

# Dielectric Relaxation and Mechanical Properties of Natural and Chloroprene Rubber with Some Nitroaniline Additives

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## SYNOPSIS

A systematic dielectric study over a frequency range from 400 to 10 MHz was carried out on natural rubber and chloroprene rubber with a small amount of nitroaniline additives. Three absorption regions were obtained as a result of the analysis of the curves relating dielectric loss  $\epsilon''$  with frequency. The first absorption region in the lower-frequency range could be attributed to the Maxwell–Wagner effect. The second absorption region which could be attributed to the ingredients added to rubber was found to be nearly the same for doped and undoped natural rubber samples, while it decreases by the addition of aniline derivatives to chloroprene rubber samples. The third region which is associated with Debye losses is found to be also nearly the same for natural rubber samples, while it decreases by the addition of aniline derivatives to chloroprene rubber; this decrease is interpreted according to the peptizing effect of aniline derivatives. For the expired chloroprene rubber gum, aniline derivatives were added and the mechanical and electrical behavior was improved to approach the unexpired one. © 1996 John Wiley & Sons, Inc.

## INTRODUCTION

Natural rubber and synthetic rubber are widely used in industry. A survey on the dielectric properties of natural and synthetic rubbers and the factors affecting these properties were given previously.<sup>1–11</sup> Additives may change the position and magnitude of the relaxation peaks.<sup>12–16</sup> On the other hand, the change in the relaxation spectrum gives some information about the reorientation and packing of the polymeric segments.

The object of this study was to investigate the effect of nitroaniline additives on the permittivity and relaxation mechanisms of natural as well as chloroprene rubbers. These anilines were chosen to act as structural peptizers in order to improve the electrical as well as mechanical properties of the rubber mixes, especially when added to the expired gum.

## EXPERIMENTAL

The mixing was carried out on a laboratory mill with the following dimensions: outside diameter 460 mm, working distance 250 mm, speed of the slow roll 16 rpm, and gear ratio 2. The aniline derivatives were added after the mastication of the rubber to decrease the toughness and the nerve of the rubber; then, the other ingredients were added to complete the mix. The curing characteristics of the rubber mixes were determined using a Monsanto oscillating disc rheometer Model 100 at  $162 \pm 1^\circ\text{C}$ . The mechanical properties were determined using a tensile testing machine (Zwick 1425, Germany).

The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  in the frequency range 400 to 10 MHz were measured. An LCR Meter type AG-4311B Ando electric LTD were used for the frequency range between 400 and 100 kHz. The capacitance  $C$  and the loss tangent ( $\tan \delta$ ) were obtained directly from the bridge from which  $\epsilon'$  and  $\epsilon''$  were calculated. The cell was calibrated using standard samples with known permittivity

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according to the method discussed before.<sup>16</sup> The experimental error in  $\epsilon'$  amounts to 3%, while for  $\tan \delta$  it is 5%. For frequencies between 50 and 10 MHz, a circuit magnification meter ( $Q$  meter) type TF 1246 from Marconi Instruments was used to measure  $\epsilon'$  and  $\tan \delta$ . The experimental error for  $\epsilon'$  and  $\tan \delta$  amounts to 3 and 5%, respectively.

To measure the conductivity  $\sigma$  of the samples, a power supply unit OM 451/01 from Philips was used, giving a stable dc voltage between 0 and 250 volts with a maximum permissible loading current of 1 mA. The potential difference ( $V$ ) between the plates holding the sample and the current ( $I$ ) flowing through it were measured by multimeter-type URI/BN 1050 from Rhode and Schwarz.

