Effects of Carbon Blacks on Electrical Properties of EPDM Compounds

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ABSTRACT: The correlations between mechanical and electrical properties of ethylene propylene diene terpolymer (EPDM) compounds and carbon black types and levels applied are studied. Tensile strength increases with an increase of the carbon black, especially for carbon black with higher surface area. Tracking resistance of EPDM compounds improves when a small amount of relatively nonconductive carbon blacks are added to EPDM compounds, whereas conductive carbon blacks decrease both dielectric properties and tracking resistance of EPDM compounds. Possible reasons for these results are discussed. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 78: 2540–2546, 2000

Key words: EPDM; carbon black; mechanical properties; dielectric losses; tracking resistance; weatherability

INTRODUCTION

Polymers have been widely used to replace porcelain and glass for external housings of high voltage outdoor devices, such as insulators, bushings, cable terminators, surge arrester, and so on, since the 1960s. Polymers offer significant advantages over porcelain and glass, such as light weight, easy installation, reduced breakage, and high impact resistance. In addition, polymers facilitate a greater flexibility in production design, and also can provide improved electrical performance, especially under contaminated conditions. Their main disadvantages are chemical changes on the surface due to weathering and arcing, and material degradation in the form of tracking and erosion.[1] Therefore, attention should be paid to the compounding design to prevent these disadvantages.

Ethylene propylene diene terpolymer (EPDM) is used in a diverse range of electrical applications because of its combinations of superior electrical properties, its flexibility over a wide temperature range, and its resistance to moisture and weather.[2] The saturated polymer backbone of EPDM is the key to its superb oxidative stability and excellent weathering resistance. Because EPDM is amorphous rubber, this compound contains only about 50% of base polymer and as many as over 10 ingredients with main fillers to achieve the mechanical and processing properties. Proper selection of the ingredients in each category requires that consideration be given to the desired physical, electrical, and environmental properties, as well as cost, ease of mixing, chemical stability, and ease of processing. Frequently these considerations may require compromises on different characteristics.

It is well known that carbon black suppresses the ultraviolet (UV) degradation owing to absorption of UV radiation.[3] The previous observation indicates that EPDM rubber compounds perform well outdoors.[4,5] Aubin et al.[6] have reported on the weathering behavior of EPDM compounds used in outdoor high-voltage insulation. They have suggested that the best protection against

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Table I Specifications of Used Carbon Blacks (Data from Manufacturer)

Type	Source	N_2 surface area[a] (m^2/g)	DBPA[b] (cm^2/100g)
ISAF (N220)	LG Chemical	128	111
XC-72 (N472)	Cabot	180	178
FEF (N550)	Kumho	42	117
SRF (N774)	LG Chemical	27	72
MT (N990)	Cancarb	8	36

[a] Surface area estimated by N_2 gas absorption.
[b] Dibutyl phthalate absorption.

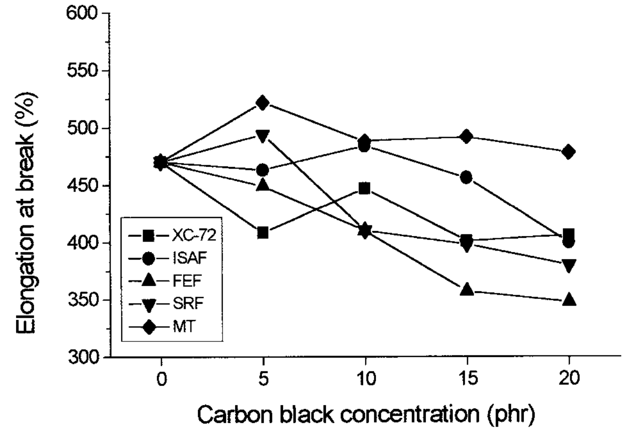

Figure 2 Effects of carbon blacks on elongation at break as a function of concentration.

weathering conditions is provided by a combination of nonconducting organic UV-stabilizers (surface protection) and of a high structure carbon black (surface and bulk protection) without decreasing the insulating properties of the compound. A similar result was observed by Lee and Kim for the cross-linked polyethylene (XLPE) samples.[7]

This paper deals with the effects of type and concentration of carbon black on mechanical properties, tracking and erosion resistance, dielectric loss, and volume resistivity.

