS AND DISCUSSION

Results show the influence of loads used, before and after irradiation. In Figures 1 and 2 are shown respectively tensile and elongation at break results for samples loaded with ATH and Carbon Black, irradiated and non-irradiated.



Figure 1: Results of tension at break in irradiated and non-irradiated samples.



Figure 2: Results of elongation at break in irradiated and non-irradiated samples.

Obtained results formulations containing carbon black presented a better performance when compared to those ones with ATH; this is based in reinforcing character of carbon black in composite compared to ATH mineral load.

After irradiation behavior of both formulations is similar for doses up to 25 kGy: it occurs a slight rise in values pointing toward the build-up of crosslinks based in scission of unsaturated fraction of EPDM rubber. For doses between 75 and 100 kGy it is observed a given values stability suggesting a competition between crosslinking and scission. At 200 kGy scission process is predominant because it takes place a reduction in mechanical properties.



Figure 3: Hardness results for irradiated and non-irradiated samples.



Hardness evaluates the resistance for a material subjected to a local distortion [14]; it depends too on other factors such as load amounts and number of crosslinks. So, hardness can be increased depending on loads incorporation.

For samples ATH loaded there was a slight reduction in hardness values at initial irradiation; within 50 kGy and 100 kGy occurred a slight increase suggesting build-up of crosslinks during the process. For doses higher than 200 kGy a reduction in hardness values confirmed scission predominance.

For samples carbon black loaded there was a higher values balance at the beginning of irradiation; nevertheless, when increasing irradiation doses it was verified a predominance of crosslinks and for doses higher than 100 kGy it was observed the initial material degradation and consequently an enhancement in chain-scission.

Samples before and after irradiation containing carbon black presented hardness values higher than those ones containing ATH; consequently, it was proved the reinforcing character of carbon black in composite.

4. CONCLUSIONS

Studies accomplished in this study indicated that both EPDM rubber composites carbon black and Aluminum Tri-hydroxide (ATH) loaded can be considered as resistant to low irradiation doses (up to 100 kGy).

EPDM composites carbon black loaded showed a better performance in mechanical properties even in irradiated samples, confirming reinforcing characteristic of carbon black.

These studies showed too the influence of the load in composites mechanical properties as well indicate that the carbon black in composite formulation promoted a rise in polymer-load interaction improving this way required properties even after the irradiation.

5. REFERENCES

[1] R. P. Sheldon, - Mechanical properties of composites, in: Composite Polymeric Materials, Sheldon, R. P.(ed), Applied Science Publishers, New York, pp.58, 1982.

[2]R. B. Seymor,-Properties of polymers, in: Properties of Solids, Seymor, R. B. (ed), ASM International, USA, pp.17, 1987.

[3]N. A. Gent,-Strength of elastomers, in: Science and Technology of Rubber, Mark, J. E.; Erman, B. &Eirich, F. R. (ed), Academic Press, New York, pp. 471, 1994.

[4]J. K. Kim, I. J, Kim, Characteristics of surface wettability and hydrophobicity and recovery ability of EPDM rubber and silicone rubber for polymer insulators, ApplPolymSci, **79**, pp. 2251–2257, 2001.
[5] M.J. Wang, Effect of polymer-filler and filler-filler interactions dynamic properties of filled vulcanizates, Rubber Chem. Tech., 71, pp.520–589,1998.

[6]S. Wolff, M. J. Wang (ED.). Carbon Black, 2nd. New York: Ed. Marcel Dekker, 1993.

[7] S. J. PARK, K. S. CHO, S. K. RYU, Filler–elastomer interactions: influence of oxygen plasma treatment on surface and mechanical properties of carbon black/rubber composites. Carbon, **41**, pp. 1437-1442, 2003.

[8] S. C.Domenech, Novos materiais condutores para estudos biomecânicos: produção de misturas físicas poliméricas constituídas por polianilinas e borracha de EPDM. 2002. 211 p. Tese (doutorado),

Curso de Pós-Graduação em Engenharia Química. Universidade Federal de Santa Catarina, Florianópolis.

[9] The Vanderbilt Rubber Handbook, 1a ed., LosAngeles, R.T. Vanderbilt Company, 1978.

[10] G Spadaro, A Valenza, Influence of the irradiation parameters on the molecular modifications of an isotactic polypropylene gamma-irradiated under vacuum. Polym.Degrad. Stab. **67**, pp.449–454, 2000.

[11] R.L Clough, K.T Gillen, Investigation of cable deterioration inside reactor containment.NuclTechnol, **59**, pp. 344-354,1982.

[12] Norma ASTM D 412–08, Standard Test Methods for Vulcanized Rubber and Thermoplastic Rubber and Thermoplastic Elastomers – Tension, Vol. 09.01, Filadelfia (USA): American Society for Testing and Materials, 2008.

[13] Norma ASTM D 2240–08, Standard Test Method for Rubber Property – Durometer Hardness, Vol. 09.01, Filadelfia (USA): American Society for Testing and Materials, 2008.

[14] V. Shah, "Mechanical properties", in: Handbook of Plastics Testing Technology, Shah, R. P. (ed), JohnWiley & Sons, New York, p.7, 1984.

ACKNOWLEDGEMENTS

The authors thank CBE Embrarad for the irradiation process, Pirelli for the elastomers, Basile Chemistry for the raw materials, IPEN/CNEN-SP and CAPES agencies for their financial support.