## RESULTS AND DISCUSSION

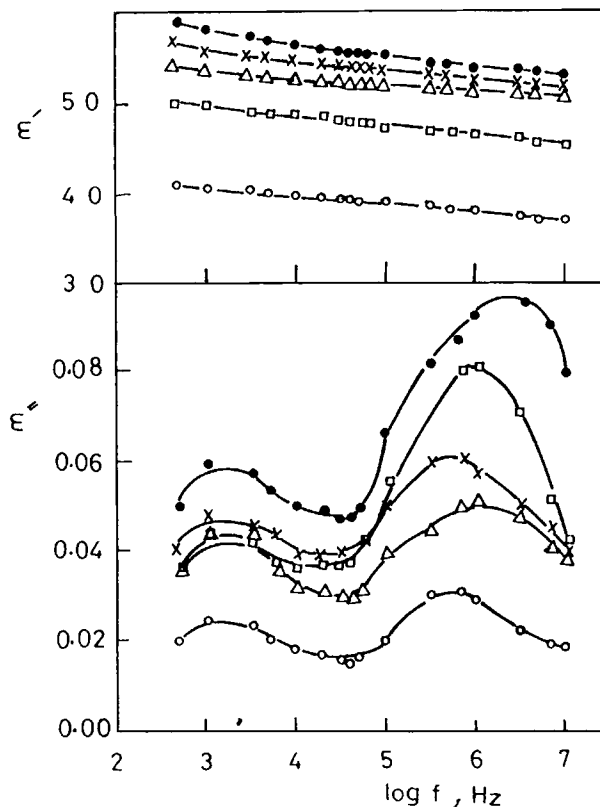
### Natural Rubber

The values of the permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  obtained for natural rubber containing 5 phr peroxide and 2.5 phr of *ortho*-, *meta*-, *para*-nitroaniline and *N,N*-dinitroaniline are illustrated graphically vs. the log applied frequency ( $f$ ) in Figure 1. All measurements were carried out at a room temperature of 25°C. From Figure 1, it is seen that  $\epsilon'$  increases with the addition of the aniline derivatives in the whole range of the tested frequencies. This increase is found to be in the order *ortho* < *para* < *meta* < *N,N*-dinitroaniline. On the other hand,  $\epsilon''$  is found to decrease by increasing the applied frequency and shows an anomalous dispersion.

The curves of  $\epsilon''$  as a function of  $\log f$  given in Figure 1 show that more than one relaxation process occurs. To a first approximation, analysis of the absorption curves was done in terms of the superposition of three absorption regions represented by three Fröhlich curves according to the Fröhlich equation<sup>17</sup>:

$$\epsilon''(\omega) = \sum_{i=1}^n \frac{\epsilon_s - \epsilon_\infty}{P_i} \arctan\left(\frac{\sinh P_i/2}{\cosh \ln(\omega\bar{\tau}_i)}\right)$$

where  $P_i$  is a parameter describing the width of distribution of relaxation times and equals  $\ln(\tau_1/\tau_2)$  and  $\bar{\tau}$  is the mean relaxation time and equals  $\sqrt{\tau_1\tau_2}$ .  $\epsilon''$  is the dielectric loss, while  $\epsilon_s$  and  $\epsilon_\infty$  are the static permittivity and the permittivity at infinite frequency, respectively.



**Figure 1** The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  for natural rubber with peroxide: (O) free; (●) *ortho*; (Δ) *meta*; (×) *para*; (□) *N,N*-dinitroaniline.

The values of the relaxation times obtained from the analyses are listed in Table I. An example of the analyses for the free natural rubber sample is shown in Figure 2. From this figure, it is interesting to notice that the three absorption regions are accurately defined as they lie within the experimental measurements.

The first absorption region which lies approximately at  $f = 1.5$  kHz is found to be present for all the investigated samples. This region could be attributed to the Maxwell-Wagner effect, the origin of which is an ac current which is in phase with the applied potential. This current results from the difference in the permittivity of peroxide and natural rubber. To ensure this,  $\epsilon'$  and  $\epsilon''$  for peroxide were measured and the obtained values of  $\epsilon'$  and  $\epsilon''$  together with the dc conductivity are illustrated graphically in Figure 3. From this figure, it is clear that  $\epsilon'$  and  $\epsilon''$  are much higher than those for rubber.