EXPERIMENTAL

Sample Preparations

In this study, samples were classified into two categories; one was prepared for the evaluation and concentration of carbon black, and another was for the study of effects of semireinforcing furnace (SRF) black. The former sample was composed of 100 parts per 100 resin (phr) of Nordel 1040 (DuPont), 100 phr of alumina trihydrate (ATH), dicumyl peroxide (DCP) as a curing agent, paraffinic oil, wax, antioxidant (AgeRite Resin D of Vanderbilt), and zinc oxide. As shown in Table I, general-purpose (GP) grade black with 0 to 20 phr was used in this study. All components were first compounded in a continuous kneader at 80°C for 10 min, followed by two-roll milling at 80°C for another 10 min. Cross-linked samples of desired thickness were prepared by compression molding at 170°C for 10 min using a Carver Laboratory's hot press. Typical thickness of samples is 2.0 mm for mechanical tests, 1.0 mm for dielectric loss and volume resistivity tests, and 6.0 mm for tracking tests.

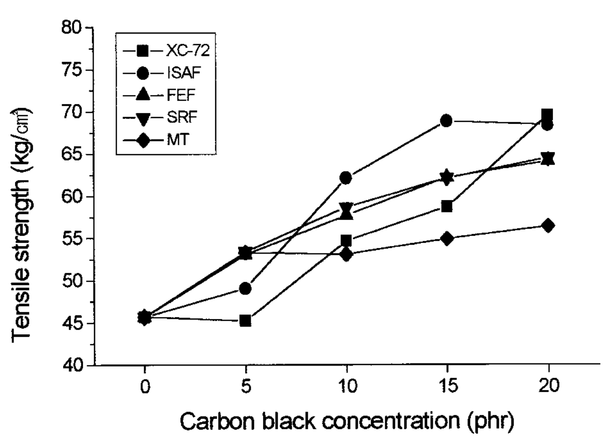

Figure 1 Effects of carbon blacks on tensile strength as a function of concentration.

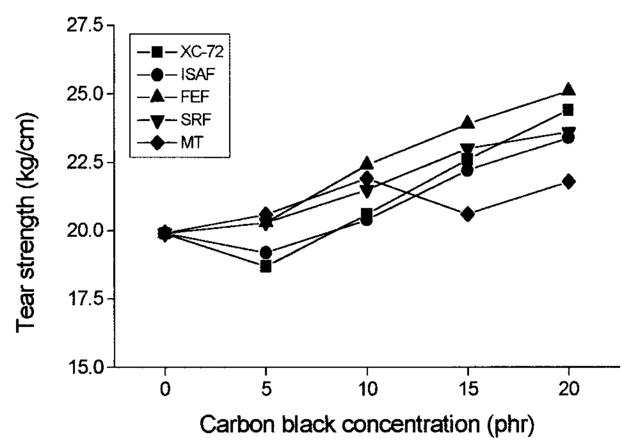

Figure 3 Effects of carbon blacks on tear strength as a function of concentration.