The second absorption region, with a relaxation time  $\tau = 1.6 \times 10^{-6}$  s for the free sample and  $1.7 \times 10^{-6}$  s for those containing different types of aniline derivatives, may be due to the polarity of either

**Table I Relaxation Data of Natural Rubber (NR) with Peroxide and the Different Aniline Derivatives at Room Temperature**

$\epsilon''_m$	$\tau_I \times 10^4$ (s)	$\epsilon''_m$	$\tau_{II} \times 10^6$ (s)	$\epsilon''_m$	$\tau_{III} \times 10^8$ (s)
<u>NR</u>					
0.026	1.06	0.019	1.59	0.026	7.95
<u>NR + <i>o</i>-nitroaniline</u>					
0.062	1.06	0.025	1.70	0.097	8.40
<u>NR + <i>m</i>-nitroaniline</u>					
0.045	1.06	0.025	1.70	0.054	8.40
<u>NR + <i>p</i>-nitroaniline</u>					
0.050	1.06	0.040	1.70	0.054	8.40
<u>NR + <i>N,N</i>-dinitroaniline</u>					
0.046	1.06	0.034	1.70	0.085	8.40

peroxide or the anilines or both added to the rubber. From Table I, it is clear that the addition of aniline derivatives increase only the height of this absorption region without any shift or effect on the relaxation time within the experimental error.

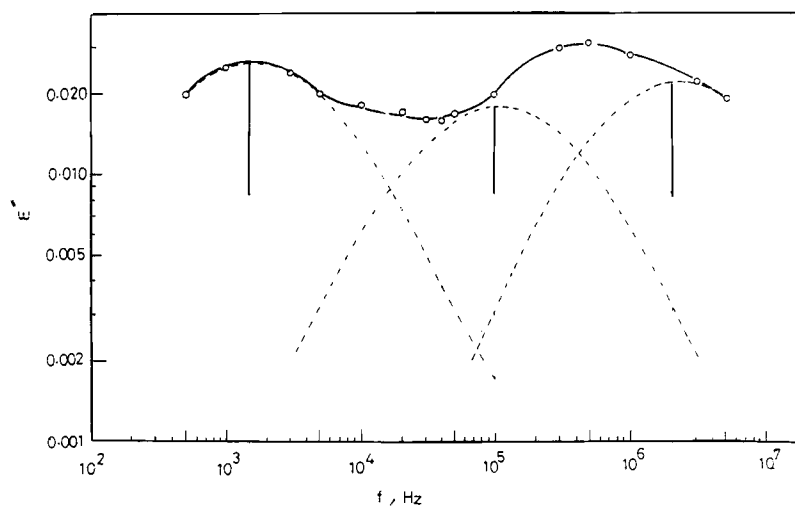
The third relaxation time, which is  $7.95 \times 10^{-8}$  s for natural rubber and  $8.4 \times 10^{-8}$  s for the other samples containing anilines, is associated with the Debye dispersion and corresponds to pure Debye losses due to the dipole orientations caused by movements of the main backbone carbon. The value of the relaxation time associated with this process is found to be nearly the same for the free samples and those containing anilines. Also, it is interesting to find that the addition of anilines increases only the height of this relaxation process without any

shift or effect on the value of the relaxation time. This means that these anilines have no peptizing effect on the natural rubber matrix. This conclusion is found to be comparable with that noticed by Mansour et al.<sup>18</sup> for rodlike molecules in polyisoprene.

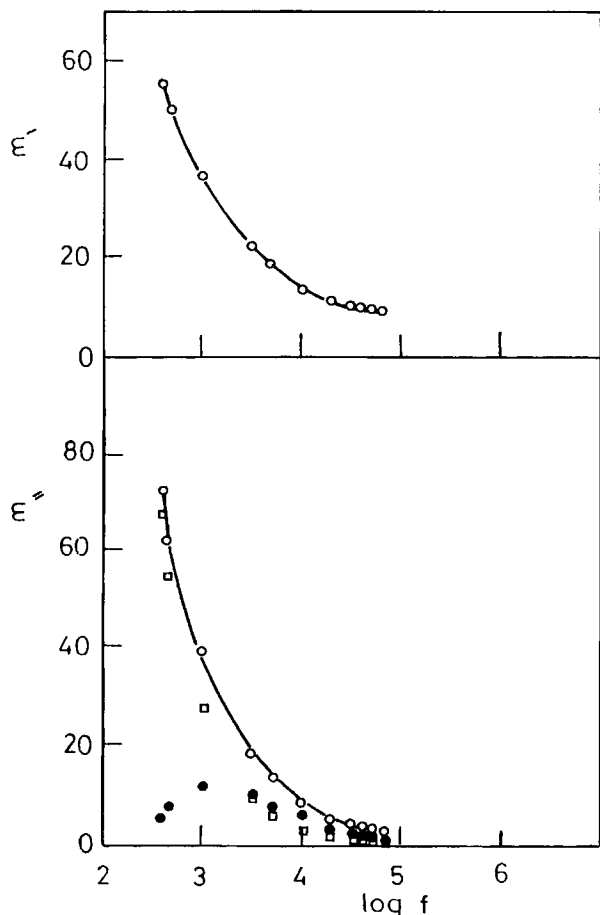
### Chloroprene Rubber

Chloroprene rubber was chosen as a synthetic rubber, in order to study the effect of addition of the nitroaniline derivatives as peptizers on its relaxation properties. The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  for chloroprene rubber containing 5 phr peroxide and 2.5 phr of *ortho*-, *meta*-, *para*-nitroaniline, or *N,N*-dinitroaniline were investigated over a frequency range from 400 to 10 MHz. The data obtained are illustrated graphically vs.  $\log f$  in Figure 4(I). From this figure, it is seen that  $\epsilon'$  increases by the addition of aniline derivatives in the order  $o > p > m > N,N$ -dinitroaniline and decreases by increasing the applied frequency, showing an anomalous dispersion. This result is close to that found here in the case of natural rubber. The variation of  $\epsilon''$  with  $\log f$  shows that more than one relaxation process occurs. These curves were analyzed into three Fröhlich terms.<sup>17</sup> The data obtained from the analysis are listed in Table II. From this table, it is interesting to notice that the three absorption regions are accurately defined as they lie within the experimental measurements.

The first absorption region which has a maximum at about 320 Hz may be due to either dc conductivity



**Figure 2** The absorption curves of natural rubber free from aniline: (O) fit of the experimental  $\epsilon''$  values using three Fröhlich terms.



**Figure 3** The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  for peroxide: (O) measured dielectric loss  $\epsilon''_m$ ; ( $\square$ ) loss due to dc conductivity  $\epsilon''_\sigma$ ; ( $\bullet$ )  $\epsilon''_m - \epsilon''_\sigma$ .

and/or the Maxwell-Wagner effect. No direct current was seen to flow through the samples, indicating that there is no dc conductivity. Moreover, it was ascertained that this effect is not due to bad contact between the sample and the condenser plates as the measurements were repeated with aluminum foil stuck to the two faces of the samples and no change in the results was noticed. As the value of the relaxation time is about the same for peptized and unpeptized samples, it is certain that the absorption region detected in that range of frequency is attributed mainly to the Maxwell-Wagner effect. The origin of this effect is an ac current which is in phase with the applied potential. This current results from the difference in conductivities and dielectric constants of chloroprene rubber and peroxide.

The second absorption region could be attributed to the large aggregates which are expected to be formed by the addition of peroxide to rubber. From

Table II, it is interesting to notice that not only an increase in the height of the relaxation process occurs, but also a decrease in the relaxation time is noticed. This decrease is found to be in the order *ortho* < *meta* < *para* < *N,N*-dinitroaniline.

The third relaxation time, which is  $7.96 \times 10^{-8}$  s, for samples free from anilines is associated with the Debye losses due to dipole rotations caused by movements of the main backbone. This region is in fair agreement with Schneider et al.<sup>19</sup> and Hanna et al.<sup>10</sup> From Table II, it is seen that this third relaxation process decreases by the addition of the aniline derivatives.