Table II Initial Tracking Voltage (kV) in Hourly Increments by 500 V Steps

Concentration Type	0 phr	5 phr	10 phr	15 phr	20 phr
XC-72	3.5–4.0	2.5–3.0	<2.5	<2.5	<2.5
ISAF	3.5–4.0	2.5–3.0	<2.5	<2.5	<2.5
FEF	3.5–4.0	>4.5	<2.5	<2.5	<2.5
SRF	3.5–4.0	4.0–4.5	2.5–3.0	<2.5	<2.5
MT	3.5–4.0	>4.5	>4.5	3.0–3.5	2.5–3.0

Tests

Tensile properties were measured by a universal testing machine following ASTM D638 at a crosshead speed of 500 mm/min. Tracking tests were carried out following ASTM D2303. The details of the specimen dimensions and electrode and sample positioning followed the standard. The end point criterion was the erosion depth of 3 mm or leakage current of 60 mA flows for 2 s. For the determination of the initial tracking voltage, samples were tested from 2.5 kV to 4.5 kV in hourly increments until failure occurred in 500 V steps. At least 10 specimens of each sample were tested and a range of failure voltages was recorded. For the determination of the time to track, samples were tested at a constant voltage of 4.5 kV, and the time to failure was measured. In this case, for samples, which did not show tracking trace for up to 6 hours at 4.5 kV, degree of erosion was calculated from the weights before and after tracking tests. Tan δ and dielectric constant were measured by Tettex's Schering Bridge and volume resistivity was measured by Insulation Resistivity Meter of Hewlett-Packard. Weathering behavior was evaluated using a Q-Panel's fluorescent UV-B lamp following ASTM G53. Its operating cycles were 6 hours UV at 60°C and 2 hours humidity at 50°C.

RESULTS AND DISCUSSION

It is known that factors to adjust the electrical conductivity in polymer-carbon black compounds are particle size, high structure, high porosity, and low volatile content of carbon blacks.[8] It is also known that the conductivity of carbon black is mainly related to structure and/or porosity, which can be inferred from DBPA (dibutyl phthalate absorption).[9] DBPA number is a measure of the structure of the carbon black aggregate. Generally, higher DBPA means higher structure and

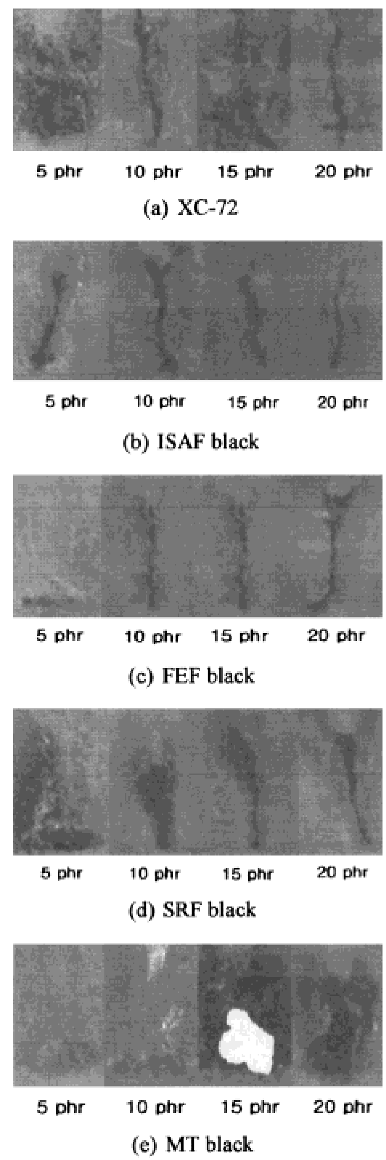

Figure 4 Tracking phenomena on carbon black filled EPDM as a function of concentration. (a) XC-72 black; (b) ISAF black; (c) FEF black; (d) SRF black; (e) MT black.