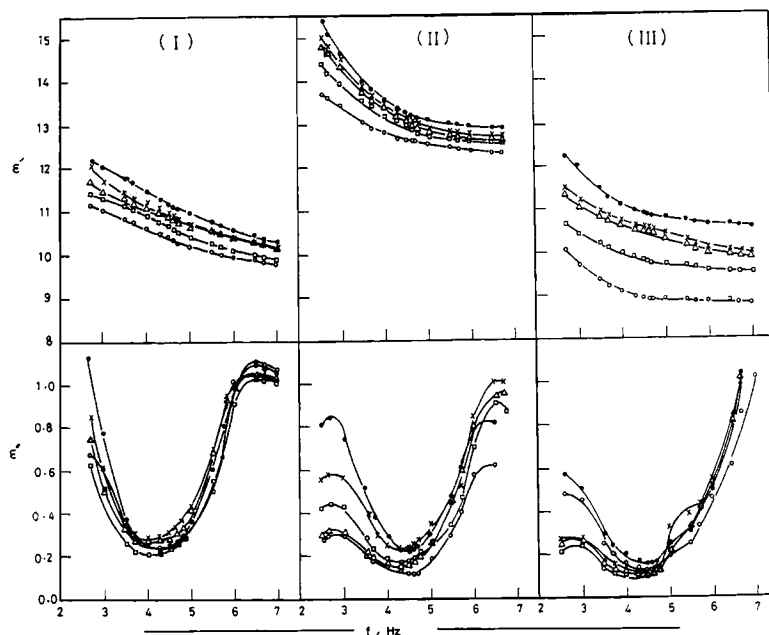
This decrease, which is noticed in the cases of the second and third relaxations, could be attributed to the peptizing effect of the low molecular weight additives on the chloroprene rubber. This leads to a decrease in the glass temperature<sup>20,21</sup> which may expand the range within the rubber that retains its flexibility and deformability.

On the other hand, the increase of the height of the relaxation process is evidence of the coupling between the reorientations of the added molecules and that of the rubber segments. This finding was observed in literature by several authors.<sup>22,23</sup>

From the above investigations, it could be concluded that the addition of the investigated anilines to natural rubber affect only the height of the relaxation processes and have no peptizing effect on its matrix, while the addition of anilines to chloroprene rubber (synthetic one) shifts the relaxation maxima to higher frequencies and, consequently, decreases the relaxation time due to the peptizing effect of these low molecular weight molecules added to the synthetic chloroprene rubber.

To study the effect of the addition of aniline derivatives in the presence of other different ingredients on the electrical and mechanical properties of chloroprene rubber, a formulation containing the different additives was prepared and is given in Table III. The aniline derivatives were added to the rubber mix in an equivalent ratio (2.5 phr). The rheometric characteristics were determined and are listed in Table IV. From the obtained data, it is clear that these compounds have a mild peptizing effect on the neoprene mix, especially on *o*-nitroaniline and *N,N*-dinitroaniline, which is shown from the decrease in the minimum torque and the increase in the scorch time.

On the other hand, the investigated compounds have no remarkable effect on the mechanical properties of the vulcanizates as shown from the mechanical properties. The aging of the vulcanizates



**Figure 4** The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  for chloroprene rubber with (I) peroxide, (II) the full ingredients, and (III) expired chloroprene rubber with the full ingredients: (○) free; (●) *ortho*; (△) *meta*; (×) *para*; (□) *N,N*-dinitroaniline.

at 100°C for 48 h led to a decrease in both tensile strength and elongation at break; however, the aniline derivatives enhance to some extent the aging properties, especially of *m*-nitroaniline and *p*-nitroaniline.

The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  for those samples were measured at 25°C and are illustrated graphically versus  $\log f$  in Figure 4(II). From this figure, it is seen that the values of  $\epsilon'$  increase in the order noticed for chloroprene samples containing peroxide. The dependence of  $\epsilon''$  upon frequency

shows that more than one relaxation process occurs. The values of  $\epsilon''$  were analyzed using the Fröhlich equation<sup>4</sup> and the obtained data are listed in Table V. It is interesting to notice that the three absorption regions are accurately defined as they lie within our experimental range of frequency. The first absorption region which is found at about 700 Hz could be attributed to the Maxwell-Wagner effect and is found to be independent of the addition of anilines. This region was not noticed before.<sup>10,19</sup> The second relaxation region is ascribed to the large aggregates which are expected to be formed by the addition of different ingredients to rubber. This region is found to be higher in comparison with that when chloroprene is mixed only with peroxide. The third relaxation, which is due to the pure Debye losses, is found

**Table II** Relaxation Data of Chloroprene Rubber (CR) with Peroxide and the Different Aniline Derivatives at Room Temperature

$\epsilon''_m$	$\tau_I \times 10^4$ (s)	$\epsilon''_m$	$\tau_{II} \times 10^6$ (s)	$\epsilon''_m$	$\tau_{III} \times 10^8$ (s)
<u>CR</u>					
0.750	4.97	0.18	4.97	1.150	7.96
<u>CR + <i>o</i>-nitroaniline</u>					
1.200	4.97	0.220	2.27	1.300	3.98
<u>CR + <i>m</i>-nitroaniline</u>					
0.800	4.91	0.390	1.59	1.250	3.97
<u>CR + <i>p</i>-nitroaniline</u>					
0.950	4.91	0.430	1.33	1.250	3.97
<u>CR + <i>N,N</i>-dinitroaniline</u>					
0.680	4.91	0.200	1.22	1.200	3.97

**Table III** Formulation of Chloroprene Rubber (CR)

Ingredients	phr
CR	100
Stearic acid	1.5
Zinc oxide	5.0
Magnesium oxide	4.0
Silitan Z	20
Different nitroaniline derivatives	2.5

to be decreased by the addition of the different aniline derivatives as in the case of chloroprene rubber-peroxide mixes. This decrease in the relaxation times which accompanied by increase in the heights of the relaxation processes is due to the peptizing effect of these low molecular weight molecules added to the rubber. This could be attributed to the shift in the glass transition temperature to the lower range which may occur by the addition of the peptizers, as was found in the case of chloroprene-peroxide mixes.

Chloroprene rubber usually suffers from scorching during storage. Thus, it is of great interest to study the effect of the investigated anilines on the vulcanization process as well as on the mechanical and electrical properties of the expired chloroprene rubber. For this purpose, the same formulations shown in Table III were prepared using the expired chloroprene rubber.

The addition of the compounds under investigation to the expired chloroprene mixes reduces the toughness of the rubber on the mill and improves the mixing and processing. This effect is clear from the rheological data shown in Table VI where the minimum torque was decreased and the scorch time was increased.

The presence of aniline derivatives has a renewable effect on the mechanical properties. This is clear from the sharp increase in both the tensile strength and the elongation at break. The aging of the vulcanizates at 100°C for 48 h shows a high stability of the vulcanizates containing the aniline derivatives against aging.

The permittivity  $\epsilon'$  and dielectric loss  $\epsilon''$  were measured at the same range of frequency and are

**Table V Relaxation Data of Chloroprene Rubber (CR) with the Full Ingredients and the Different Aniline Derivatives at Room Temperature**

$\epsilon''_m$	$\tau_I \times 10^4$ (s)	$\epsilon''_m$	$\tau_{II} \times 10^6$ (s)	$\epsilon''_m$	$\tau_{III} \times 10^8$ (s)
<u>CR</u>					
0.300	2.27	0.13	5.31	0.640	7.96
<u>CR + <i>o</i>-nitroaniline</u>					
0.820	2.27	0.220	4.68	0.940	6.37
<u>CR + <i>m</i>-nitroaniline</u>					
0.330	2.27	0.140	3.98	0.950	6.12
<u>CR + <i>p</i>-nitroaniline</u>					
0.590	2.27	0.230	3.79	1.000	6.12
<u>CR + <i>N,N</i>-dinitroaniline</u>					
0.460	2.27	0.160	3.62	0.900	5.31

illustrated graphically in Figure 4 (III). Comparing the data of  $\epsilon'$  with those for the unexpired ones, it is interesting to notice that  $\epsilon'$  values for the chloroprene sample mixed with the whole ingredients except nitroanilines are found to be lower. By adding nitroanilines to the mixes,  $\epsilon'$  values become higher and close to those for the unexpired ones.

The dependence of  $\epsilon''$  upon the frequency was analyzed in the same way discussed before where three absorption regions were detected, and the obtained data are listed in Table V. These relaxation processes are comparable with those found for the unexpired ones. The first absorption region, which is found to be the same for all the investigated samples, is attributed to Maxwell-Wagner effect. The second absorption region, which is attributed to the