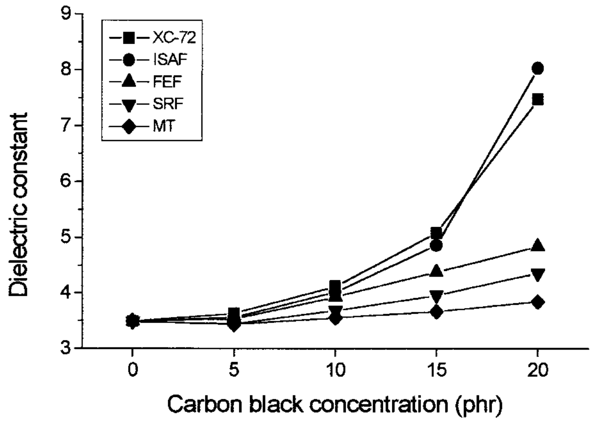

Figure 5 Effects of carbon blacks on dielectric constant as a function of concentration.

bulkiness, resulting in an increase of conductivity.[10]

As shown in Figures 1 to 3, in the case of EPDM samples with carbon black having higher surface area such as ISAF and XC-72, tensile and tear strength increase, whereas elongation at break decrease with increasing carbon black concentration. For the samples with medium thermal (MT) carbon having smaller surface area, tensile properties remain almost same even though some scatter points in data are seen. In the case of EPDM with SRF and fast extruding furnace (FEF) black, the tensile strength and tear strength show a moderate increase with an increase of carbon black content. The resulting reinforcement effects of carbon blacks agree well with their particle size and structure. Smaller particles such as XC-72 and intermediate super abrasion furnace (ISAF) blacks could produce a more uniform distribution in the cross-section, leading to a higher tensile strength.

In general, carbon black has a negative effect on the electrical properties of polymeric insulating materials. Clabburn et al.[11] have reported that 2% carbon black lowers the initial tracking voltage from 3 to 1.5 kV in polyethylene. For this reason, the carbon black concentration is limited by required electrical insulating properties such as tracking resistance, volume resistivity, and withstand voltage.

The effects on tracking resistance of carbon concentration are shown in Table II and Figure 4 for five types of blacks. In samples with ISAF and XC-72 black, tracking and erosion resistances significantly decrease with increasing carbon black concentration. It is thought to be that these types of carbon blacks provide a higher conductivity on a specimen derived from their high structure and small particle size leading to form a conductive path more easily. In that case, once a hot spot roots at the bottom electrode, failure by tracking usually occurs within a few minutes. For the samples with FEF and SRF blacks, tracking occurs with combination of the erosion in wide ranges and the conducting path, whereas the sample with MT black fails mainly from erosion. The tracking resistance for the samples with FEF, SRF, and MT black is improved by adding 5 phr. Levels higher than 5 phr reveal the excessive impairment of original tracking resistance for the samples with FEF and SRF blacks. The major point of interest shown in these data is the substantial improvement in resistance to tracking obtained by the use of 5 phr carbon black. It could be explained that carbon black provides for a linear voltage distribution, i.e., mitigation of concentration of the electric field due to conductivity of carbon black on the surface of the samples.[12]

The changes of dielectric constant, tanδ, and volume resistivity as a function of carbon black concentration are shown in Figures 5 to 7. As expected, for the samples with XC-72 and ISAF, increased amounts of carbon black reduce the dielectric properties such as dielectric constant, tanδ, and volume resistivity, whereas for the samples with SRF, FEF, and MT black, dielectric properties remain or slightly decrease with increasing carbon black concentration. These results could be explained by the fact that XC-72 and ISAF blacks have a lot of oxidative groups in their surfaces such as quinone (>═O), phenol (>OH), carboxylic acid (>—COOH), and lactone

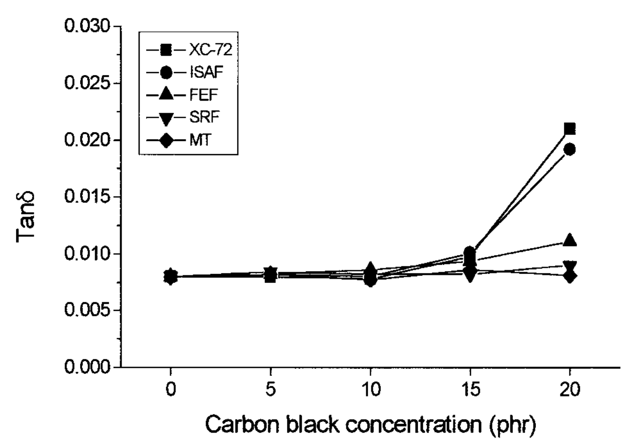

Figure 6 Effects of carbon blacks on tanδ as a function of concentration.

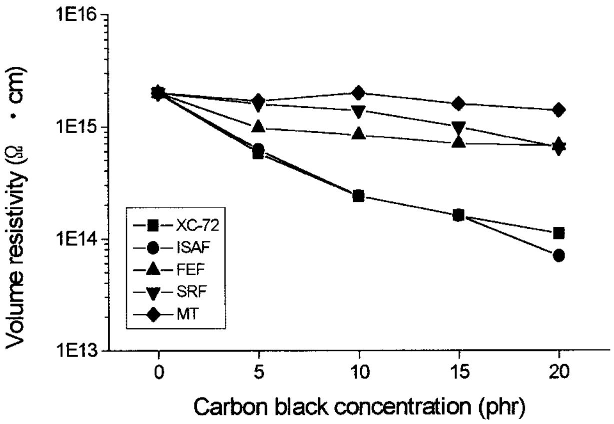

Figure 7 Effects of carbon blacks on volume resistivity as a function of concentration.

(>—COO—),[9] which result in a higher chance of static energy accumulation and higher polarity.

To explore the enhancement of tracking resistance by adding a carbon black in detail, another set of samples was prepared. ATH of 150 phr was added to improve the tracking resistance of EPDM compounds. SRF black was chosen for the reason of its relatively low conductivity, and its concentrations were 0, 0.5, 1.0, 1.5, 2.0 phr, respectively. These samples were designated CB-0, CB-0.5, CB-1.0, CB-1.5, and CB-2.0 for the virgin samples, and WCB-0, WCB-0.5, WCB-1.0, WCB-1.5, and WCB-2.0 for the samples accelerated weathered in fluorescent UV-B lamp for 2000 hours, respectively. For the purpose of quantitative analysis among the samples, the "time to track" method was adopted with a constant voltage, and the time to failure was recorded. Furthermore, effects of SRF levels up to 2.0 phr on weather resistance were also evaluated. Table III shows the effect of SRF black concentration on the tracking resistance. It can be seen that tracking failure does not develop up to CB-1.5 sample at 4.5 kV for 6 hours, whereas tracking failure occurred after 2 hours for the CB-2.0 sample. It could be said that a higher carbon black concentration results in a higher leakage current due to conductivity of carbon black, leading to an easy carbonization of the surface of the sample. The amount of erosion by measurements before and after tracking test decreases with increasing carbon black concentration up to 1.5 phr.

To clarify this effect, measurements of leakage currents during tracking tests were performed. Figure 8 illustrates a leakage current record of unaged EPDM samples having carbon black concentration of 0 and 1.5 phr. The leakage current surges reach 40 mA for the CB-0 sample and 35 mA for the CB-1.5 sample, respectively. Moreover, width of the peak and difference between maximum and minimum value of the CB-1.5 sample is much less than the CB-0 sample. Acceler-

Table III Tracking and Erosion Resistance Before and After 2000 Hours Weathering in Fluorescent UV-B Lamp (Time to Track Method at 4.5 kV for 6 Hours)

Sample	Tracking	Erosion (Wt Loss, %)
	Before weathering	
CB-0	no tracking	2.2
CB-0.5	no tracking	1.3
CB-1.0	no tracking	0.8
CB-1.5	no tracking	0.3
CB-2.0	tracking (30 min)	—
	After weathering	
WCB-0	no tracking	2.1
WCB-0.5	no tracking	1.4
WCB-1.0	no tracking	0.9
WCB-1.5	no tracking	0.3
WCB-2.0	tracking (25 min)	—