**Table IV The Rheometric Characteristics and the Mechanical Properties of the Neoprene Rubber**

Property	Free	<i>o</i> -Nitroaniline	<i>m</i> -Nitroaniline	<i>p</i> -Nitroaniline	<i>N,N</i> -Dinitroaniline
<u>Rheometric characteristics at 162°</u>					
$M_L$ (dN m)	10	7	9	9	8.5
$M_H$ (dN m)	30	25	27	28	33
$t_{s2}$ (min)	2	3	2.5	2.5	3
$t_{e90}$ (min)	25.5	27	20	23	31
CRI (min <sup>-1</sup> )					
<u>Mechanical properties</u>					
TS (MPa)	9.50	9.01	8.35	8.15	10.23
$M$ 100% (MPa)	0.58	0.67	0.45	0.73	0.64
$E$ (%)	800	825	800	775	800
<u>Mechanical properties after aging 48 h at 100°C</u>					
TS (MPa)	6.34	7.86	7.96	7.10	7.88
$M$ 100% (MPa)	0.85	0.48	0.43	0.46	0.61
$E$ (%)	520	660	680	670	660

**Table VI The Rheometric Characteristics and the Mechanical Properties of the Expired Neoprene Rubber**

Property	Free	<i>o</i> -Nitroaniline	<i>m</i> -Nitroaniline	<i>p</i> -Nitroaniline	<i>N,N</i> -Dinitroaniline
<b>Rheometric characteristics at 162°</b>					
$M_L$ (dN m)	8.00	3.57	4.00	4.00	5.00
$M_H$ (dN m)	38	20	23	25	28
$t_{s2}$ (min)	1.5	2.5	2.5	2.5	2.5
$t_{c90}$ (min)	30.5	31.5	24.5	29.5	27.5
CRI (min <sup>-1</sup> )					
<b>Mechanical properties</b>					
TS (MPa)	5.26	11.6	12.2	9.43	13.46
$M$ 100% (MPa)	1.28	1.00	1.05	1.29	1.07
$E$ (%)	450	900	825	700	800
<b>Mechanical properties after aging 48 h at 100°C</b>					
TS (MPa)	4.26	10.79	12.03	8.78	11.91
$M$ 100% (MPa)	2.64	0.42	0.45	0.65	0.58
$E$ (%)	160	840	825	670	775

rotation of the large aggregates which are expected to be formed by the addition of the different ingredients to rubber, is found to be nearly the same but slightly decreases when adding *o*-nitroaniline and *N,N*-dinitroaniline. In addition, a detectable increase in the height of this process is noticed. The third relaxation process, which is due to the Debye losses, is associated with dipole rotations caused by movements of the main backbone of the rubber and is found to decrease by the addition of the aniline derivatives. This decrease is found to be maximized by the addition of *o*-nitroaniline and *N,N*-dinitroaniline.

Comparing Tables V and VII, it could be noticed that the relaxation time of the chloroprene rubber

**Table VII Relaxation Data of Expired Chloroprene Rubber (CR) with the Full Ingredients and the Different Aniline Derivatives at Room Temperature**

$\epsilon_m''$	$\tau_I \times 10^4$ (s)	$\epsilon_m''$	$\tau_{II} \times 10^6$ (s)	$\epsilon_m''$	$\tau_{III} \times 10^8$ (s)
<b>CR</b>					
0.560	2.27	0.10	6.31	0.230	15.90
<b>CR + <i>o</i>-nitroaniline</b>					
0.520	2.21	0.123	4.82	0.500	7.96
<b>CR + <i>m</i>-nitroaniline</b>					
0.260	2.21	0.110	5.31	0.320	10.61
<b>CR + <i>p</i>-nitroaniline</b>					
0.270	2.21	0.120	5.31	1.300	12.24
<b>CR + <i>N,N</i>-dinitroaniline</b>					
0.270	2.13	0.110	4.82	0.340	7.96

free from aniline is much smaller than that for expired free one. This means that the aging of the gum increase the rigidity of it and, consequently, the relaxation time. On the other hand, the addition of nitroanilines, especially *o*-nitroaniline and *N,N*-dinitroaniline, to the expired rubber leads to the value of  $\tau$  equal to that for the novel free one. This means that the addition of these two types of peptizers may lower the glass transition temperature as mentioned before, which may lead to the expansion of the range within which the rubber retains its flexibility and deformability.

From the above investigation, it could be concluded that addition of either *o*-nitroaniline and *N,N*-dinitroaniline, which act as peptizers, in small quantities to the expired chloroprene rubber will modify its electrical and mechanical properties and make it nearly the same as the unexpired one. This result is considered to be an important and interesting result in this field of work.

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