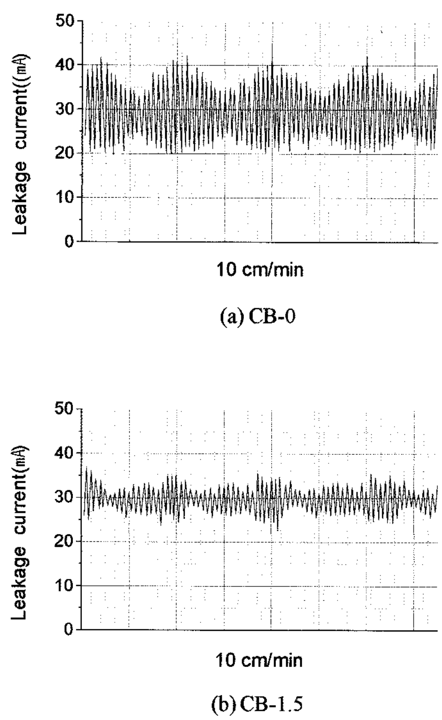

Figure 8 Leakage current records during tracking test (after 3 hours) for the virgin sample.

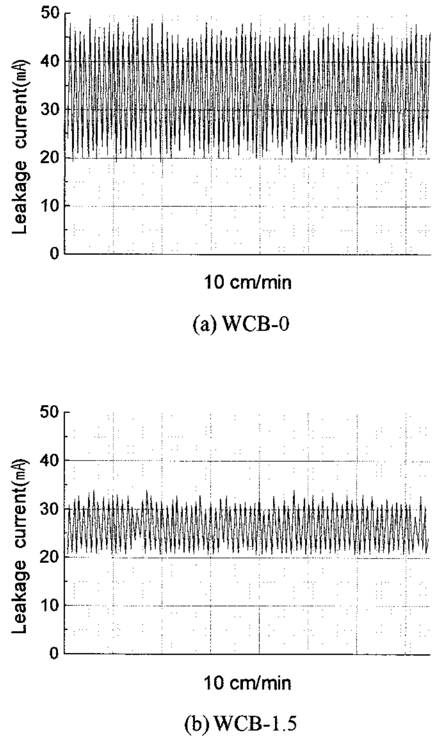

Figure 9 Leakage current records samples during tracking test (after 3 hours) for the 2000 hours aged sample.

ated weathering in fluorescent UV-B lamp for 2000 hours shows no deleterious effect on the tracking endurance of all the samples as shown in Table III. However, from the leakage current records of Figure 9, it can be seen that the magnitude and width of the peaks for the WCB-0 and WCB-1.5 samples slightly increase compared with the CB-0 and CB-1.5 samples, respectively.

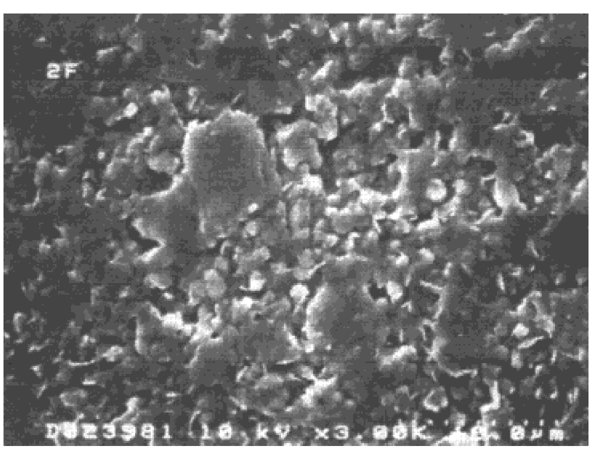

Figure 10 SEM photograph for the WCB-0 sample.

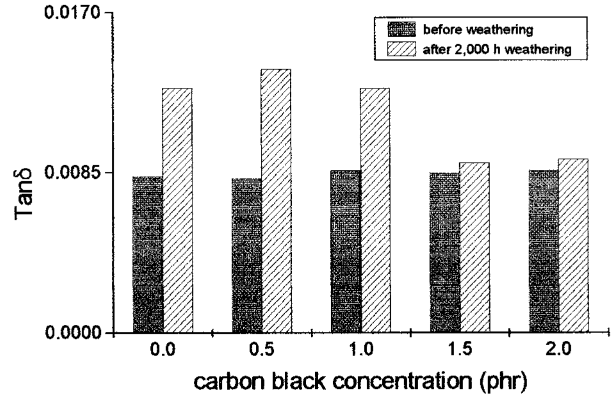

Figure 11 Tanδ of EPDM as a function of carbon black concentration before and after weathering.

This result could be explained by the surface change of the samples, i.e., increase of surface roughness from the formation of porosity after weathering as shown in scanning electron microscopy (SEM) photograph (Fig. 10), leading to increase of leakage current. From Table III and Figures 8 and 9 it is clear that the tracking and erosion resistance of the EPDM compounds are unaffected by accelerated UV weathering for 2000 hours, although a small increment of leakage current is observed on the surface of the samples.

On the other hand, it is well known that deterioration of olefin polymers by photo-oxidation encourage the formation of carbonyl group, resulting in an increase of polarity in the compound, followed by an increase of tanδ and dielectric constant.[13] Changes of tanδ and dielectric constant of EPDM compounds before and after weathering of 2000 hours are shown in Figures 11 and 12. For the unaged samples, dielectric losses are unaf-

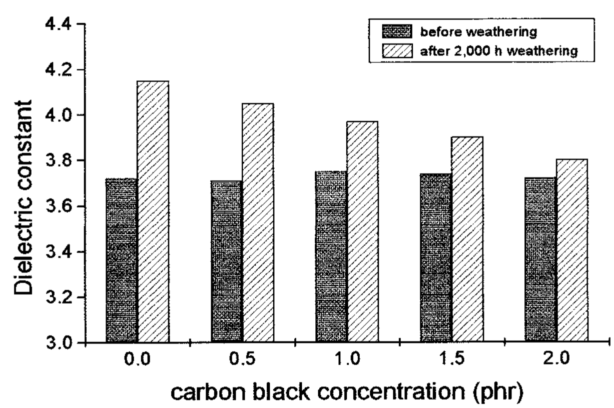

Figure 12 Dielectric constant of EPDM as a function of carbon black content before and after weathering.

fected by adding the SRF black up to 2.0 phr. In the case of UV aged EPDM, as expected, the marked increases of tanδ and dielectric constant are observed after 2000 hours accelerated weathering at lower concentration of carbon black. Again, it is assumed that such increase is due to the formation of carbonyl groups and water absorption during humidity cycles in UV equipment. As the carbon black level becomes higher, the rate of increment decreases after weathering. These results indicate that the addition of carbon black has a positive effect to suppress the photodegradation in EPDM/carbon black compounds.

CONCLUSIONS

It is necessary to apply an optimum carbon black type and concentration to maximize the performance and the lifetime of the EPDM compounds to resist the serious environment in the field. The results from this study suggest that the samples evaluated can be categorized into three categories. EPDM with XC-72 and EPDM with ISAF have a higher tensile strength, low dielectric properties, and low tracking resistance with increasing carbon black concentration. For the samples with FEF and SRF, tensile strength and dielectric properties moderately increase and decrease with increasing carbon black concentration, respectively. EPDM with MT black does not affect the mechanical and dielectric properties up to 20 phr of concentration, and improve the tracking resistance up to 10 phr of concentration. Addition of SRF black up to 1.5 phr has a positive effect to improve both tracking and weathering resistance. Enhancement of tracking resistance could be explained as follows: conductivity of carbon black suppresses the electrical field, leading to a linear voltage distribution during tracking test. Thus, tracking resistance and erosion resistance are improved by adding a small amount of carbon black.

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